Review Article

An effective approach on physical and dielectric properties of PZT-PVDF composites

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Abstract

The 0-3 connected ferroelectric composites made of PZT and PVDF have been examined for its different physical and dielectric properties. The standard methods presented in earlier literatures for the composite preparation have been compared to find an effective method. The distinction in the physical properties like density and porosity of the piezocomposite was studied and their SEM morphological facts are scrutinized. The dielectric constant of PZT-PVDF composite has been predicted by using standard models and the results are compared. Also the relaxation behavior of polymer at higher temperature has been justified.

Keywords:PZT-PVDF, dielectric properties, scanning electron microscope, ferroelectric ceramic, ferroelectric polymer.

1.Introduction

'Smart materials'' are materials that show reproducible and stable responses through significant variations of at least one property when subjected to external stimuli [1]. Smart materials, which exhibit piezoelectricity, find a diverse range of applications in the industry [2]. In the past decade, great efforts have been made to develop flexible piezoelectric ceramic-polymer nano composites with high permittivity, which can be potentially utilized for preparing to embed micro capacitors to meet the requirement of the miniaturization trend of integrated circuits [3].

Piezoelectric ceramics are used in a variety of applications because of its excellent piezoelectric, dielectric properties, with very high pyroelectric constants, and strong electromechanical coupling [4]. However, its inherent properties, such as poor mechanical properties and the large difference in acoustic impedance with water restrict its applications in areas such as vibration sensing, impact detection, fiber-reinforced structures in aircraft and energy harvesting [5]. Among different piezoelectric ceramic materials, PZT is found to useful for the wide range of applications because of the presence of spontaneous polarization behavior.

Piezoelectric polymers provide relatively low piezoelectric strain coefficient (d_{33}), low dielectric constants, but offers unique advantages over ceramic such as low density, high breakdown strength and ease of processing. They have good mechanical flexibility and therefore can be formed easily on to the curved surfaces [6-7]. Ferroelectric polymers, such as poly (vinylidene fluoride) (PVDF) and its copolymers have been mainly used in various applications, especially in the field of sensor, transducers and actuator devices. PVDF is being widely investigated due to its outstanding pyro and piezoelectric properties among the polymeric systems. PVDF is chemically inert, tough, creep resistant, and has good stability when exposed to sunlight [8]. In addition, it has a low density along with low dielectric permittivity resulting in a very high voltage coefficient.

Piezoelectric ceramic-polymer composites combine superior properties of both polymer and ceramic phases [9], which offer many advantages over those of the constituent materials. These materials combine the excellent pyroelectric and piezoelectric properties of ceramics with the mechanical flexibility, chemical stability and processing possibility of polymers resulting in relatively high dielectric

permittivity, thermal conductivity and breakdown strength, which are not attainable in a single phase piezoelectric material [10-11].

The piezocomposites are classified into 10 different types according to their connectivity (such as 1-1, 2–2, 1–3, 0–3 etc.) which was reported by Newnham *et al* [12]. Connectivity is defined as the number of dimensions through which the material is continuous [13]. Among different connectivity patterns 0-3 and 1-3 type composites have been found to be more reliable for plenty of applications. Composites with 0-3 connectivity is the simplest form of the composite, they are easy to produce, and hence the volume fraction of the ceramic phase can be varied in a wide range [14]. The preparation of thin film composites with well-established techniques such as spin coating method can be made possible with 0-3 connectivity type.

Piezoelectric materials are excellent candidates for technological applications such as sensors and actuators because of their ability to couple electrical and mechanical signals. Piezoelectric ceramic–polymer composites are being increasingly utilized for their specific dielectric, ferroelectric, piezoelectric, pyroelectric, electro-optic, and superconducting properties [1, 15]. Composites of ferroelectric polymers (e.g. PVDF) and ceramics (e.g. PZT) have been widely utilized in underwater hydrophones, biomedical imaging with ultrasound and non-destructive testing applications [16]. Piezoelectric materials are versatile materials for the applications such as energy harvesting and sensing by applying a mechanical force to the material which produces a charge and voltage across the material (i.e. piezoelectric effect) and vice versa (i.e. Inverse piezoelectric effect). The direct piezoelectric effect has been widely used in sensor design, and the inverse piezoelectric effect has been applied in actuator design [5].

In the present work, an attempt has been made to correlate the variance of theoretically predicted values of dielectric constant values of the piezo ceramic composite. Standard models like Furukawa, Bhimasankaram, Yamada, Maxwell Wagner, Rayleigh model, and Effective medium theory have been considered for the study. Among different methods of composite preparation the samples prepared using a cold press, hot press, solvent casting, and spin coating method have been considered for this study and their surface morphological studies were scrutinized. The predicted dielectric constant of the composites obtained by standard models was analyzed. In addition to the above mentioned properties the temperature dependence of the dielectric constant of the composites was extensively studied.

2. Theory
2.1 Sample preparation
The most common method for the preparation of piezoelectric ceramic, polymer composites is chemical mixing of ceramic and polymer materials using a suitable chemical solution which is termed as a solvent. Literature survey revealed that there are different chemical compounds have been used as a solvent for the composite preparation. Cyclohexanone (CYX), dimethyl sulfoxide (DMSO) and dimethyl formamide (DMF) are commonly used as a solvent for PZT-PVDF composite preparation [17-18]. The ceramic and polymer ratio in the composite is well-ordered by the factor called volume fraction (ϕ). Prepared composite materials were made into samples of a desired dimension. The dimension of the sample will depends upon the application and sample preparation method. The prepared composite materials, undergoes heat treatment until solvent evaporate completely which may lead to the formation of voids in the composite.

Different methods are involved in the sample preparation. The summary of different methods available for composite sample preparation have been given by Jain *et al* [2]. Short description of prominent methods such as cold press, hot press, solvent casting and spin coating methods are described as follows.

2.1.1 Cold press method
In this method the prepared ceramic-polymer composite is filled into the specially designed die or mold of a desired dimension. The filled composite material is pressed under high pressure of the order of few megapascal (Mpa) for a fixed time period [19]. The samples prepared in this method suffers the disadvantage of having low dielectric and piezoelectric properties than compared to the samples prepared by hot press method [20]. The reduced dielectric and piezoelectric behavior of these samples is due to the presence of voids in the composite sample.

2.1.2 Solvent casting
This method is used to prepare thin or thick film samples over the specified area. The mixed ceramic-polymer composite in a solvent solution (e.g. DMSO) is stirred well using a magnetic stirrer under a fixed temperature to ensure the homogeneous mixture of ceramic particles into the polymer matrix. The solution is cast onto a glass substrate and heated in a furnace to eliminate solvent solution [21]. Thin or thick film composites will be obtained in this process, but it's tough to obtain thin films of uniform thickness. This method also suffers with the disadvantage of obtaining void free samples. The detailed description of this method has been provided by Jain *et al* [8].

2.1.3 Spin coating

Thin film composite samples of uniform thickness can be obtained by using the spin coating method [22]. In this method the composite material to be coated is deposited onto a substrate (e.g. glass) and allowed to rotate for a fixed time period [1]. The solvent used for the composite preparation is usually volatile, and simultaneously evaporates [2]. The thickness of thin film samples depends on the factors such concentration of the solvent, solution and the angular speed and time of spinning.

2.1.4 Hot press method

Hot press method is found to be one of the prominent and versatile methods for the preparation of composite samples in the form of pellets as well as thick or thin films. In this method the die or mold which is filled with coagulated composite material is heated using in-situ heater, and made into pellets under optimized temperature and pressure [16-17, 23-24]. The void free samples can be obtained by this method, but still hot press method is a non-continuous and time-consuming process [22].

2.2 Electroding and poling

The prepared composite samples were undergoing electroding and subsequent poling process. The electroding can be done on both sides of the samples by using a conductive silver paste [1, 17, 21, 24]. Gold or aluminum can also be used for electroding the samples and it has been represented in very few literatures [22, 25-26].

Electroded samples were poled under the optimized poling temperature and time by applying an electric field of the order of a few kilovolt to megavolt range. The poling field of the sample will vary according to the method of poling. The piezocomposites can be poled by either contact poling (thermal poling) or corona poling. The selection of poling method depends on the sample preparation as well as its application. In general poling has been performed by keeping the sample inside the silicon oil to avoid arcing and short circuiting. In certain cases corona poling can be done under high vacuum conditions. The detailed description about poling can be found in the earlier studies [22, 25, 27-28].

2.3 Theory of measurements

The experimental density of the composite can be found by using the conventional formula. The density values are calculated using the rule of mixtures [25, 29] which is given by equation (1),

$$\rho = \rho_1(1-\phi)+\rho_2\phi \qquad (1)$$

where ϕ is the volume fraction of PZT. ρ_1 and ρ_2 is the density of polymer and ceramic phase respectively. The porosity (p) and the relative density (ρ_{rel}) of the composites can be calculated from the experimental

and calculated density of composites using an equation,

$$p = 1- \rho_{exp}/\rho_{cal} = 1- \rho_{rel} \qquad (2)$$

The surface morphological properties can be analyzed by using scanning electron microscope (SEM) studies. The theoretical values of the dielectric properties of the composites were predicted by using the standard models. The obtained values have been compared with one another to choose the best model which is closely related to the experimental values given in the earlier literature studies. The theoretical models chosen for the present study have been given below.

2.4 Theoretical models for dielectric constant

In this study the ε_m, ε_i and ϕ represents the dielectric constant of the matrix (polymer phase), dielectric constant of filler (ceramic phase) and ceramic volume fraction respectively for all the models considered. The short description of selected models and its corresponding numerical formulation have been given here. The detailed description of these models has been presented in Jain *et al* [2].

2.4.1 Furukawa model

Based on the assumption that the ceramic particles are spherical and uniformly dispersed in the polymer matrix, a theoretical model had been proposed by Furukawa to predict the dielectric constant of composites and it is given by Furukawa *et al* [30],

$$\varepsilon = \frac{1 + 2\phi}{1 - \phi}\varepsilon_m \qquad (3)$$

2.4.2 Bhimasankaram model

This model is based on the fact that the ceramic particles are randomly dispersed in the uniform polymer matrix and also it is assumed that the each ceramic particle get polarized individually [31]. The dielectric constant of this model can be estimated by using the relation,

$$\varepsilon = \frac{\varepsilon_m (1 - \Phi) + \varepsilon_i \Phi \left[\frac{3\varepsilon_m}{\varepsilon_i+2\varepsilon_m}\right]\left[1 + \left[\frac{3\Phi(\varepsilon_i-\varepsilon_m)}{\varepsilon_i+2\varepsilon_m}\right]\right]}{(1 - \Phi) + \Phi \left[\frac{3\varepsilon_m}{\varepsilon_i+2\varepsilon_m}\right]\left[1 + \left[\frac{3\Phi(\varepsilon_i-\varepsilon_m)}{\varepsilon_i+2\varepsilon_m}\right]\right]} \qquad (4)$$

2.4.3 Yamada model

This model made one of the most general attempts of describing the dielectric behavior of composites. It is based on the properties of the individual materials and also consider a factor (n), which is related to the shape and relative orientation of the filler, while the other models considered only the spherical particles [32].

$$\varepsilon = \varepsilon_m \left[1 + \frac{n.\Phi.(\varepsilon_i - \varepsilon_m)}{n.\varepsilon_m + (1-\Phi)(\varepsilon_i - \varepsilon_m)}\right] \qquad (5)$$

Where, $n = 4\pi/m$ is the shape parameter attributed to the shape of the ellipsoidal particles. In the present

study it has been considered as n=8.5 with reference to the Yamada *et al* [32].

2.4.4 Maxwell-Wagner model

The dielectric constant of the composites having similar properties of one another can be found by using the Maxwell-Wagner equation [33] and it is given by,

$$\varepsilon = \varepsilon_m \left[\frac{2\varepsilon_m + \varepsilon_i + 2\Phi(\varepsilon_i - \varepsilon_m)}{2\varepsilon_m + \varepsilon_i - \Phi(\varepsilon_i - \varepsilon_m)} \right] \qquad (6)$$

2.4.5 Rayleigh model

This model was developed by Rayleigh based on his theory deduced from inferences taken from both the Maxwell–Garnett and Furukawa models for biphasic composite materials containing minor spherical filler [2 ,31].

$$\varepsilon = \varepsilon_m \left[\frac{2\varepsilon_m + \varepsilon_i - 2\Phi(\varepsilon_i - \varepsilon_m)}{2\varepsilon_m + \varepsilon_i + \Phi(\varepsilon_i - \varepsilon_m)} \right] \qquad (7)$$

2.4.6 Effective medium theory

In effective medium theory model, the dielectric property of the composite is obtained by averaging the permittivity values of the constituents.

In this model it is assumed that a random unit cell consisting of each filler (ceramic) particle surrounded by a concentric matrix (polymer) layer.

$$\varepsilon = \varepsilon_m \left[1 + \frac{\Phi.(\varepsilon_i - \varepsilon_m)}{\varepsilon_m + n(1 - \Phi)(\varepsilon_i - \varepsilon_m)} \right] \qquad (8)$$

The above equation is the EMT model for the prediction of effective dielectric constant of polymer–ceramic composite. The only restriction of using this model is that the particle size should be very small which is suitable to nanocomposite [34], n is the surface factor which is taken as 0.16 in this study.

2.5 Variation of dielectric constant with temperature

The temperature dependence of the dielectric constant of the piezo ceramic-polymer composites have been extensively studied in most of the earlier literatures. The PZT-PVDF composites of 0-3 connectivity prepared using different methods under different condition have been considered for this study. The composite samples prepared in the different poling field [17], dissimilar particle size of PZT [35], altered volume fraction [4, 23]and measured at diverse frequencies [18] have been considered for the study.
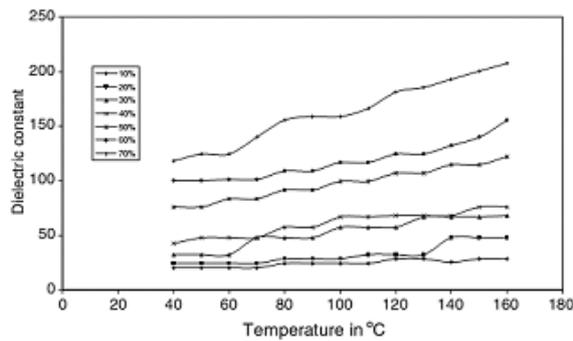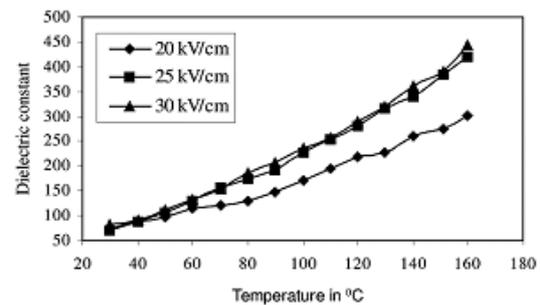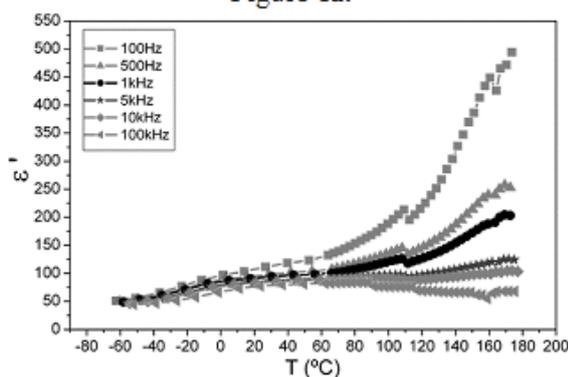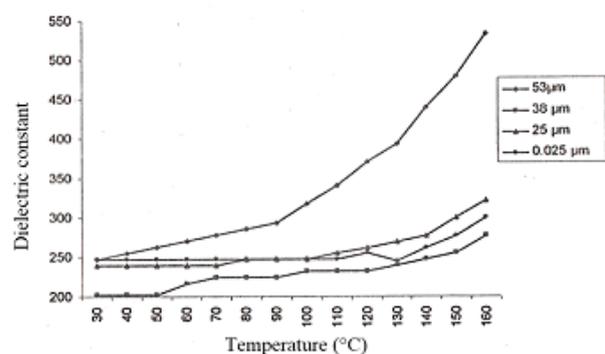

Figure 1a.

Figure 1b.

Figure 1c.

Figure 1d.

Fig. 1(a-d): Variation of dielectric constant of PZT-PVDF composite with temperature for different volume fraction of PZT [23], poling field [17], frequency [18] and at different particle size [35].

3. Results and discussion

3.1 Density and porosity

The theoretical and experimental density of the composites prepared using different methods of

preparation have been analyzed and compared. The density of composites prepared by solvent cast method is much lower than the hot press method have been reported by Venkatragavaraj *et al* [21]. Zhang De-

Qing *et al* [20] stated that the density of the hot pressed sample was higher than the cold pressed sample. The theoretical density of the hot pressed composite samples was related to the experimental density [17] and particularly at higher pressure the density values are lie closer to each other [23]. The voids in the composite samples were minimized at higher temperature and pressure, which causes the improved density in the hot press method.

The literature studies [20-21] revealed that the porosity of composites prepared using cold press and solvent casting method were relatively higher than compared to the composites prepared using the hot press method due to the presence of voids. The porosity can also be minimized using the spin coating method due to the reduced thickness of the layer which helps to decrease formation of voids.

3.2 Surface analysis of the composite

Surface morphological studies of the PZT-PVDF piezocomposites with 0-3 connectivity has been analyzed. The SEM micrograph studies presented in the earlier literatures revealed that the PZT particles are uniformly dispersed in the polymer matrix and the number of PZT particles in the polymer matrix

increases with the increase in the PZT volume fraction. It is also found that there is no agglomeration found in the samples irrespective of the preparation methods [1, 7, 18, 20, 36]. From these discussions it can be concluded that the chemical mixing of polymer and ceramic materials using a suitable solvent (e.g. Cyclohexanone) at a fixed temperature is the root cause for the homogeneous distribution of ceramic particles. Stirring the composite solution using a magnetic stirrer and the sonication process will help to achieve more uniform distribution of PZT particles.

3.3 Dielectric properties

3.3.1 Dielectric constant

An attempt has been made to predict the dielectric constant of the composites using some of the standard models and the comparison of those predicted values was represented in the Fig. 2. These results were correlated with the experimental values in the former literatures. Rayleigh and Maxwell-Wagner models were flops to follow the variation of the dielectric constant with the volume fraction of ceramic phase, whereas other models considered in this study shows the relative variation in dielectric constant with ϕ.
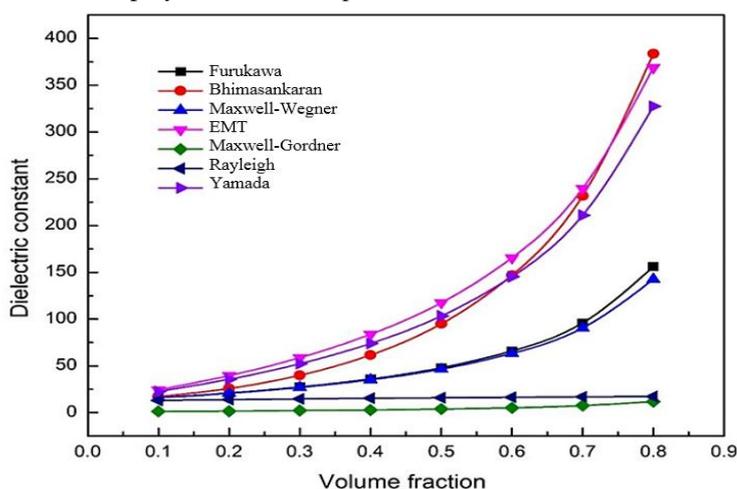

Fig 2: Comparison of predicted dielectric constant of different models.

In comparison with existing experimental studies, it is found that the each model has its own limitations and fails to explain experimentally observed variations of dielectric constant. Furukawa's model assumes that the piezocomposite is electrically inhomogeneous due to space charge effects. This assumption leads to the deviation in the experimental results from the theoretical values estimated by Furukawa *et al* [30]. Bhimasankaram *et al* [31] considered the effect of oriented dipoles in the surrounding medium. Hence Bhimasankaram's model helps to obtain significant results closer to the experimental results at higher volume fraction of PZT, but still it fails to explain the deviations at lower volume fractions. Significant effect have been made

by Yamada *et al* [32] to follow the changes in the particle size of the PZT but still Yamada model is lacking with the shape of the particle. The usage of effective medium theory has been constrained to the nanocomposites only. From these observations it should be noted that a model which is applicable from macro to nano scale has yet to be developed for the predictions of dielectric constant of piezocomposites.

3.3.2 Temperature dependence of dielectric constant

The variation of dielectric constant with the changing temperature of the composite materials has been analyzed extensively in numerous literatures [4,17-18,23,35]. The temperature dependence of the dielectric constant for the piezocomposites prepared at

different volume fraction, frequency, particle size and poling field have been represented in the Fig. 1. From these observations, it is found that the dielectric constant increases with increase in temperature irrespective of the parameters considered in the sample preparation. It can be concluded that the variation in the dielectric constant is mainly attributed due to the relaxation behavior of polymer (PVDF) phase at higher temperatures.

4. Conclusion

The dielectric and physical properties of ferroelectric composites made up of PZT-PVDF with 0-3 connectivity have been investigated. Among different methods chosen for the sample preparation, hot press method and spin coating method are found to be a more reliable method. The voids in the piezocomposite are reduced by a hot press method which helps to obtain highly densified piezocomposite samples with low porosity. The spin coating method is found to be a prominent method for the preparation of thin film samples.

SEM micrographs revealed the homogenous dispersion of PZT particles in polymer matrix and it is achieved mainly because of the chemical mixing of PZT and PVDF phases in a solvent. In comparison and detailed study about the theoretical predictions of dielectric constant revealed that a model has yet to be developed to resolve the experimental variation of dielectric constant with the predicted one. The temperature dependence of dielectric constant studies revealed that the relaxation of polymer at higher temperature leads to the relative increase in the dielectric constant of the composite.

Acknowledgements

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