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Dielectric Properties of 0-3 PZT/PVDF/Graphite Composite

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Abstract— We prepared the composites of PZT/PVDF/Graphite composites having 0-3 connectivity using hot press method. These samples were poled under different conditions i.e. at different poling temperatures and fields. We characterized these composites for their dielectric, piezoelectric and pyroelectric properties. We are reporting and discussing only the dielectric properties of these composites in this paper. The highest dielectric constant was found to be around fifty for composite with almost equal percentage of ceramic and polymer.

Index Terms: PZT, PVDF, Graphite, polymer, ceramic, composites, piezoelectric, pyroelectric, dielectric

1 INTRODUCTION

The piezoelectric materials can be divided in three classes viz. inorganic, organic and composites. In inorganic class there are ceramics and crystals having high dielectric constant, high piezoelectric coefficient, high electromechanical coupling but poor mechanical properties and relatively high acoustic impedance restricting their use to only certain applications. In organic class there are polymers, such as Polyvinylidene Fluoride PVDF, having relatively low acoustic impedance, which could provide a good acoustic matching to water or tissues, but its piezoelectric properties are inferior to those of ceramics. In composite class mainly there are ceramic/polymer composites having superior piezoelectric and dielectric properties as compared to polymer as well as superior mechanical properties as compared to ceramics. Thus they have their use in many applications owing to their better and tunable properties.

These composites can be further divided into different classes based on the connectivity scheme of their constituents. Composites of the 0–3 connectivity type comprising piezoelectric ceramic (FC) inclusions in an extended polymer matrix exhibit a variety of useful physical properties and are widely used in modern piezoelectric technology, acoustics, and other fields [1–4]. Different composites of PZT with various types of polymers such as PVDF, PVC, PVA and copolymers have been widely studied and reported in literature.

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But poling 0-3 composites is a big challenge and improper poling results in reduced useful properties. It has been reported in the literature that 0–3 composites cannot be poled fully due to the screen effect of the polymer matrix, so the total properties of the composite are reduced, which limits the practical application of the 0–3 ferroelectric composite [5, 6]. It was found by Sakamoto et al that doping the composite with a conductor results in improved poling and thus enhances the properties [7]. Other researchers have also found similar experimental phenomena when the electrical conductivity of matrix material was raised [8–11]. The uses of semi/conducting fillers not only increase conductivity, but also induce Maxwell–Wagner polarization which leads enhanced dielectric properties [12]. In present work we have prepared the 0-3 composites of PZT, PVDF and Graphite and studied their dielectric, piezoelectric and pyroelectric properties.

2 MATERIALS AND METHODS

2.1 Sample preparation:

The samples containing different volume percentage of PZT, PVDF and Graphite were prepared, in free standing film form, by the hot press techniques. These were then electrode with standard silver paste from Du-Pont. The samples were then poled in Silicone oil bath at different poling voltages for one hour. The optimum value of poling field and temperature was found to be 9 kV/mm and 1000C respectively. The optimum concentration of Graphite was found to be 1% by volume.

2.2 Characterization:

The dielectric properties i.e. dielectric constant and dielectric loss was estimated by measuring the C, D (capacitance and dissipation/loss factor represented by tanδ) parameters at different frequencies and also at a constant frequency with varying temperature. The capacitance (C) and loss factor (D) was measured with the help of Agilent precision LCR meter (4284A). For measurements under varying temperature conditions the sample were kept in silicone oil bath for uniform heating. The samples were short circuited for one hour before taking any measurements to avoid any stray capacitance effects. Then dielectric constant and dielectric loss for the samples were calculated using standard relations given in [13].

3 RESULT AND DISCUSSION

The dielectric permittivity and loss of composites depend not only on the permittivity of each phase in the mixture but also their volume fractions, shape, size, porosity, interphase polarizability and interphase volume fractions [14-16]. Figure 1 depicts frequency dependence of dielectric constant and dielectric loss of the composite on frequency for different volume fractions of different phases. It is observed that the dielectric constant slightly decreases with frequency. The reason for this decrement is the fact, that as frequency increases the different polarization mechanisms are not able to follow the change and they cease to contribute towards the dielectric constant. But as the decrease is not very much the dielectric loss almost remains constant except in the low frequency domain. And the reason for this may be the space charge effect and high dielectric loss associated with PZT.

It can be observed from figure 2 that the both the dielectric constant and loss increases with the increase in ceramic loading at all frequencies. An increase in the content of ceramic fillers in the composites increases the interfacial area between the ceramic phase and polymer phase. As a result, the influence of interfacial polarization on the dielectric permittivity and dielectric loss can be significant. Accordingly the relative permittivity and loss increase with ceramic loading. [17]

In all cases the effective dielectric constant is higher than that of pure PVDF and lower than that of pure PZT [18]. It is well known that PVDF has five different polymorphisms, of which four phases designated as α, β, γ and δ are stable at room temperature and α phase, which is nonpolar, is the most commonly present [19]. Due to the nonpolar nature of PVDF and the constrained polymer chain hindering the contribution of the electrical polarization, the value of dielectric constant is lower than that of PZT. The low frequency dispersion increases with the increase in PZT content.

The dielectric constant increases with increasing poling field upto 9kV/mm of poling field and decreases beyond that as shown in figure 3. This may be due to the fact that the poled sample has been polarized effectively and reached saturation at 9kV/mm of the field.

Figure 4 illustrates the temperature dependence of dielectric constant and dielectric loss of composite at 100 kHz of frequency. The dielectric constant increases with increasing temperature. The reason for this is the fact that higher temperature facilitates easier movement of the different dipoles. Thus the dipoles are able to follow the change in frequency more aptly. Another reason is the fact that polymer matrix expands with temperature and this results in increased interfacial polarization [20]. The increment in the dielectric loss with temperature is because of the increased conductivity of polymer matrix due the presence of graphite.

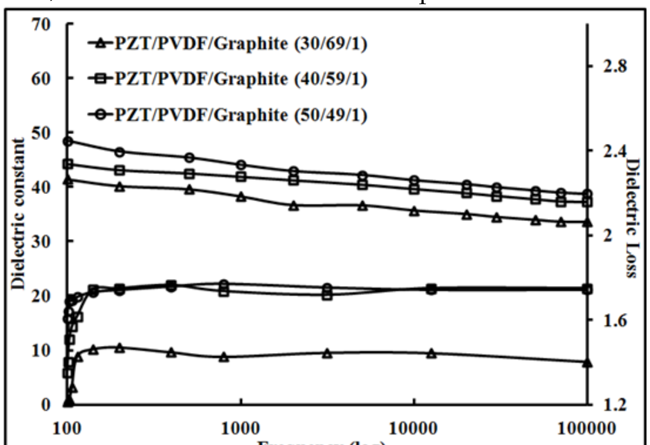

Figure 1: Variation of dielectric constant and loss with frequency for different composites poled at 9 kV/mm

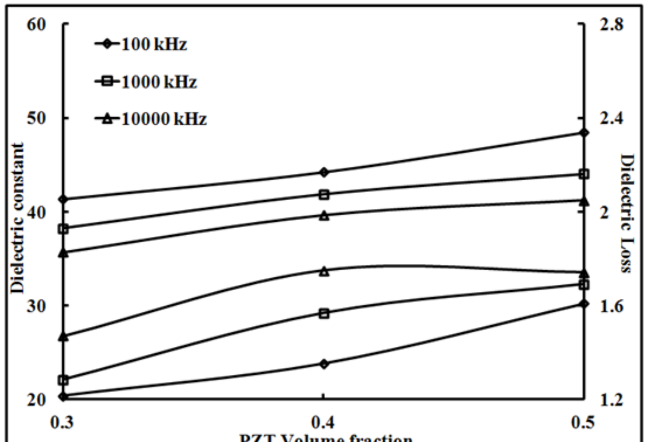

Figure 2: Variation of dielectric constant and loss with PZT volume fraction at different frequencies

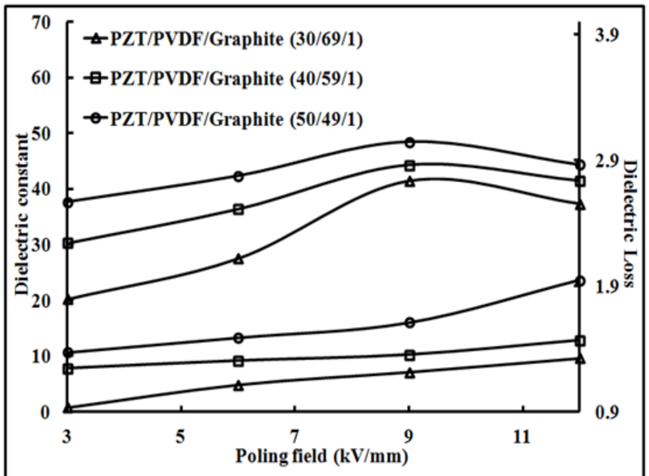

Figure 3: Variation of dielectric constant and loss with poling field for different composites at 100 kHz

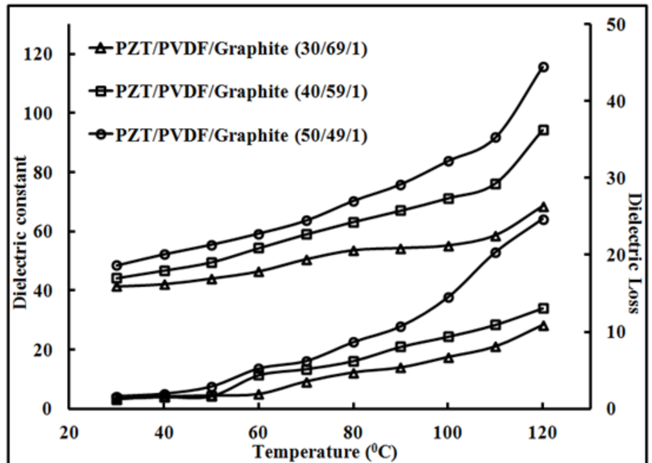

Figure 4: Variation of dielectric constant and loss with temperature at 100 kHz for different composites poled at 9 kV/mm

4 CONCLUSION

We prepared the 0-3 composites of PVDF and PZT and doped them with 1% of graphite. The graphite doping was done to increase the conductivity of the matrix as this increased conductivity will enhance the poling of ceramic filler.

The dielectric constant of almost composite having almost equal loading of ceramic and polymer was found to be about fifty. Both the dielectric constant and loss almost remained constant with frequency and increased with ceramic & polymer loading, poling field and temperature.

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