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Piezoelectric Properties of Diphase Ceramic-Polymer 0-3 Composites for Flexible Sensor Applications

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Abstract: ***Ferroelectric ceramic/polymer composites have the compliance of polymers which overcome the problems of brittleness in ceramics. By imbedding piezoelectric ceramic powder into a polymer matrix, 0-3 composites with good mechanical properties and high dielectric breakdown strength can be developed. The obtained composites of 0-3 connectivity would exhibit the piezoelectric properties of ceramics and flexibility, strength and lightness of polymer. These composites can be used in vibration sensing and transducer applications specially as piezoelectric sensors. In this paper, we have described the piezoelectric properties of 0-3 composites films of PZT- PVA prepared by hot press method. Composites were poled by applying different poling voltages. The piezoelectric properties of composite were analyzed by measuring piezoelectric coefficient (d_{33}) as a function of the PZT loading fraction. The results so obtained were found in good agreement with theoretical results as calculated from other models. Effect of poling time on piezoelectric properties of these composites was also studied and analyzed.***

Keywords: Piezoelectric effect, piezoelectric coefficient, polymer-ceramic composites, and hot press method.

1. Introduction

Composites made of ferroelectric ceramic and polymer offer several advantages over pure ceramics. Diphasic composites of PZT and different polymers and copolymers have been widely studied for various applications due to their appreciable piezoelectric properties. These composites can be easily made to large area to increase sensitivity as sensors; they are flexible that intimate contact with the objects to be detected is possible, this is particularly important in vibration sensing and transducer applications. The ceramics provides functional effects and the polymer matrix provide good protection of the ceramic phase while maintains the compliant mechanical property. The obtained composites exhibit piezoelectric properties of ceramics and flexibility, strength and lightness of polymers but electromechanical coupling coefficient, pyroelectric and piezoelectric charge coefficient, relative permittivity and other effective coefficients of these flexible composites are relatively low, which hinders the realization of practical use of the polymer composites.[1,2] Hence, extensive research is going on for optimizing properties of these composites using different ceramics and polymer matrices.[3-5] Different composites of PZT with various types of polymers such as PVDF, PVC and copolymers have been widely studied and reported in literature [6-8]. The properties of composite also depend on the morphological properties of ceramics like grain size, grain boundaries, pores, crystallinity and micro-crabs etc. In 0-3 connectivity composites, ceramic particles are dispersed in polymer matrix. These composites with 0-3 connectivity are amenable to mass production. Ceramic –polymer composites of 0-3 connectivity prepared by hot press technique. This technique of producing composite films is cost effective, easy and fast. In this paper, we have described and analyzed the piezoelectric properties of PZT/PVA composites films prepared by hot press method.

2. Experimental Details

The PZT powder was prepared by mixed oxide method in our laboratory. A measured quantity of PVA was dissolved in the solvent. A proportionate quantity of PZT powder by weight was dispersed in this solution and mixed thoroughly. The solution was stirred and given ultra-sonification treatment for proper mixing. The solution was then heated at 40°C for one hour until it became viscous and finally the solvent evaporates. The dried thick composite film so obtained was then cut, placed in the die and was hot pressed at 12 MPa and 200° C temperature for one hour using hot press technique. This technique of producing composite films is cost effective, easy and fast.

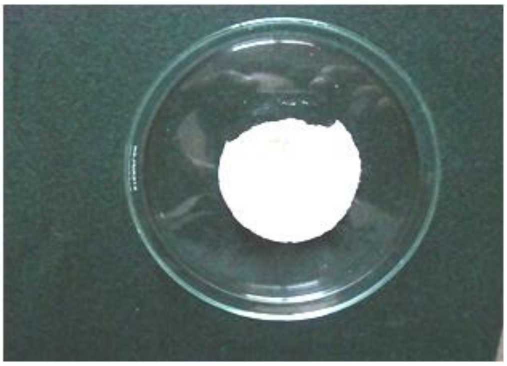

Figure 1: PZT – polymer composite obtained by hot press technique

The disc shaped 0-3 composites of PZT-PVA of thickness 200 μm with various ceramic volume fractions (ϕ = 10 % to 60%) were obtained as shown in figure 1

The electrodes on both sides of this composite were made by standard silver paste. For piezoelectric activation, the ceramic-polymer composites were poled under different poling conditions along the thickness direction in a silicon oil bath employing DC poling set up in the laboratory. To pole only the ceramic inclusions inside the composites, the poling is processed at temperature higher than the curie temperature (around 100°C) of polymers. The composites to be poled were first heated to 120°C. A field upto 15KV/mm was applied to the sample for 1 hr. The electric field was switched off at 120°C before cooling the sample at room temperature. As the field was switched off while the polymer is still in a paraelectric phase, hence, only the ceramic phase is poled. At room temperature, the sample have been poled under 15 KV/mm for another 1 hr. The composites were poled in a silicon oil bath at different poling voltages, time and poling temperatures. Piezoelectric characterization of these poled composites was carried out employing Piezo-Test PM100 computer interfaced system. We have also studied effect of poling time on Piezoelectric Charge Coefficient d_{33} . Effect of ceramic volume fraction on d_{33} of various composites was also studied.

3. Result & Discussion

Fig. 2 shows the variation of piezoelectric coefficient of PZT-PVA composites with different ceramic volume fractions. Piezoelectric ceramic gives the major contribution to piezoelectric properties in the composites. As volume fraction of ceramic increases, the contribution from the ceramic dominates the d_{33} and thus $d_{33 \text{ increases}}$ with increasing volume fraction.

The theoretical values of d_{33} coefficient were calculated using the relation given by Furukawa et.al as [9-11]

$$d_{33} = \frac{15\phi\varepsilon_p}{(2+3\phi)(1-\phi)\varepsilon_c}$$

Where ϕ is the PZT volume fraction, ε_p and ε_c are the dielectric constants of polymer and ceramic phases respectively. The experimentally observed values of d_{33} were found in good agreement with theoretical values as calculated by this model.

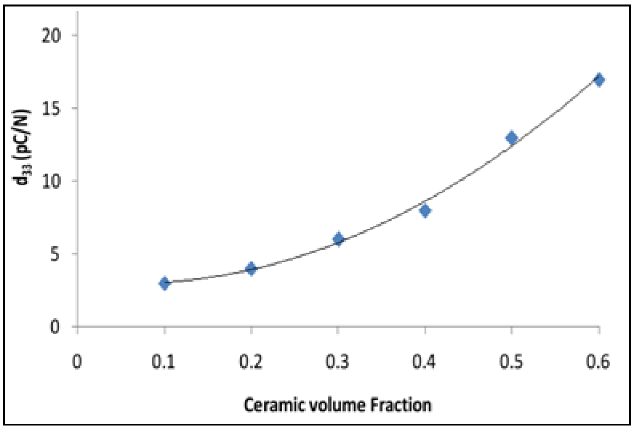

Figure 2: Variation of d33 for different ceramic volume fractions of PZT-PVA composites

We have also studied the variation of d_{33} and the poling time t at 2KV/mm. Under the condition of the same poling field and the poling temperature, the values of the d_{33} increase remarkably with increasing the poling time in the initial period of poling and saturates around 40 minutes and further as shown in fig 3. This is because the reversal of the 180° ferroelectrics domain takes place mainly at the initial stage of poling, which cannot give rise to local stress and is performed in short time. However, at anaphase, the reversal of 90° ferroelectrics domain plays an important role, which is accompanied by producing of the local stress and strain. This means that it is not easy to make the 90° ferroelectrics domain reversal.

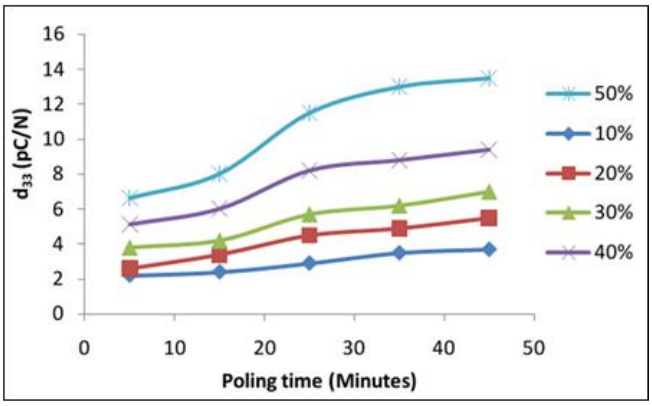

Figure 3: Variation of d_{33} with poling time for PZT-PVA composites

Therefore, the degree of poling can be improved by extending the poling time, but when the poling time exceeds 40 min, the d_{33} tends to be constant after a period of time because the orientation of the ferroelectrics domains is completed directionally.

We have also observed that these piezoelectric composites have limited temperature dependence compared to both PZT ceramics and polymers. We found that d_{33} of the composite remains almost constant (slight variation) with increasing temperature. This leads to a more stable output signal of a sensor made from a composite material, thus enhancing the sensorial properties of the component.

4. Conclusion

0-3 composites of PZT- PVA with different PZT volume fraction have been designed by hot press method. These composites were poled and then were characterized for their piezoelectric properties. The composites with high PZT volume fraction were found to have higher piezoelectric coefficient. The highest value of d_{33} obtained in this work is about 16.1 pC/N for composite with 60 % of volume fraction of PZT in the composite. We have also studied the effect of poling time on the piezoelectric properties of these composites. It was observed that piezoelectric coefficient increases with poling duration and then gets level off subsequently.

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