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Dielectric constant of barium titanate/cyanoethyl ester of polyvinyl alcohol composite in comparison with the existing theoretical models

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Abstract

A unique method has been introduced to measure the dielectric constant of polymer/ceramic composites using an effective medium instead of using the general methods of preparing bulk sintered pellets or films. In this work, a new and a simple method has been applied to measure the dielectric constant of polyvinyl cyanoethylate/barium titanate composites. The results are obtained by dispersing the ceramic powders in the polymer of a relatively low dielectric constant value. The dielectric constant of the composites is measured with varying ceramic volume percentages. The obtained results are compared with the many available theoretical models that are generally in practice to predict the dielectric constant of the composites. Then these results are extrapolated to comprehend the dielectric constant values of ceramic particles as these values form the base for the design of the composite. The precision and simplicity of the method can be exploited for predictions of the properties of nanostructure ferroelectric polymer/ceramic composites

Keywords: Dielectric constant; Barium titanate; Polymer/ceramic composite; Dielectric theoretical models

1. Introduction

Passive components in an electronic system are those electrical elements which support the active components and are characterized as resistors, inductors, and capacitors. Discrete passives are considered to be the major barrier of the miniaturization of electronic system. Assimilation of passives provides the components with the advantages like better electrical performance, higher reliability, lower cost, and improved design options [1]. Currently, interest in passive components is increasing for miniaturization and better electrical performance of electronic packages. Among various kinds of passives, focus is on decoupling capacitors, which are used for simultaneous switching noise suppression [2]. The science of embedded capacitor is a sophisticated technology with the congregation of both performance and functionality requirements for future electronic devices. One of the major hindrances for implementing this technology is the lack of dielectric materials with promising dielectric properties. Polymer based composite is considered as a solution to the problem mentioned hitherto. Developing a composite with compatible high dielectric constant material is the major challenge of the integral capacitor technology. Polymer/ceramic composites can be used in forming capacitors because they combine the process ability of polymers and high dielectric constant of ceramics. One of the promising embedded capacitor materials is a polymer/ceramic composite which is a ceramic particle-filled polymer. It is a material utilizing both high dielectric constant of ceramic powders and good process ability of polymers. Particularly epoxy/ceramic composites have been investigated and stud-

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ied due to their compatibility with printed writing boards (PWB) [3–9]. It is very important for composite material design to precisely understand the dielectric constant of ceramic particles. Many methods and models, with several quantitative rules, have been developed to predict the dielectric constant of heterogeneous two component composites counting on the basis of dielectric properties of each component, i.e., both ceramic and polymer [10–13]. However, while different models have been developed, usually little or no experimental evidence was provided to support the derived equations. So ambiguity still prevails in which model is more useful for the prediction of the effective dielectric constant of the composites.

Polymers filled with ceramics have been studied for use as dielectric materials in thick film capacitors [14]. Ceramic particle size influences the effective dielectric constant of composite dramatically. Precise prediction of the effective dielectric constant of polymer/ceramic nanocomposites forms the focal point for the design of composite materials. Many theoretical models have been proposed in the literature for simulating the electrical properties of the composites. Mostly, composite dielectrics are statistical mixtures of several components. The models mentioned are empirical models to describe the polymer/ceramic nanocomposite property. Other efforts also have been made to predict the dielectric properties of composite using percolation theory [15–18]. The major interest in the physics of disordered materials lies in relating the macroscopic property of interest like permittivity, conductivity, etc. The effective-medium theory (EMT) is also used to set up a numerical model that can precisely predict the dielectric constant of polymer/ceramic nanocomposite [22]. The major factors that affect the dielectric properties of barium titanate ceramics are the grain size, phase contents and the types of dopants used [19–21]. Thus the dielectric property of composite can be treated in terms of an effective medium whose dielectric permittivity can be obtained by a suitable averaging over the dielectric permittivity of the two constituents [25]. For polymer/ceramic composites, the perovskite-type barium titanate is in the powder form instead of the sintered form. The removal of grain boundaries, elimination of constrained forces from neighboring grains and a drop in domain density due to decrease in the particle size will reduce the dielectric constant of BT powders [23,24]. Hence, sintered and unsintered powders of BT show a different dielectric behavior. Our work deals with a composite medium composed of dispersed unsintered ceramic within the polymer with the sole intention of minimizing voids or pores. Though this is a relative way of characterizing the composite for dielectric constant values, the method seemed interesting and reliable for measuring the dielectric constant values of the composites. Then these results are used to extrapolate linearly only to achieve the dielectric constant value of the unsintered ceramic powder. The method that is followed is basic and uncomplicated when compared to many methods that are already in existence.

2. Materials and procedure

The polymer/ceramic composites are prepared using the commercial ceramic powder, Cabot BT-8 (BT), (hydrothermal powder with a mean particle size of 0.2 μm obtained from Cabot Performance Materials, Boyertown, PA), a cyanoethyl ester of polyvinyl alcohol (CEPVA) kindly provided by Plastpolymer J.S.Company via St. Petersburg State Institute of Technology, Russia, castor oil, (Eur. Pharm. grade), having a density of 0.957 g/cc obtained from Acros Organics and BYK-W 9010 from BYK-Chemie which is a dispersant used for a better dispersion of the ceramic powder. *N*,*N*-dimethyl formamide (DMF) from Fisher and 2-methoxyethanol from Aldrich were used as the solvents for the polymer without any prior treatment and further purification. DMF and 2-methoxyethanol were mixed in a 1:1 volume ratio and the solid polymer was dissolved maintaining the temperature at around 65 °C. The amount of solid polymer added was adjusted to get a final polymer concentration of 30% by weight. After dissolution, the solution was cooled to room temperature and magnetic stirring was continued for 12 h in a teflon jar to obtain a clear transparent pale yellow solution. The solution was stable over a period of several weeks and did not show any signs of turbidity. In a different teflon jar, a 50% by weight suspension of the commercial ceramic powder was prepared by agitation by magnetic stirring in a similar mixed solvent of 1:1 DMF and 2-methoxyethanol. The use of castor oil as a matrix was due to its frequent availability, chemical/thermal inertness, relatively high dielectric constant and good binding action. The main reason for using CEPVA as the polymer matrix was because of its high dielectric constant, 22, and that of castor oil is 5.

Initially, castor oil and the BT ceramic powder were mixed in different proportions. These mixtures had a variable ceramic content of 10–50% by volume on the dry basis. Next, different amounts of both polymer solution and the BT suspensions were then mixed and taken into different small containers and the mixtures were adjusted to contain the final polymer/ceramic weight ratios shown in Table 1. The prepared mixtures were then gently dried at 80°–90 °C under both continuous magnetic stirring and mildly reduced pressure to get rid of the solvents where viscous slurries/pastes were obtained. The dried composites were then kept under reduced pressure and used for further characterization. The capacitor is fabricated using the same procedure that was followed in our earlier work [26] and is characterized for capacitance. Dielectric constants of the

Table 1

Composite	Ceramic wt%	Polymer wt%
A	70 with 29 vol%	30
B	75 with 34 vol%	25
C	80 with 41 vol%	20
D	85 with 49 vol%	15