

Studies of Dielectric Constant of Some Polymer/Ceramic Composite Materials: A Conformational Study

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Abstract- Polymer/ceramic composites are the most promising embedded capacitor material for organic substrates application. Predicting the effective dielectric constant of polymer/ceramic composites is very important for design of composite materials. In the present paper the dielectric constant has been evaluated by various mixing rules/models for polymer-ceramic composites and compared with experimental values taken from literature. Results have been discussed on the basis the value of average percentage deviation of dielectric constant.

Keywords- Dielectric Constant, Mixing Rule, Polymer/Ceramic Composition, Average percentage deviations, material.

I. INTRODUCTION

The advances in natural science follow two inseparably interrelated paths: experimental and theoretical. Accumulation of an appropriate amount of experimental data allows drawing generalized conclusions, deriving appropriate formulas and formulating laws that govern the studied phenomena. This leads to the possibility to predict and program expected effects for practical use. Obviously, such a research cycle requires, particularly obtaining information about molecular interaction and stereochemical effects, the theoretical results to be verified by experimental results.

Engineers consistently demand new material systems for specific applications. This demand dictates that material scientists develop new material systems. The modern applications require diverse and specific properties in materials which cannot be met in single-phase materials. The composites contain two or more chemically different materials or phases. In these materials, it is possible to tailor electrical and mechanical properties catering to a variety of applications.

Dielectric constant of a composite is determined for applications and for understanding the nature of the interactions between the constituents of the composite. There are many theoretical studies on dielectric constant of composites in order to describe its dependence on the volume fraction of the filler particles, the dielectric constant of polymer and filler and the possible interaction between both constituents [1-6]. Accuracy of different theoretical models is necessary for designing of ceramics polymer composites for various applications.

In the literature, different theoretical models and mixing rules like Jayasundere and Smith, Lichtenecker logarithmic, Maxwell Garnett, Sillar and Yamada are typically applied to different ceramic/ polymer composite systems, with the models introducing different physical fundamentals for describing the interactions between the ceramic and the polymer [7-10].

II. THEORY

The dielectric behavior of polymer/ceramic composition systems has been analyzed by many scientists and many equations have been derived based on experimental results and theoretical derivation. An attempt has been made to compute dielectric constant theoretically for the polymer – ceramic composition.

The most commonly used equation is the **Lichtenecker logarithmic** law of mixing and is written for a two-component system as

$$\log \varepsilon = v_p \log \varepsilon_p + v_c \log \varepsilon_c \quad (1)$$

$$\log \varepsilon = \log \varepsilon_p + v_c (1 - k) \log \varepsilon_c / \varepsilon_p \quad (2)$$

Jayasundere and Smith [11] have worked together in deriving an equation which was modified from the well-known Kerner equation by including interactions between neighboring spheres for the measurement of dielectric constant of binary composites and the equation is shown in equation

$$\varepsilon_{eff} = \frac{v_p \varepsilon_p + v_c \varepsilon_c \left[\frac{3\varepsilon_p}{\varepsilon_c + 2\varepsilon_p} \right] \left[1 + \frac{3v_c(\varepsilon_c - \varepsilon_p)}{\varepsilon_c + 2\varepsilon_p} \right]}{v_p + v_c \left[\frac{3\varepsilon_p}{\varepsilon_c + 2\varepsilon_p} \right] \left[1 + \frac{3v_c(\varepsilon_c - \varepsilon_p)}{\varepsilon_c + 2\varepsilon_p} \right]} \quad (3)$$

The Maxwell-Garnett mixing rule was initially used in a system where metal particles are encapsulated in an insulating matrix [12]. But in recent times the same mixing rule is applied for ceramic particle inclusions. This mixing rule is then modified and the effective dielectric constant for a polymer/ceramic composite incorporating homogeneous distribution of spherical ceramic material can be determined by the equation developed by Maxwell and Wagner [13] which is known as **Maxwell- Wagner** mixing rule

$$\varepsilon_{eff} = \varepsilon_p \left[\frac{2\varepsilon_p + \varepsilon_c + 2v_c(\varepsilon_c - \varepsilon_p)}{2\varepsilon_p + \varepsilon_c - v_c(\varepsilon_c - \varepsilon_p)} \right] \quad (4)$$

Yamada have studied the polymer/ceramic binary system and proposed a model using the properties of its constituent materials [14]. Considering the system to comprise ellipsoidal particles dispersed continuously, the dielectric constant is given by the equation

$$\varepsilon_{eff} = \varepsilon_p \left[1 + \frac{\eta v_c(\varepsilon_c - \varepsilon_p)}{\eta \varepsilon_p + (\varepsilon_c - \varepsilon_p)(1 - v_c)} \right] \quad (5)$$

$$\varepsilon_{eff} = \varepsilon_p \left[1 + \frac{v_c(\varepsilon_c - \varepsilon_p)}{\varepsilon_p + n(\varepsilon_c - \varepsilon_p)(1 - v_c)} \right] \quad (6)$$

where $n = 0.2$ and $v_p =$ volume fraction of polymer, $v_c =$ volume fraction of ceramic, $\varepsilon_p =$ dielectric constant of polymer, $\varepsilon_c =$ dielectric constant of ceramic.

For the case of spherical dielectric inclusions embedded in an isotropic dielectric medium, Sillars found the following equations,

$$\varepsilon = \varepsilon_1 \left[1 + \frac{3v_2(\varepsilon_2 - \varepsilon_1)}{2\varepsilon_1 + \varepsilon_2} \right] \quad (7)$$

III. RESULTS AND DISCUSSION

Dielectric Study

Dielectric constants of BaTiO₃/polymer and BaTiO₃/epoxy were calculated from various existing mixing rules. Maxwell – Garnett and Rayleigh’s models have the same solutions, so just Maxwell – Garnett is shown. Dielectric constant of pure polymer and BaTiO₃ ceramics have been taken from literature [15,16]. The correlation between theoretical models is presented in Figures 1 to 3. In each figure, the dielectric constant of the composites is plotted as a function of volume fraction of the inclusions. The theoretical predictions are drawn and are represented along with the experimental results for different particle size in order to investigate the agreement between them.

The experimental and predicated values of dielectric constant (ε) using five mixing rules for BaTiO₃/Poly (ethylene glycol) diacrylate (PEGDA) composite at different frequency are presented in Figure 1. It is clear from the Figure 1 (a) and (b) that out of five mixing rules/models, Lichtenecker model predicts ε values best in term of average percentage deviations, while Jayasundere and Yamada model are near to the experimental values over the whole composite range and Maxwell- Garnett and Sillar models show a very distinct behaviour as compared with other models. It is observed from Figure 1 (c) that dielectric constant evaluated using Jayasundere and Lichtenecker models are close to experimental results, however other models/mixing rules show more deviation from the experimental values.

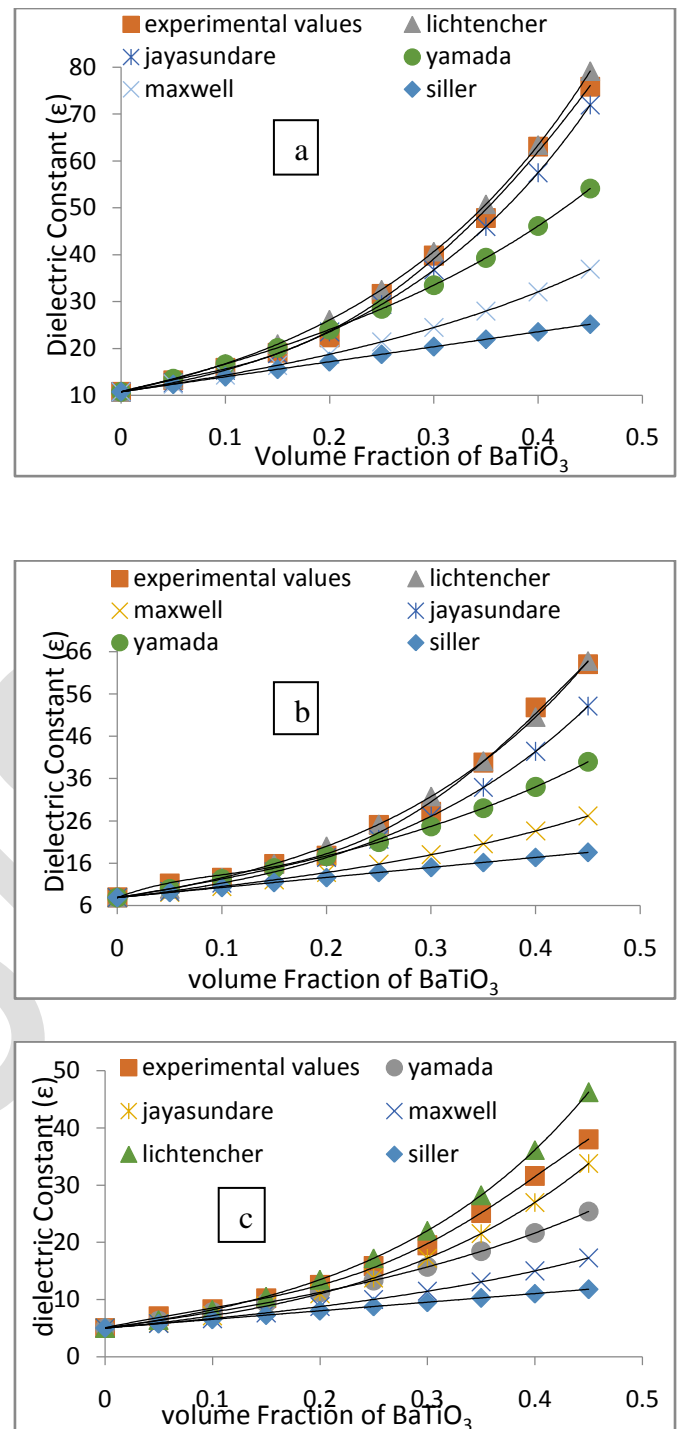


Figure 1. Comparison of the experimental and theoretical values of Dielectric constant of BaTiO₃/PEGDA with volume fraction of BaTiO₃ at (a) 1KHz (b) 1 MHz and (c) 1GHz frequency

Figure 2 is the graphical depiction of the dielectric constant of BaTiO₃/ trimethylolpropane triacrylates (TMPTA) computed by various mixing rules and shows the relative deviation from the experimental data. The values of ε calculated from the lichtenecker logarithmic law are found to be in good agreement with the experimental values of the BaTiO₃/TMPTA composite materials.

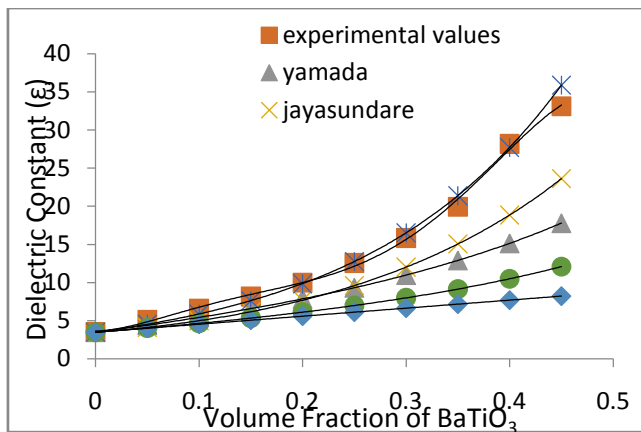


Figure 2. Comparison of the experimental and theoretical values of Dielectric constant of BaTiO₃/TMPTA with volume fraction of BaTiO₃ at 1GHz frequency

Figure 3 shows the effective dielectric constant of two composites BaTiO₃/ epoxy with the 20 volume % ceramic filler. The ceramic – epoxy composites were fabricated using Ba_{1-x} Sr_x TiO₃ (x=0.2 and x=0.4) powder mixed with Bisphenol an epoxy. Different mixing rules such as Maxwell- Garnett, Sillar, Jayasundere & Smith, Yamada and Lichtenecker models used to compute ϵ values and compared with the experimental results.

The Lichtenecker model fits better with the experimental results for both composites (Figure 3). In case of BST (0.2) epoxy composite it is noticed that Lichtenecker model gives small deviation.

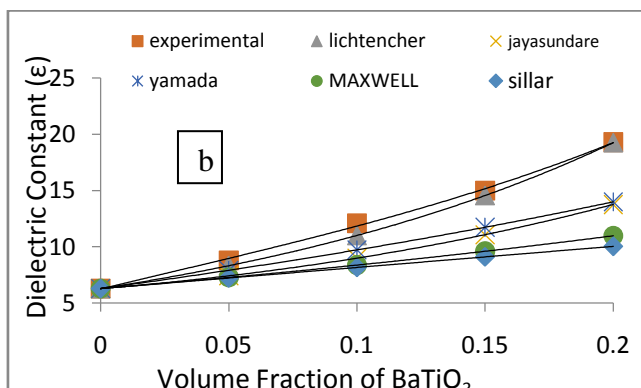
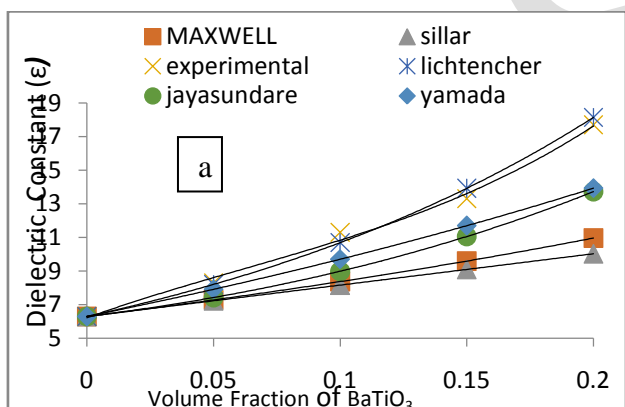


Figure 3. Comparison of the experimental and theoretical values of Dielectric constant of BaTiO₃/Epoxy thick film with volume fraction of BaTiO₃ at (a) BST (0.2) (b) BST (0.4)

Table 1 Average percentage deviations of the values of dielectric constant calculated using different mixing rules.

Compositions	Lichtenecker	Jayasundare	Yamada	Maxwell	Sillar
BaTiO ₃ /TM A	1.313	22.608	25.831	39.942	45.733
BaTiO ₃ /PEGDA at 1 GHz	-6.886	12.109	16.228	32.644	39.510
BaTiO ₃ /PEGDA at 1 MHz	-1.158	10.948	14.878	31.496	38.378
BaTiO ₃ /PEGDA at 1 kHz	-4.901	3.556	7.896	25.923	33.427
BaTiO ₃ / Epoxy thick film (0.4)	-3.517	19.223	15.755	12.698	27.516
BaTiO ₃ / Epoxy thick film (0.2)	-0.633	14.080	10.420	10.400	23.132

The average percentage deviations of dielectric values for different composites of polymer/ceramic are given in table 1. A close perusal of table 1 reflects that Lichtenecker model is best suitable for all the compositions with the minimum percentage deviation - 0.633 and maximum percentage deviation -6.886 for the composition ceramic/epoxy thick film (0.2) and BaTiO₃/PEGDA respectively, while Jayasundare and Yamada models give slightly large deviation for all composition except BaTiO₃/PEGDA at 1 kHz.

IV. CONCLUSION

The Lichtenecker logarithmic rule and Jayasundere rules used for estimation the dielectric constant show good agreement with the experimental values of dielectric constants of polymer / ceramic compositions, BaTiO₃/PEGDA at 1 GHz, BaTiO₃/PEGDA at 1 MHz, BaTiO₃/PEGDA at 1 KHz, BaTiO₃/TMPTA, BaTiO₃ / Epoxy thick film (0.4) and BaTiO₃ / Epoxy thick film (0.2) while Maxwell- Garnett and Sillar have very distinct behaviour when compared with other rules. Out of five mixing rules Lichtenecker is best suited in term of average percentage deviations, while Jayasundere and Yamada are near to the experimental values over the whole composite range.

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