SOME OF DIELECTRIC PROPERTIES OF POLYMER/FERROELECTRIC COMPOSITES

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ABSTRACT

$BaTiO_3$, synthesized in polycrystalline form, using stoichiometric mixture of oxide with traditional ceramic technique and characterized by X-ray diffraction, tetragonal structure of prepared powder were found. Moreover, particle size of ceramic powder determined using laser particle size analyzer. The particle size distribution was found in the range (0.427-3.195)µm. The epoxy ceramic composite system was prepared. $BaTiO_3$ with content 10%, 20%, 30%, 40%, 50% wt and 60% wt. respectively used as filler. Permittivity of the composites was investigated at frequencies from 2KHz to 5 MHz The dielectric loss tangent (tanδ) was determined as a function of frequency, compositions. It was found that the weight percent of $BaTiO_3$ filler in ceramic/epoxy composite effect on the dielectric properties. At low frequencies, $BaTiO_3$/Epoxy composites have much higher dielectric constant. Dielectric constant increases considerly with the addition of $BaTiO_3$ fillers, however, the higher conductivity of $BaTiO_3$ than epoxy lead to an increase in ac conductivity with increases $BaTiO_3$ content, the increase in both properties was attributed to the presence of the high permittivity ferroelectric phase

Keywords: Polymer composite, ferroelectric, $BaTiO_3$, dielectric behavior, XRD

الخلاصة

تم تحضير مركب متعدد التبلور$BaTiO_3$ باستخدام خليط متجانس للاكاسيد المكونة، واستخدمت التقنية التقليدية في تحـضير الـسيراميك وتشخيص المركب باستخدام تقنية حيود الأشعة السينية وقد وجد المسحوق السيراميكي المحضر رباعي التركيب، علاوة على ذلك تم قياس

المقاس الحبيبي للمسحوق السيراميكي باستخدام محلل المقاس الحبيبي الليزري. وجد توزيع المقاس الحبيبي في مدى مابين (0.42-3.195) مايكرون. وحضر نظام متراكب للبيوكسي- سيراميك، اضيف تيتانيت الباريوم بنسبة وزنية 10%،20%،30%،40%،50% و60% على التوالي كمالئ. فحص ثابت العزل كدالة للتردد مابين 2 كيلوهرتز -5 ميكاهرتز"،معامل الفقد ايضا دالة لنفس التردد بالإضافة الى نسبة المكونات. وجد بان النسبة المؤية الوزنية لمتراكبات اليوكسي/سيراميك يوثر على خواص العزل. عند الترددات المنخفضة فان ثابت العزل اعلى لمتراكبات الييوكسي /تيتانيت الباريوم. ثابت العزل يزداد باضافة تيتانيت الباريوم، الزيادة في كلتا الخاصيتين تعزى الى السماحية العالية للطور الفيروكهربائي.

INTRODUCTION

Development of electronic devices working at high operating frequencies, such as fast computers, integrated capacitors, cellular phones[Bhattacharya et al 2000, Popielarz et al 2001], etc., require new high-dielectric constant materials that combine good dielectric properties with both mechanical strength and ease of processing [Chanmal et al 2008]. In particular, the high dielectric constant materials are required for making embedded capacitors for integrated electronic devices [Hamami et al 2006]. The unique combination of dielectric and mechanical properties is hard to device in one component material. Pure polymers are easy to processes into mechanically robust components but generally suffer from low dielectric constant [Dimos et al 1994].On the other hand, typical high constant materials, such as ferroelectric ceramic are brittle and require high-temperature processing which is often not compatible with current circuits integration technologies. The ideal solution would be a high dielectric constant material that is mechanically robust and processable at ambient temperatures. The unusual dielectric properties of Barium Titanate make this ferroelectric compound at the ambient temperature a significant material for electrical engineering. Because of their high permittivity and weak dielectric losses, ferroelectric materials are promising for microwave applications, such as capacities with strong permittivity, accordable filters and resonators, and modulators of frequency. As a consequence of their wide application there is continued interest in the development of new synthetic method .

Indeed, the best-known peroviskites of the ABO_3 type, $BaTiO_3$ ceramic are interest material to their ferroelectric and dielectric properties [Muhammad et al 2005. Nowosielski et al 2008].

In this paper the effect of filler amount to make polymer/ceramic composite, on the capacitor properties such as dielectric constant, dielectric loss factor, were investigated using $BaTiO_3$ particles as a ceramic and epoxy resin as a polymer.

EXOERIMENTAL WORK

The matrix material used for fabrication of composite consist of low temperature curing epoxy resin system was Quickmast 105(DCP).It has low viscosity, and corresponding hardener. Resin and hardener were mixed in a ratio 1:3 weight as recommended by the supplier.

$BaCO_3$ supplied by BDH chemicals Ltd Pool (England) with purity >99% and TiO_2 supplied by redel-De Haen Germany Company with high pure (more than 99%) used as starting materials for prepared $BaTiO_3$ by the conventional ceramic technique [Mostaghaci et al 1986]. Particle size analyzes of prepared $BaTiO_3$ carried out using a particle size analyzer from Shimadzu made in Japan.

Resin and hardener were mixed in appropriate proportion after very slow mixing to avoid formation of bubbles in the resin system. The $BaTiO_3$ ceramic powder were mixed with the epoxy resin with weight ratios (10wt%,20wt%,30wt%,40wt%,50wt%, and 60wt%). Mixture was stirred for prolonged time at slow speed to ensure good mixing and wetting of all components by resin and get uniform quality dough. The dough was then transferred to the moulds and was allowed to cure at room temperature for 24 hour then used oven at 50° C for 2 hours to prepare the composite samples.

The real and imaginary parts of the dielectric constant as electrical conductivity measurements were carried out using LCR meter model 4194A made in Japan.

The electrical measurements were carried out in the range from 20 KHz to 5MHz, using LCR meter. The dielectric constant ε_r was calculated using the formula

$$\varepsilon_r = \frac{C \times d}{\varepsilon_o \, A} \tag{1}$$

Where, C is capacitance of pellet in Farad, d the thickness of pellet in meter. A, is the cross section area of the flat surface of the pellet and ε_o is a constant of permittivity of free space. Also, the value of loss factors for test samples is given by tan δ.

In addition, Ac conductivity was carried out for all composite samples at room temperature in the same range of the frequency, $\sigma_{a.c}$ measurement can be calculated by following the equations [Viswanathan et al 1990, Sucher 1963]

$$\sigma = 2\pi f \varepsilon_o \varepsilon_i \tag{2}$$

Where, σ is the conductivity $(\Omega.m)^{-1}$,ε_i is imaginary part of dielectric constant (F/m), f is the frequency (Hz).

$$\omega = 2\pi f \tag{3}$$

ω: is angular velocity

RESULTS AND DESSCUSION

The X-ray diffraction pattern for preparing ferroelectric ceramic powder examined in the range of (2θ) was (20°-60°). Existence of tetragonal $BaTiO_3$ phases with reflection planes (100), (101), (111), (002), (200), (102), (201), and (211) as shown in Fig.(1),one can compare these values with Miller indices of standard tetragonal $BaTiO_3$ using table (1).

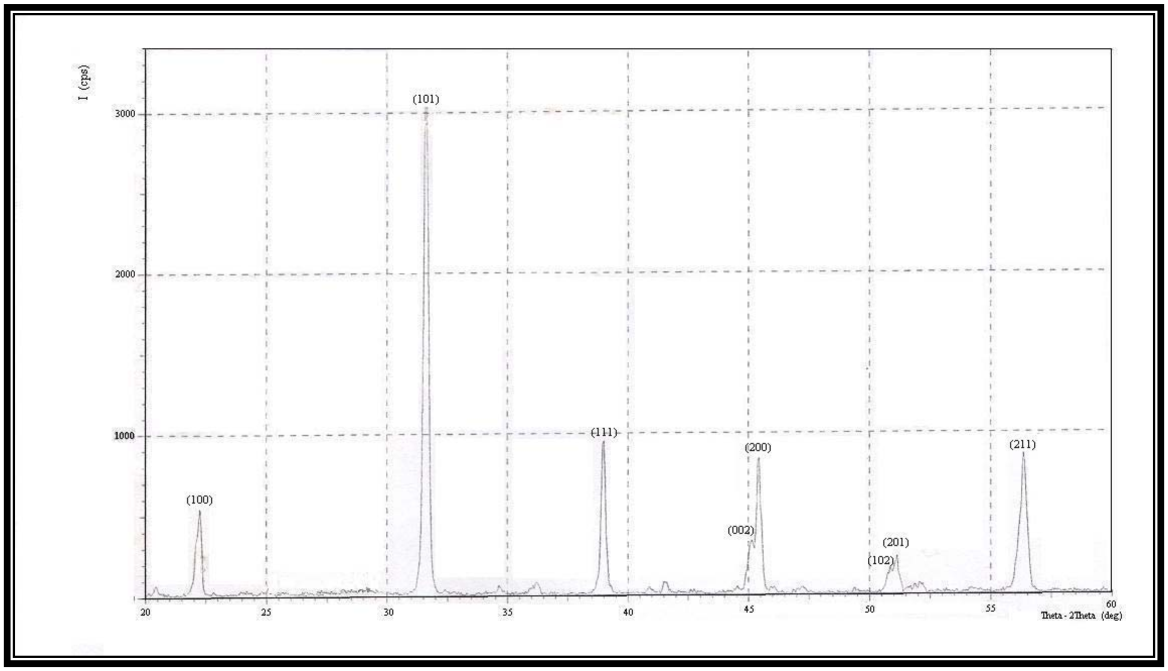


Fig (1):X-Ray diffraction pattern of prepared $BaTiO_3$ powder.

Table (1): Miller indices of standard tetragonal $BaTiO_3$

2θ	Int	hkl
22.262	25	100
31.497	100	101
38.887	46	111
44.855	12	002
45.377	27	200
50.613	6	102
50.976	8	201
56.251	35	211

The particle size distribution curve of prepared $BaTiO_3$ powder as shown in Fig. (2). The size distribution appears to be between (0.427 μ-3.195μ). The influence of Barium titanate additives on the dielectric constant of composite was studied in the frequency range of (20KHz-5MHz). Figure (3) shows the dependence of the dielectric constant of the composites on frequency for different weight fractions of Barium titanate at ambient temperature. In the case of $BaTiO_3$/Epoxy matrix, the dielectric constant decreases linearly with the frequency for all the $BaTiO_3$ concentrations. At low frequencies, $BaTiO_3$/Epoxy composites have much higher dielectric constant for all samples. It is perceptible that higher concentration of ferroelectric filler lead to higher dielectric constant composites. An important observation is that dielectric constant increases considerly with the addition of $BaTiO_3$ fillers in epoxy resin, thus resulting is the higher dielectric constant of the composites.

It obviously that there is a large contrast between the dielectric constant of the pure polymer and components, especially at high $BaTiO_3$ filler content.

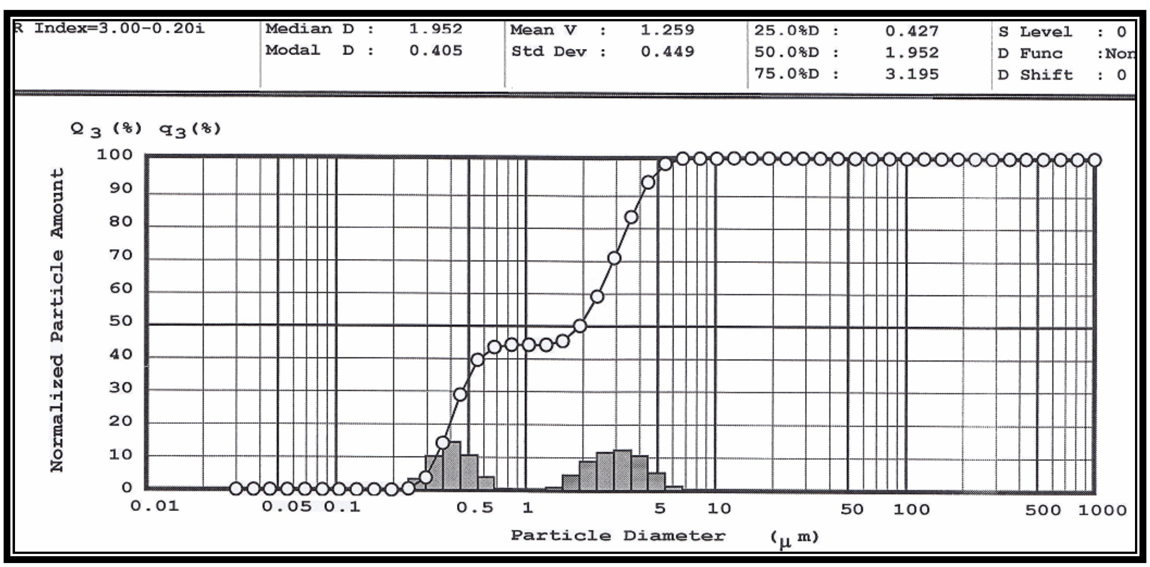
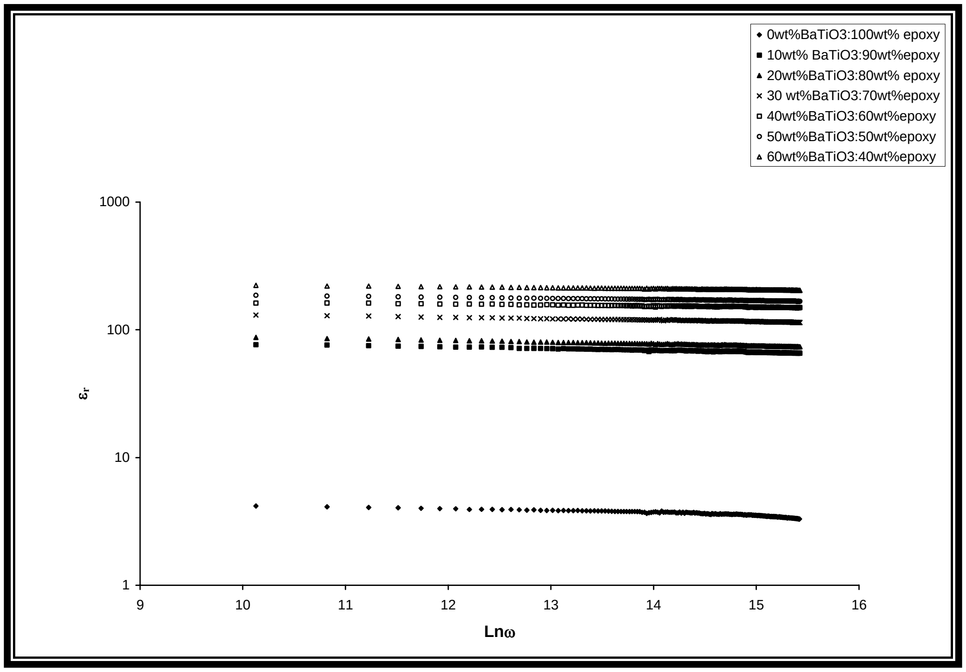


Fig (2): Particle size distribution of prepared $BaTiO_3$ powder.



Fig(3):Variation in dielectric constant to weight fraction for $BaTiO_3$-Epoxy system as a function of frequency.

The increase in conductivity for epoxy-$BaTiO_3$ composites was due to an increasing weight fraction of $BaTiO_3$, However, $BaTiO_3$-epoxy Composites consist of two relatively insulating phases, as soon from the low frequency conductivities of pure $BaTiO_3$ and epoxy. Using microstructural electrical networks [Almond et al 2004] it has been shown that the ac conductivity of insulating phases, $\omega\varepsilon\ \varepsilon_{\circ}$, can contribute to the conductivity in power law region, where ε is the permittivity of the phase (epoxy or $BaTiO_3$) and $\varepsilon_{\circ}$ is permittivity of free space. Since the permittivity of $BaTiO_3$ is much greater than epoxy, the higher $\omega\varepsilon_r\varepsilon_{\circ}$ of $BaTiO_3$ lead to an increase in ac conductivity with increase $BaTiO_3$ content as shown in Fig.(4).

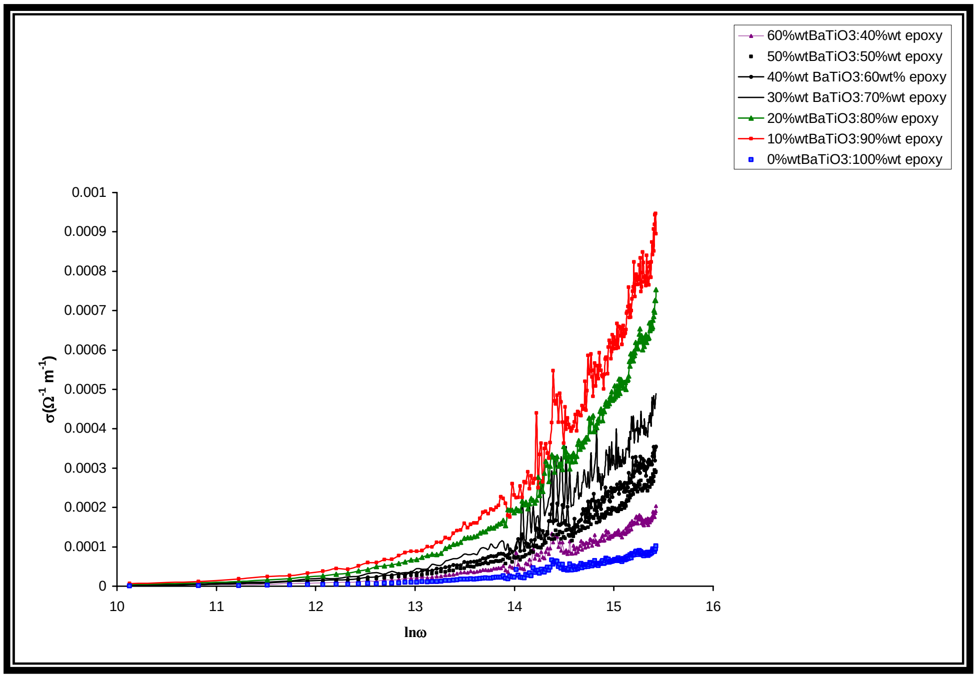


Fig.(4):Variation in conductivity to weight fraction for $BaTiO_3$-Epoxy system as a function of frequency.

The magnitude of dielectric loss is an important material parameter for making capacitors. Ideally, in a capacitor the dielectric losses should be as low as possible. Figure (5) shows the variation of loss tangent (tanδ) with frequency at ambient temperature for all the preparing samples, the value of dielectric dispersion decrease with increase frequency in all the samples.

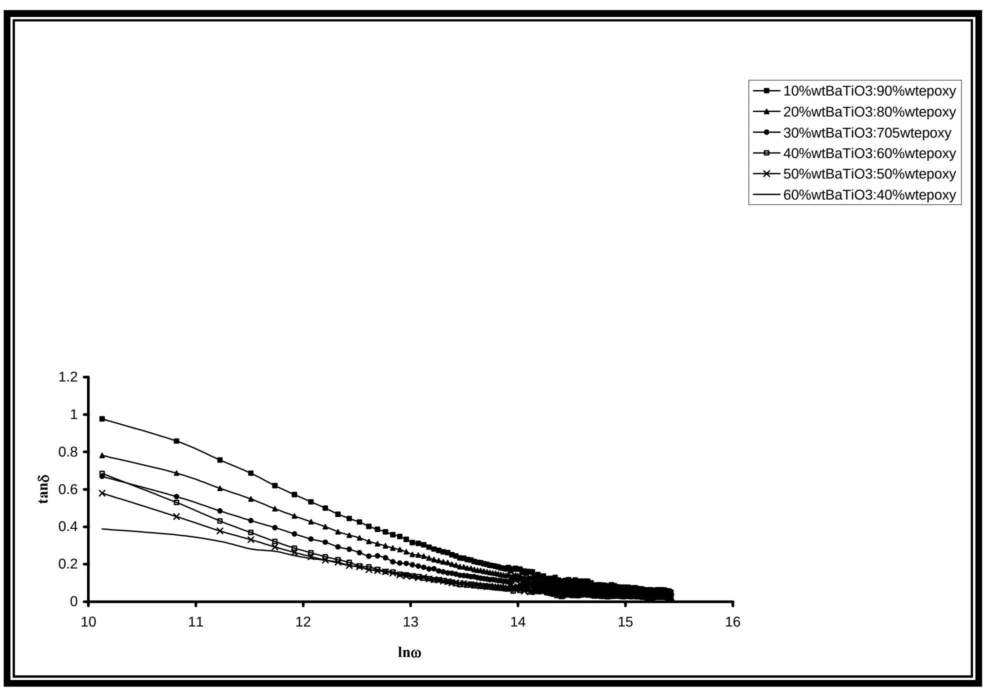


Fig.(4):Variation in loss factor to weight fraction for $BaTiO_3$-Epoxy system as a function of frequency.

CONCLUSIONS

- This study points to the fact that upon introduced of $BaTiO_3$ filler, there is an improvement in the dielectric properties
- Frequency dependence of dielectric constant, desperation factor and resistively in pure resin and polymer ceramic composites with various concentration of ferroelectric powder as a filler has been studied in the frequency range 20 KHz-5 MHz at ambient temperature. The experimental results indicate that addition of filler particles to epoxy leads to increase in the dielectric constant of the composite specimens The decrease in dispersion factor with increase of frequency is due to the orientation polarization.

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