

SYNTHESIS AND TAILORING THE OPTIMIZATION AND DIELECTRIC PROPERTIES OF PMMA/PEG/BARIUM TITANATE HYBRID NANOSTRUCTURES FOR ENERGY STORAGE AND ELECTRONICS APPLICATIONS

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Abstract. The present work aims to improve the dielectric properties of poly-methyl methacrylate (PMMA)-polyethylene glycol (PEG) blend doped with barium titanate (BaTiO₃) nanoparticles to be useful in electronics and dielectric fields. The PMMA/PEG/BaTiO₃ nanocomposite films were fabricated by using the casting method. The morphological and dielectric characteristics of PMMA/PEG/BaTiO₃ nanocomposite films were tested. The dielectric characteristics were examined by LCR meter at frequencies ranging from 100 Hz to 5 MHz. The results of dielectric characteristics confirmed that there is a growth in the dielectric parameters of PMMA/PEG blend with increasing BaTiO₃ NPs content. The dielectric properties of PMMA/PEG/BaTiO₃ nanocomposite films were distorted with a boost of the frequency. Finally, dielectric characteristics show that the PMMA/PEG/BaTiO₃ nanocomposite films can be utilized in various electrical and nanoelectronics applications with high energy storage, lightweight, low cost, and flexibility.

Keywords: BaTiO₃, dielectric properties, PMMA/PEG, nanoelectronics, blend.

1. Introduction

Researchers are very interested in polymer nanocomposites because of their impressive mechanical

and thermal characteristics, as well as their great performance. Improved mechanical strength and dimensional stability, good optical, magnetic, and electrical properties, thermal stability, meaningful flame retardancy, chemical resistance, increased anti-scratch and wear resistance, *etc.*, are just a few of their outstanding properties¹. High-dielectric materials are becoming more popular due to the rising need for electronic devices and capacitors. The processing simplicity, flexibility, and strong mechanical characteristics of polymer materials are their strengths, but their dielectric qualities are often lacking. Since then, there has been a flurry of activity in the field of high dielectric composite preparation via the use of high dielectric fillers². It is now common practice to combine many polymers into a single material to create novel polymeric materials with enhanced physical and chemical characteristics. Several variables, including the mixing technique, the ratio of polymers used, the mixing temperature, and the amount of mixing of each polymeric component, affect the finished product characteristics. Due to its shape and internal structure, which are governed by the degree of polymer dispersion, the immiscible polymer phase has a significant impact on the material's rheological and mechanical characteristics. Nanoparticles interact with the functional groups in the polymer mixture, improving their characteristics when added to the mixture^{3, 4}. From a technical standpoint, organic-inorganic hybrid optical materials are very useful. Once the connections between inorganic element properties and polymer matrices are understood, hybrid compositions with extraordinary characteristics may be created. Another important topic of applied materials science is polymeric nanocomposites, which are known for their unique set of characteristics. Interesting optical properties of polymer composites include a high or low refractive index, absorption or emission spectra that are suited to the material, and severe optical nonlinearities. Due to their unique characteristics, hybrids have the potential to be used in optoelectronic devices⁵.

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Coatings and materials for many components in aviation, marine, car, and biomedical applications are two common uses of polymeric composites in human life. Composites are better than other materials because they possess ideal electromagnetic and elastic characteristics in addition to excellent resistance to fire, impact, and abrasion. They also have excellent mechanical qualities. Since the advent of 3D printing, there has been a meteoric rise in the popularity of polymeric composites. New opportunities for the development of smart materials in a variety of applications are opened up by this technology. Thermoplastic, thermosetting, and elastomer polymers are only a few examples of the many possible matrix materials for the production of polymeric composites^{6,7}.

Interesting electrical properties are produced by combining insulating polymers with conducting fillers. Powerful interfaces are formed when the insulating polymer matrix and conducting filler contact, and these interfaces both actively and passively affect the composite's dielectric behavior⁸. A revolutionary shift has been occurring in electronics, with a focus on investing in gadgets that are lightweight, soft, and flexible as opposed to big, heavy, and stiff. So, new commercial goods like screens, solar cells, and biological sensors might be developed in the rapidly expanding area of flexible electronics. These technologies have the potential to completely transform our daily lives when embedded in clothes and other commonplace products. This creates new possibilities for the electronics sector, and the constant development of thin-film materials and devices in manufacturing will eventually lead to the pervasiveness of flexible electronics⁹.

Polymethyl methacrylate (PMMA) is a cheap, amorphous polymer that is chemically stable, very transparent, and very hydrophilic. Researchers in the vast subject of material science love it for its outstanding gelatinizing and high solvent qualities, as well as its superb outdoor weather ability and excellent environmental stability. Its surface resistance is strong, and it resists acids well. Some of its characteristics, such as thermal stability and ionic conductivity, are limited, despite its great attributes. This polymer's popularity in several technical and industrial domains is owing, in part, to its exceptional optical quality, which is a result of its transparency in the visible range. Films made of PMMA that include inorganic or organic elements have found widespread use due to the desirable qualities they impart, including photoluminescence, nonlinear optical characteristics, and photoconductivity^{10,11}.

PMMA, or poly(methyl methacrylate), is a kind of acrylate polymer. Thanks to its great impact strength, weather resilience, and scratch resistance, this optically clear thermoplastic has found widespread application as a replacement for inorganic glass. Doping and combining

with metallic nanoparticles cause it to carry electric charges, despite its intrinsic insulating nature. How the incorporated nanoparticles interact with the polymer matrix, as well as their size, shape, concentration, and type, may have a profound impact on the characteristics of polymer nanocomposites. Because of its cheap cost and outstanding physical and chemical characteristics, PMMA has garnered interest as a conducting polymer for usage in various applications as optical components, optical sensors, and optoelectronics devices^{12,13}. A linear polymeric substance derived from ethylene oxide, polyethylene glycol (PEG) has many applications in the biomedical and industrial fields. Its desirable properties, including high hydrophilicity, excellent biocompatibility with cells, low toxicity, and lack of immunogenicity, have made it one of the most studied polyethers. The inclusion of hydroxyl groups (OH) in PEG's structure allows it to dissolve in both hydrophilic and hydrophobic solutions. Since the hydroxyl group is thought of as an electron donor, it possesses the capability to create hydrogen bonds with organic molecules, allowing it to interact with them¹⁴⁻¹⁶. The non-toxicity, elevated dielectric permittivity, and ferroelectric properties of barium titanate (BaTiO₃) make it a useful material for a wide variety of applications, including but not limited to: sensors, electrical and microwave devices, ceramic capacitive interlayer structures, supercapacitors, and movable energy storage fields. New avenues for research into potential future applications have emerged from topical investigations in nanocomposites of BaTiO₃ and polymer^{17,18}. Introducing nanoparticles into bio to enhance their optical and dielectric characteristics, PMMA and PEG polymers are being worked on¹⁹⁻²¹. There are many theoretical studies on different nanostructures that have been investigated²²⁻²⁹. The properties of nanocomposites and composites materials were studied to use these materials in various fields³⁰⁻⁴³. With cheap cost, high energy storage, low factor of dispersion, and superior corrosion resistance compared to other nanocomposites, this study seeks to develop films of PMMA/PEG/BaTiO₃ for use in various electrical and nanoelectronic applications.

2. Materials and methods

The materials utilized in the present work are polymethyl methacrylate (PMMA) and polyethylene glycol (PEG) as raw materials for the polymeric blend, and barium titanate (BaTiO₃) nanoparticles as an additive. The PMMA/PEG polymeric blend and BaTiO₃ nanoparticles doped PMMA/PEG polymeric blend were manufactured by the casting technique. The pure PMMA/PEG blend film was obtained by dissolving 1 g of PMMA and PEG with 52:48 (w/w) ratio in chloroform (30 mL). 2.5 wt. %

and 5 wt. % of BaTiO₃ NPs were added to PMMA/PEG polymer blend solution. The PMMA/PEG/BaTiO₃ nanocomposite films were fabricated with a thickness of 100 μm. The BaTiO₃ NPs distribution in the PMMA/PEG medium was tested by optical microscope. The dielectric characteristics of PMMA/PEG/BaTiO₃ nanocomposite films were measured in the frequency range from 100 Hz to 5M Hz using a LCR meter type (HIOKI 3532-50 LCR HI TESTER). The dielectric constant (ϵ') was defined by Eq. (1)⁴⁴⁻⁴⁶

$$\epsilon' = C_p \cdot d / \epsilon_0 \cdot A, \quad (1)$$

where C_p is the capacitance and d is the sample thickness.

The dielectric loss (ϵ'') was calculated by Eq. (2)^{47,48}

$$\epsilon'' = \epsilon' \cdot D, \quad (2)$$

where D is the dispersion factor.

The A.C conductivity was calculated by Eq. (3)^{49,50}

$$\sigma_{AC} = \omega \cdot \epsilon'' \cdot \epsilon_0, \quad (3)$$

where ω is the angular frequency.

3. Results and discussion

The PMMA-PEG-BaTiO₃ nanocomposite (NC) was designed using Gauss View 5.0 software, initially each compound individually, as shown in Fig. 1, while Fig. 2 is specific to the nanocomposite under study. One of the main advantages of PMMA-PEG-BaTiO₃NC is its tunable dielectric properties. By adjusting the concentration of BaTiO₃ nanoparticles in the composite, the dielectric constant of the material can be tailored to suit specific applications, as it is a ceramic material that exhibits ferroelectric properties, making it useful in electronic devices and sensors. All of this makes PMMA-PEG-BaTiO₃NC ideal for use in electronic devices such as capacitors and sensors. Moreover, the addition of PEG improves the compatibility of the material with biological systems, making it suitable for use in biomedical applications. In addition to its mechanical flexibility, the presence of PEG in the composite imparts a degree of flexibility to the material, allowing it to be easily shaped into different shapes. Chemical bonding can be observed in PMMA-PEG-BaTiO₃NC interactions between PMMA and PEG polymers, in addition to the incorporation of BaTiO₃ molecules into the matrix, while physical bonding occurs between PMMA, PEG, and BaTiO₃ molecules (Figs. 1 and 2). So, we notice the high compatibility between PMMA and PEG due to their similar chemical structures; both are polymeric materials containing carbon, hydrogen, and oxygen atoms. The presence of oxygen in both polymers facilitates intermolecular interactions, such as hydrogen bonding and dipole interactions, which can promote good compatibility.

Hence, chemical compatibility between components is crucial for their successful integration through PMMA-PEG-BaTiO₃NC. This confirms that the polymer components (PMMA and PEG) provide a compatible matrix for the dispersion of BaTiO₃ nanoparticles, and it is also possible to work on modifying the surface of the BaTiO₃ particles to enhance their compatibility with the polymer matrix by introducing functional groups related to the polymer chains, which allows for better dispersion and adhesion between surfaces⁵¹.

Figs. 3 and 4, respectively, show the dependence of dielectric constant (ϵ') and dielectric loss (ϵ'') of PMMA/PEG/BaTiO₃ nanocomposite films on frequency. It could be noticed that the boost of BaTiO₃ NPs concentration in the PMMA/PEG polymer blend enhances the dielectric constant and dielectric loss with improved electrical properties due to the major contribution of polarization for interfacial type related to the accumulation of trapped charges at the interface.

The increase in the dielectric can be attributed to the condensed pinning effect of defects on the movement of the field walls associated with the deposition of spatial charges at the interfaces. Therefore, a smaller excitatory field was created mainly in the group of field walls, and the dependence of ϵ' on the excitatory field is weak. In addition, these data confirm that the interfaces of nanocomposites play an important role in the growth of the dielectric properties. Consequently, the improvement of the dielectric constant values largely leads to the improvement of the BaTiO₃ NPs particles dispersion in the PMMA/PEG polymer matrix. When BaTiO₃ NPs were added, heterogeneous differences in the ϵ' value were demonstrated, which are associated with changes in the interactions between the components of nanocomposites and complexation between them. In addition, it is illustrated that at low frequencies, the increased values of ϵ' can be associated with the interfacial type of polarization. The interfacial type of polarization occurs at low frequencies and decreases with increasing frequency. At elevated frequencies, the rotational motions of polar molecules in nanocomposites are not fast enough to reach equilibrium with the help of an electric field. As a result, ϵ' decreases with increasing frequency. Increased dielectric loss values were found at low frequencies, which may be due to the Maxwell – Wagner effect.

In addition, the decrease in ϵ' and ϵ'' with the increase in frequency is associated with the removal of the spatial charge polarization from the total polarization, when this type of polarization becomes an additional contribution at low frequencies⁵²⁻⁵⁸. The ϵ' and ϵ'' values of PMMA/PEG polymer blend increase with the increase in the BaTiO₃ NPs content, which is associated with an increase in the number of charge carriers⁵⁹⁻⁶⁴, as shown in Fig. 5. This figure demonstrates the BaTiO₃ distribution in

the PMMA/PEG polymer matrix. When BaTiO_3 NPs content increases, nanoparticles form a network of tracks inside the PMMA/PEG polymer matrix, hence, charge carriers can pass through the nanocomposite films⁶⁵⁻⁷⁰.

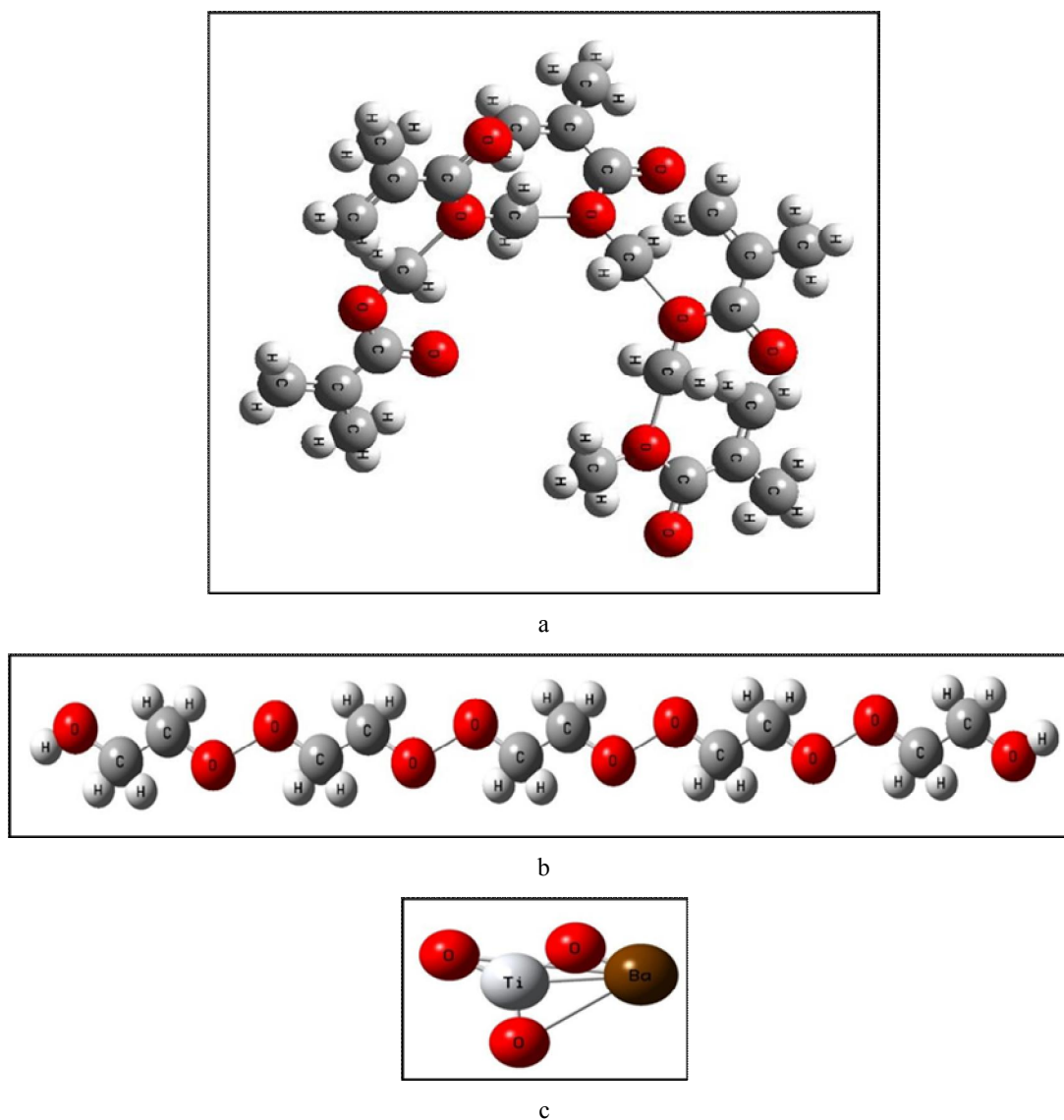


Fig. 1. Structure of poly-methyl methacrylate (PMMA) (a), polyethylene glycol (PEG) (b) and BaTiO_3 (c)

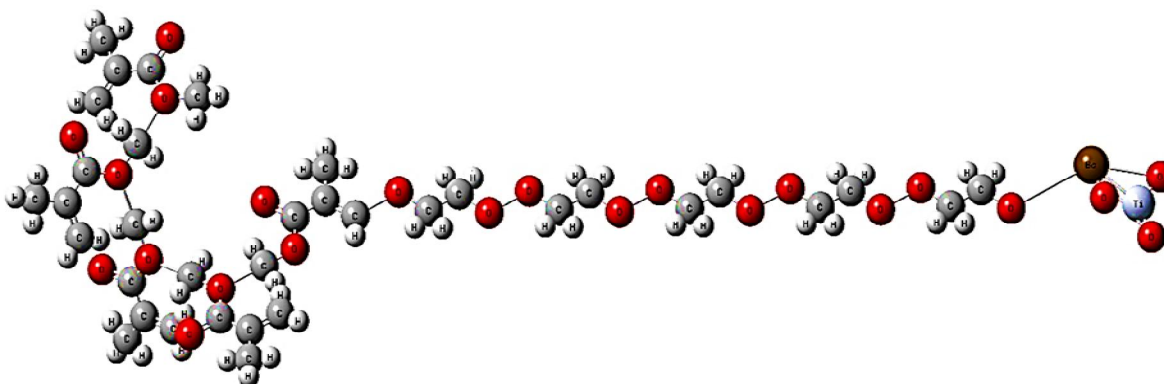


Fig. 2. Structure of PMMA-PEG- BaTiO_3 nanocomposites

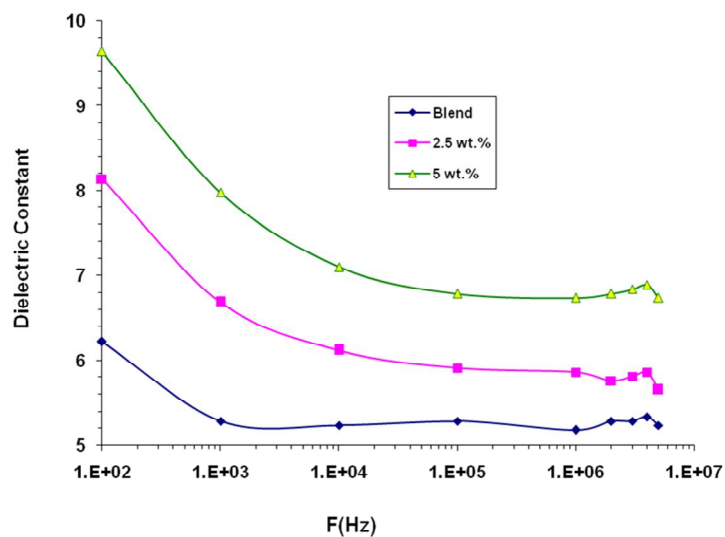


Fig. 3. Dependence of dielectric constant on frequency for PMMA/PEG/BaTiO₃ nanocomposites films

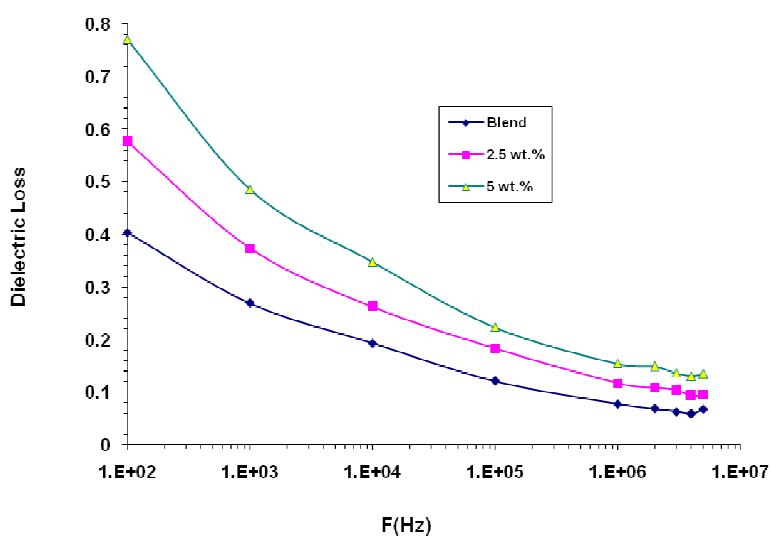


Fig. 4. Dependence of dielectric loss on frequency for PMMA/PEG/BaTiO₃ nanocomposites films

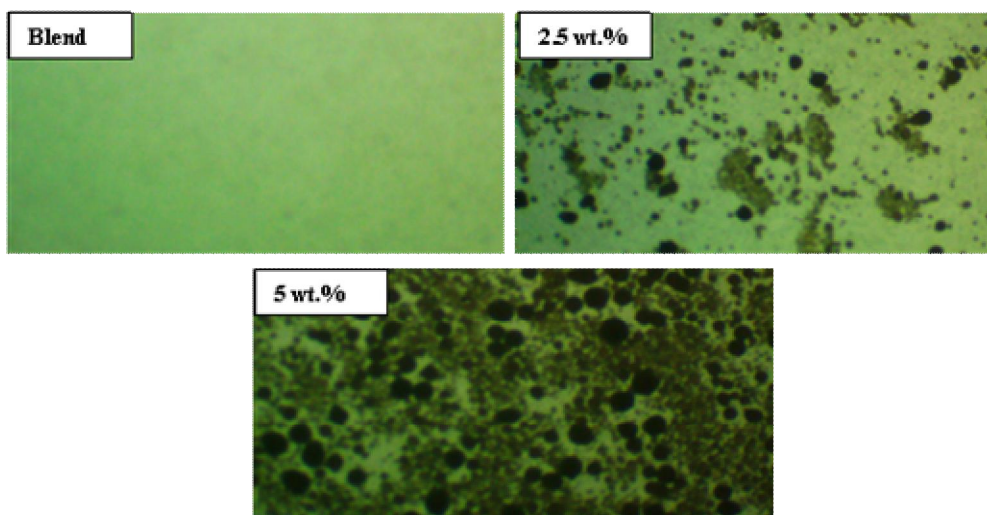


Fig. 5. Optical microscope images for PMMA/PEG/BaTiO₃ nanocomposite films

Fig. 6 illustrates the dependence of AC electrical conductivity for PMMA/PEG/BaTiO₃ nanocomposite films on frequency. The AC electrical conductivity boosts with increasing frequency and BaTiO₃ NPs content. Usually, the number of charge carriers, which have a high relaxation time due to the upper energy barrier and respond at low frequency, could be less in number; consequently, the conductivity is lower at low frequency. On the other hand, the number of charge carriers with short barrier heights is greater and they respond simply with elevated frequency and confirmed upper conductivity at elevated frequency. The increase in BaTiO₃ NPs content improves the number of electrons and leads to an increase in the conductivity^{71–74}.

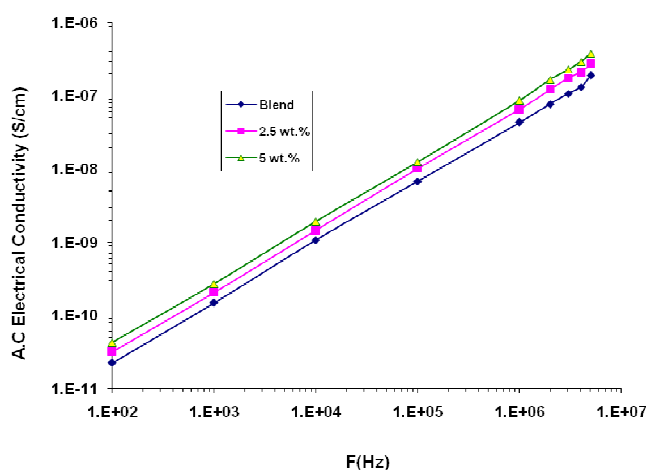


Fig. 6. Dependence of AC electrical conductivity on frequency for PMMA/PEG/BaTiO₃ nanocomposite films

4. Conclusions

In this work PMMA/PEG/BaTiO₃ nanocomposite films were prepared. The PMMA/PEG/BaTiO₃ nanocomposite films were obtained by the casting method. The morphological and dielectric characteristics of PMMA/PEG/BaTiO₃ nanocomposite films were investigated. The results of the dielectric characteristics confirmed the increase in the dielectric parameters of PMMA/PEG blend with an increase in BaTiO₃ NPs content. The dielectric constant of PMMA/PEG blend increased by about 35.4 %, while the electrical conductivity increased by about 47.6 % at 100 Hz when the SrTiO₃ content reached 5 wt. %. The dielectric properties of PMMA/PEG/BaTiO₃ nanocomposite films distorted with increasing frequency. Finally, the dielectric characteristics of PMMA/PEG/BaTiO₃ nanocomposite films can be used in various electrical and nanoelectronic applications with high energy storage, lightweight, low cost, and flexibility.

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СИНТЕЗ ТА ОПТИМІЗАЦІЯ ДІЕЛЕКТРИЧНИХ ВЛАСТИВОСТЕЙ ГІБРИДНИХ НАНОСТРУКТУР ПММА/ПЕГ/ТИТАНАТ БАРІЮ ДЛЯ НАКОПИЧЕННЯ ЕНЕРГІЇ Й ЕЛЕКТРОННИХ ЗАСТОСУВАНЬ

Анотація. Дослідження спрямоване на покращення діелектричних властивостей суміші поліметилметакрилату (ПММА) та поліетиленгліколю (ПЕГ) з додаванням наночастинок титанату барію (BaTiO₃) для використання в електроніці та діелектричних галузях. Нанокмпозитні плівки ПММА/ПЕГ/BaTiO₃ виготовлено методом лиття. Перевірено морфологічні та діелектричні характеристики нанокмпозитних плівок ПММА/ПЕГ/BaTiO₃. Діелектричні характеристики досліджено за допомогою LCR-метра за частот від 100 Гц до 5 МГц. Результати визначення діелектричних характеристик підтвердили зростання діелектричних параметрів суміші ПММА/ПЕГ зі збільшенням вмісту наночастинок BaTiO₃. Діелектричні властивості нанокмпозитних плівок ПММА/ПЕГ/BaTiO₃ змінювалися з підвищенням частоти. Нарешті, діелектричні характеристики показують, що нанокмпозитні плівки ПММА/ПЕГ/BaTiO₃ придатні для різних електричних та наноелектронних застосувань з високим накопиченням енергії, малою вагою, низькою вартістю та гнучкістю.

Ключові слова: BaTiO₃, діелектричні властивості, ПММА/ПЕГ, наноелектроніка, суміш.