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FABRICATION AND TAILORING THE MORPHOLOGICAL AND ELECTRICAL FEATURES OF SILICON NITRIDE-SILICON DIOXIDE/POLYMETHYL METHACRYLATE HYBRID NANOCOMPOSITES FOR NANOELECTRONIC FIELDS

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Abstract. The goal of this research is to create PMMA and SiO_2-Si_3N_4 nanoparticles doped PMMA films with enhanced structural and electrical properties to employ in various quantum electronics fields. The casting process was used to create the (PMMA-SiO_2-Si_3N_4) nanocomposite films. In the development of nanocomposite materials, the hybrid nanocomposite films with 2.3%, 4.6% and 6.9% contents of nanoparticles were prepared. Using an optical microscope (OM), the morphology of the nanocomposites was examined. At room temperature, the electrical characteristics of (PMMA-SiO_2-Si_3N_4) nanocomposites were investigated. The results revealed that the dielectric constant and dielectric loss of (PMMA-SiO_2-Si_3N_4) nanocomposites reduced as the frequency of the applied electric field increased. The electrical conductivity of alternating current rises with rising frequency. With increasing concentrations of SiO_2-Si_3N_4 nanoparticles, the dielectric constant, dielectric loss, and AC electrical conductivity of (PMMA-SiO_2-Si_3N_4) nanocomposites were enhanced. When the SiO_2-Si_3N_4 NPs content reached 6.9% at 100Hz, the dielectric constant increased from 3.86 to 4.76 while the dielectric loss increased from 0.19 to 0.29. Finally, the obtained results demonstrated that the (PMMA-SiO_2-Si_3N_4) nanocompo sites have elevated values of dielectric constant compared with dielectric loss, which makes them suitable for a variety of quantum electronics applications.

Keywords: PMMA; conductivity; Si_3N_4; nanocomposites; SiO_2; quantum electronics.

1. Introduction

The concept of nanocomposite material has grown greatly over the years to include a wide range of systems, including one-dimensional, two-dimensional, three-dimensional, and amorphous materials comprised of clearly different components and blended at the nanoscale. Nanocomposites may be described as "materials with a nanoscale structure that improve the macroscopic properties of products." Clay, carbon, or polymer nanocomposites, or a mixture of these materials with nanoparticle building blocks, are examples of nanocomposites.[1] Conductive polymer composites are lightweight materials that can combine the properties of electrically conductive metals with polymer capabilities. These composites are employed in a variety of applications such as acoustic emission sensors, angular acceleration accelerometers, integrated decoupling capacitors, electronic packaging, electromagnetic frequency interference shields, and antistatic devices.[2] PMMA has various fields in numerous productive and technological applications. PMMA has the advantage of a distinguished combination of good optical characteristics (transparency in a broad series from the near UV to the near IR). Additionally, PMMA is extremely appropriate for several imaging & non-imaging microelectronic fields like photo-resistance for direct write e-beams, process of X-ray & deep-UV micro-lithographic processes. It is used to create a variety of optical devices like optical lenses. PMMA has good thermal stability, electrical characteristics, safety, resistance to

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weather, molding ability, and simple shaping.[3] PMMA offers several advantages for applications, including its ability to withstand high temperatures, simplicity of laboratory manipulation, lightweight, low cost, esthetic appeal, color, and absence of toxicity.[4] Because of its great features such as low density, high strength, strong thermal shock resistance, and good resistance to oxidation and corrosion, silicon nitride (Si_3N_4) ceramics are regarded as an essential contender. However, the inherent brittleness of Si_3N_4 precludes its extensive industrial utilization. Much work has been expended in recent decades to enhance the mechanical characteristics of Si_3N_4 ceramic materials, particularly fracture toughness.[5] SiO_2 (silicon dioxide), often known as silica, is a metalloid oxide that belongs to the fourteenth group of the periodic table. It is a silicic acid polymer composed of linked tetrahedral SiO_4 units. Natural and synthesized SiO_2 can be found as crystals (quartz, cristobalite, and tridymite) and amorphous forms. These ceramic materials exhibit unique features in their bulk form, such as abrasion, optical ultraviolet (UV) filtering, luminescence, and biocompatibility.[6] SiO_2 nanoparticles are commonly used in polymers to increase heat resistance, radiation resistance, optical and electrical characteristics.[7-17] The metal nitride or oxide was added to polymers to improve their electronic, dielectric, and optical properties.[18-25] The PMMA-nanocomposites were studied for different fields.[26-31] The present work aims to create Si_3N_4/SiO_2/PMMA nanostructure films with better electrical characteristics than other types of nanocomposites, making them promising nanocomposites for use in various electronic nanodevices.

2. Materials and Methods

The silicon nitride (Si_3N_4) nanopowder and silicon dioxide(SiO_2) nanopowder were obtained from US Research Nanomaterials, Inc. (Si_3N_4, 15-30 nm, purity 99.9%) and (SiO_2, 98+%, 60-70nm, amorphous). The (PMMA-SiO_2-Si_3N_4) nanocomposites were fabricated by dissolving 1 g of PMMA in 30 ml of chloroform, and mixed at room temperature for 30 minutes using a magnetic stirrer to get the PMMA solution. Then, SiO_2-Si_3N_4 nanoparticles were added to the PMMA solution to achieve the content of nanoparticles of 2.3%, 4.6%, 6.9% at SiO_2:Si_3N_4 ratio of 50:50 (w/w). The PMMA-SiO_2-Si_3N_4 NCs were cast on a glass Petri dish at room temperature. The structural properties of (PMMA-SiO_2-Si_3N_4) nanocomposites were studied using an optical microscope (OM). The dielectric characteristics were measured using an LCR meter (HIOKI 3532-50 LCR Hi TESTER) within the frequency range of 100Hz-5MHz. Permittivity (ε') is determined by Eq. (1):[32, 33]

$$\mathcal{E}' = C_p\, d/\varepsilon_o A \qquad (1)$$

where C_p is the capacitance and d is the film thickness. The dielectric constant is determined according to Eq. (2):[34]

$$\mathcal{E}' = C_P / C_o \qquad (2)$$

Where C_p means parallel capacitance and Co signifies vacuum capacitance. To compute dielectric loss by using the relation:[35]

$$\mathcal{E}'' = \mathcal{E}' D \qquad (3)$$

where D is the dispersion factor, the AC conductivity was calculated as:[36]

$$\sigma_{A.C} = \omega \mathcal{E}_o \mathcal{E}'' \qquad (4)$$

where ω is an angular frequency ($\omega = 2\pi f$).

3. Results and Discussion

Fig. 1 shows optical microscope images of PMMA and PMMA-SiO_2-Si_3N_4 nanocomposites with different SiO_2-Si_3N_4 concentrations at a magnification of 10. This figure reveals the distribution and propagation of SiO_2-Si_3N_4 nanoparticles inside the PMMA medium with different contents of SiO_2-Si_3N_4 NPs, which absorbed and scattered the incident light photons. The surface energy components of the polymer and the nanoparticles differ, which may lead to nanoparticle agglomeration. At low content, the NPs are propagated as clusters.[37-40] When the content of SiO_2-Si_3N_4 nanoparticles increases, the nanoparticles form a network of paths within the polymer matrix.[41-45]

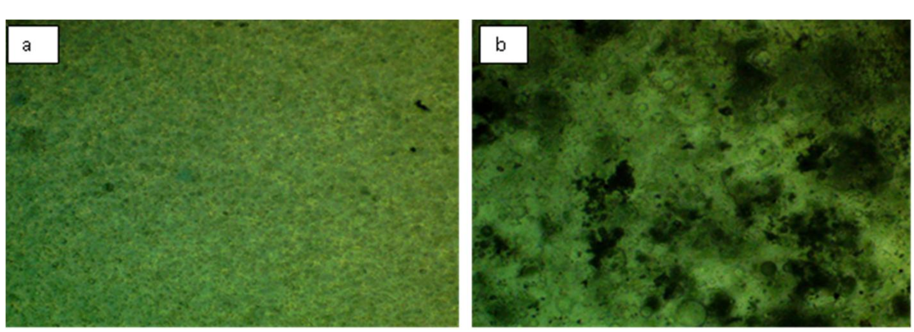

Fig 1. Optical microscopic images of PMMA-SiO_2-Si_3N_4 nanostructures at a magnification of 10x: a. Pure PMMA, b. 2.3 wt.% SiO_2-Si_3N_4 NPs,

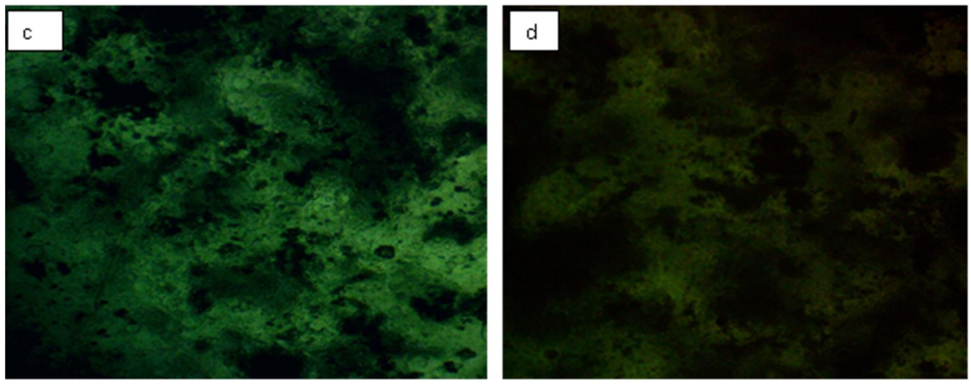

Fig. 1. Continuatio

Figs. 2 and 3 show the dependence of the dielectric constant and dielectric loss of PMMA-SiO_2-Si_3N_4 nanocomposites on frequency. The results demonstrate that the dielectric constant and dielectric loss are larger in the lower frequency range and decrease with increasing frequency for all samples, which is due to Maxwell-Wagner polarization, which generates higher dielectric constants and dielectric losses. Insulator-conductor contacts create this kind of polarization. This interfacial polarization is induced by the build-up of space charges or dipoles at the interfaces. In the lower frequency range, space charges have plenty of time to react to the applied electric field, whereas in the higher frequency range, changes in the electric field are too quick for the space charges to react, and the polarization effect does not exist, so increasing the frequency. This leads to a decrease in the dielectric constant and dielectric loss. The dielectric constant and dielectric loss increased with increasing content of SiO_2-Si_3N_4 NPs, which is related to the rise in the number of charge carriers.[47-59] At 100 Hz, when the SiO_2-Si_3N_4 NPs content reached 6.9 %, the content of the dielectric constant increased from 3.86 to 4.76, while the dielectric loss increased from 0.19 to 0.29. These values, ε' and ε'', make the (PMMA-SiO_2-Si_3N_4) nanocomposites important and potential nanomaterials for many electronic applications.

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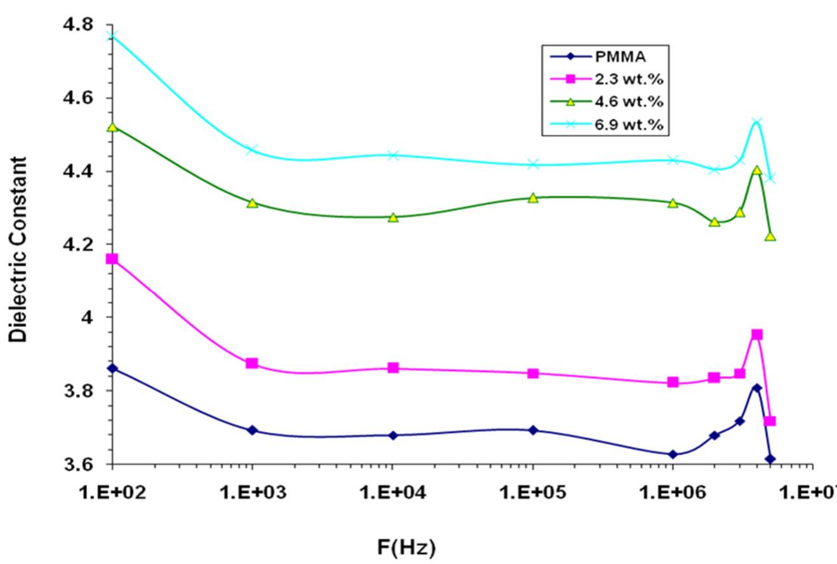

Fig. 2. Dielectric constant for nanocomposites vs. frequency

The increase in permittivity might be produced by defects influence on the motion of the field wall related to the space charges at the interfaces. Consequently, a lesser excitation field was capable of producing typically in the movement of field walls, and the ε' reliance on an exciting field is weak. Additionally, these consequences confirmed that the interfaces of nanocomposites play a significant role in the increase of the dielectric properties. As well, the interfacial type of polarization provides significant corroboration of the homogeneous distribution of SiO_2-Si_3N_4 nanostructures in the PMMA medium. Consequently, the enhancement in the ε' values is strongly caused by the increase in dispersion of SiO_2-Si_3N_4 nanostructures inside the PMMA matrix. At the low frequencies, the ε' has high values, which may be due to the interfacial type of polarization. The interfacial polarization is created at the lower frequency and decreases with the rise of the applied field frequency. At higher frequencies, the movement of polar molecular rotation of PMMA/SiO_2-Si_3N_4 nano-structures is not fast enough to obtain equilibrium with an electric field. Thus, with rising of the frequency, the ε' decreased. The higher ε'' values were demonstrated at lower frequency which may be due to the effect of Maxwell–Wagner. Furthermore, the reduce of ε' and ε'' values with an increase in the frequency related to the eliminate of space charge type of polarization from total polarization wherever this polarization type was the extra contributing at the lower frequencies.[60,61]

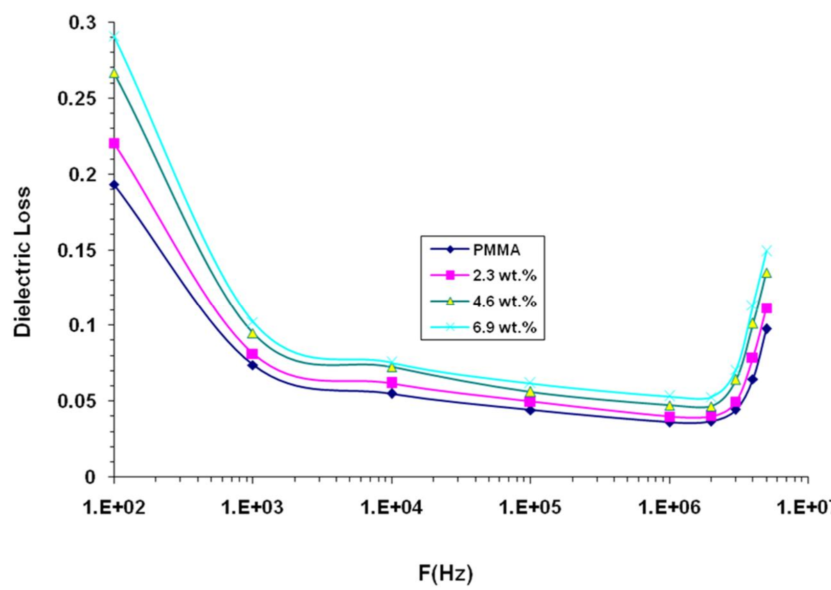

Fig 3. Dielectric loss for nanocomposites vs. frequency

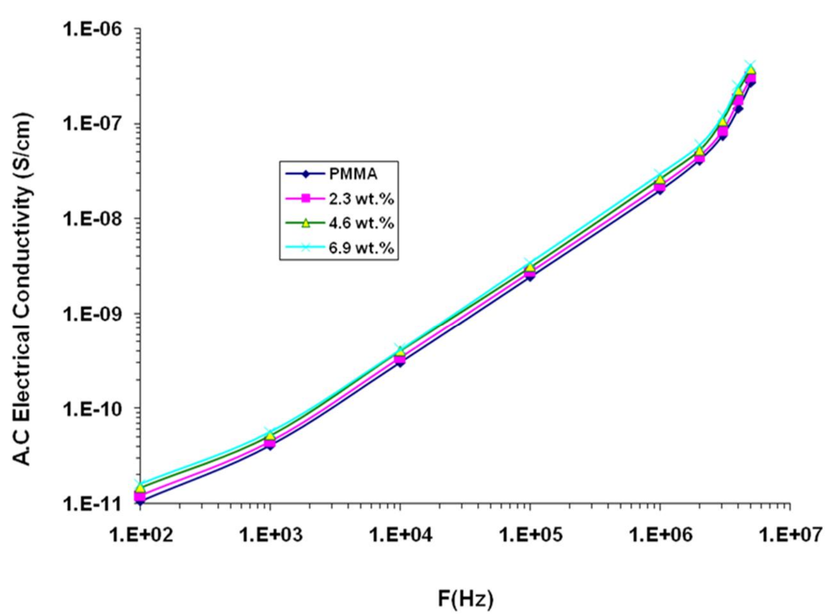

Fig 4. AC conductivity of (PMMA-SiO_2-Si_3N_4) nanocomposites vs. frequency

Fig. 4 depicts the fluctuation of AC electrical conductivity with electric field frequency for PMMA-SiO_2-Si_3N_4 nanocomposites at room temperature. As seen from the image, the AC electrical conductivity rises with frequency due to space charge polarization. This is caused by both charge carrier stimulation of higher states in the conduction band and low-frequency space charge polarization. Electronic polarization and hopping charge carriers cause an increase in conductivity at high frequencies. The primary chain motion and ion motions are the two factors that influence AC conductivity. The influence of SiO_2 and Si_3N_4 NPS concentration on the electrical conductivity of a PMMA) mix at 100 Hz. When the frequency increased from 100Hz to 2MHz, the electrical conductivity of PMMA increased from 1.07×10^{-11} to 4.09×10^{-8} S/cm and from 1.62×10^{-11} to 5.86×10^{-8} S/cm for 6.9% SiO_2-Si_3N_4 NPs. Since the charge carrier density in the polymer medium rises with the concentration of nanoparticles, the AC electrical conductivity increases.[62-70]

4. Conclusions

The current work comprises the casting technique for producing PMMA films and SiO_2-Si_3N_4 NPs doped PMMA. The PMMA-SiO_2-Si_3N_4 nanostructures' structural and dielectric characteristics were examined for use in different nanoelectronic techniques. The structural characteristics of PMMA-SiO_2-Si_3N_4 nanostructures revealed a great dispersion of SiO_2 and Si_3N_4 NPs in the PMMA medium, as well as effective integration of Si_3N_4-SiO_2 NPs into the PMMA matrix. The results showed that dielectric constant and dielectric loss of PMMA-SiO_2-Si_3N_4 nanocomposites decreased, while the conductivity increased, as the frequency of the applied electric field increased. The dielectric constant, dielectric loss, and AC electrical conductivity of PMMA-SiO_2-Si_3N_4 nanocomposites rise with increasing concentrations of SiO_2-Si_3N_4 nanoparticles, which can be advantageous for various electrochemical applications. The dielectric constant increased from 3.86 to 4.76 while the dielectric loss increased from 0.19 to 0.29 when the SiO_2-Si_3N_4 NPs content reached 0f 6.9% at 100Hz. When the frequency increased from 100Hz to 2MHz, the electrical conductivity of PMMA rose from 1.07×10^{-11} to 4.09×10^{-8} S/cm and from 1.62×10^{-11} to 5.86×10^{-8} S/cm for 6.9% SiO_2-Si_3N_4 NPs. These results make the PMMA-SiO_2-Si_3N_4 nanostructures welcome in a variety of quantum electronics applications.

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ВИГОТОВЛЕННЯ Й АДАПТАЦІЯ МОРФОЛОГІЧНИХ ТА ЕЛЕКТРИЧНИХ ХАРАКТЕРИСТИК ГІБРИДНИХ НАНОКОМПОЗИТІВ НІТРИДУ КРЕМНІЮ-ДІОКСИДУ КРЕМНІЮ/ПОЛІМЕТИЛМЕТАКРИЛАТУ ДЛЯ ГАЛУЗЕЙ НАНОЕЛЕКТРОНІКИ

Анотація. *Метою цього дослідження є створення плівок з ПММА та легованих SiO_2-Si_3N_4-наночастинками ПММА з покращеними структурними й електричними властивостями для використання в різних галузях квантової електроніки.*

Процес лиття був використаний для створення плівок нанокомпозитів (ПММА-SiO_2-Si_3N_4). У рамках розробки нанокомпозитних матеріалів були отримані гібридні нанокомпозитні плівки з вмістом наночастинок 2,3%, 4,6% та 6,9%. За допомогою оптичного мікроскопа було досліджено морфологію нанокомпозитів. За кімнатної температури було досліджено електричні характеристики нанокомпозитів (ПММА-SiO_2-Si_3N_4). Результати показали, що діелектрична проникність і діелектричні втрати нанокомпозитів (ПММА-SiO_2-Si_3N_4) зменшуються зі збільшенням частоти прикладеного електричного поля . Електропровідність змінного струму зростає зі збільшенням частоти. Зі збільшенням концентрації наночастинок SiO_2-Si_3N_4 діелектрична проникність, діелектричні втрати й електропровідність змінного струму нанокомпозитів (ПММА-SiO_2-Si_3N_4) покращувалися. Діелектрична проникність збільшувалася з 3,86 до 4,76, тоді як діелектричні втрати збільшувалися з 0,19 до 0,29, коли вміст наночастинок SiO_2-Si_3N_4 досяг 6,9% за 100 Гц. Отже, отримані результати показали, що нанокомпозити (ПММА-SiO_2-Si_3N_4) мають підвищені значення діелектричної проникності порівняно з діелектричними втратами, що робить їх придатними для використання в різних сферах квантової електроніки.

Ключові слова: *ПММА, провідність, Si_3N_4, нанокомпозити, SiO_2, квантова електроніка.*