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EFFECT OF SI_3N_4/TaC NANOMATERIALS ON THE STRUCTURAL AND ELECTRICAL CHARACTERISTICS OF POLY METHYL METHACRYLATE FOR ELECTRICAL AND ELECTRONICS APPLICATIONS[†]

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This study used the casting method to prepared PMMA/Si_3N_4/TaC nanocomposites with variant content (0,2,4,6,8) % wt of Si_3N_4/TaC nanoparticles. The structural and electrical properties have been investigated. Scanning electron microscope (SEM) indicates the homogenous, smooth and dispersed of Si_3N_4 and TaC NPs inside the PMMA matrix due to strong covalent interaction between the Si_3N_4 and TaC NPs in the PMMA matrix, which means a good method for prepared films. Optical microscope images explained that increasing nanoparticle content forms network paths inside the polymeric matrix that act as charge carriers. FTIR spectra indicate a physical interference between the polymer matrix and nanoparticles. The AC electrical properties of nanocomposites obtained that the dielectric constant and dielectric loss rise with rising content of nanoparticles and decrease with increasing frequency of applied electric field. While the A.C. electrical conductivity rises with the rising frequency and weight content of Si_3N_4/TaC nanoparticles. These results indicated that the PMMA/Si_3N_4/TaC nanostructures could be considered promising materials for electronics and electrical nanodevices.
Keywords: *Nanocomposites; PMMA; Si_3N_4; TaC; AC electrical properties*
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1. INTRODUCTION

Polymers hold the potential to revolutionize industrial thought by replacing the majority of materials with plastic ones and enabling a wide range of applications. Additionally, it entered all sectors of the economy, including the medical sector, and a variety of optical, electronic, and applications, including photovoltaic cells and optoelectronics [1]. Last year, there was a lot of interest in innovative materials called nanocomposites polymers, composed of organic and inorganic polymers and nanoparticles in a nanoscale region. These composite materials differ from pure polymers regarding their chemical and physical characteristics. Impurity polymers' effects on the properties of polymers provide them with more significant advantages and enable them to improve desirable features. In many different types of applications, that might be highly significant and helpful [2,3].

A good thermoplastic polymer known as poly (methyl methacrylate, or PMMA) has papers describing how it can be used as a gate insulator in organic transistors with thin films (OTFTs). PMMA is a great choice as a dielectric layer in organic electronics due to its excellent mechanical and thermal durability, excellent electrical resistivity (>21015.cm), appropriate dielectric characteristics, and thin film processability on vast regions by spin coating. We explore the optical, electrical, and microgravimetric properties of PMMA thin sheets to assess their chemical sensing potential. This tactility-dependent thermoplastic material is made of volatile organic compounds with high stiffness, transparency, outstanding insulating qualities, excellent planarity, and thermal stability [4,5].

Polymers' chemical and physical composition and structure influence the size of their dielectric. The unique characteristics of the molecular mobility of polymers, and consequently their chemical and physical structure, affect the parameters that describe the dielectric loss and losing tangential. Due to their low conductivity, most polymers are insulators in general. The inclusion of the proper dopants as well as thermally and electrically produced carriers affect the conductivity [6,7]. Due to its excellent oxidation resistance and strong thermal conductivity, silicon nitride (Si_3N_4) is a possible alternative to silica supports that are more frequently utilized in reactions where efficient heat transmission is required. Because it still contains NH_2 and NH groups, amorphous silicon nitride made from sol-gel is especially intriguing for solid base catalysis [8,9]. Silicon nitride is a good contender for use in material science, the microelectronics industry, solar technology, and other fields due to its physical features. Due to the silicon nitride film's exceptionally high transparent in the spectral range of 300-1200 nm, it is employed in solar cell technology as an antireflective coating and to passivate silicon surfaces [10,11].

Tantalum Carbide (TaC), which is also a very promising ultra-high temperature material, has been found to be wear-resistant as well as to have biocompatible qualities that make it suitable for biomedical applications [12,13]. TaC has drawn investigation, but thorough comprehension has eluded researchers because, like other transition-metal carbides, it appears in a broad variety of compositions, and as a result, its physical characteristics vary. Additionally, impurities can affect characteristics, and well-characterized TaC materials, particularly single crystals, are typically hard to come by. TaC's lattice dynamics and band structure, as well as its mechanical, thermal, and electrical characteristics, have all been studied [14,15]. This paper aims to preparation of the PMMA/Si_3N_4/TaC nanocomposite and study the structural and AC electrical properties.

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2. MATERIALS AND METHOD

The casting method was used to create films of poly methyl methacrylate (PMMA) doped with silicon nitride nanoparticles (Si_3N_4 NPs) and tantalum carbide nanoparticles (TaC NPs). PMMA pure film was created by dissolving 1.5 gm of this polymer in 30 ml of chloroform at room temperature with magnetic stirrer for half an hour. The nanocomposites films were created by adding Si_3N_4 and TaC NPs to a PMMA solution with concentrations of (2, 4, 6, and 8) % wt. The structural characteristics of ($PS/SiC/Sb_2O_3$) nanocomposites examined by the scanning electron microscopic (SEM) using a Hitachi SU6600 variable, Optical microscope (OM) provided by Olympus (Top View, type Nikon-73346) and the Fourier Transformation Infrared Spectroscopy (FTIR) (Bruker company type vertex-70, German origin) with variety wavenumber (500-4000) cm^{-1}. The dielectric characteristics were studied at range (f=100 Hz to 5×10^6 Hz) by LCR meter (HIOKI 3532-50 LCR HI TESTER).
The dielectric constant (έ) is given by [16,17]

$$\acute{\varepsilon} = \frac{C_p}{C_o}, \tag{1}$$

where, Cp is capacitance and C_o is a vacuum capacitance
Dielectric loss (ε'') is calculated by [18,19]:

$$\varepsilon'' = \acute{\varepsilon} D. \tag{2}$$

Where, D: is displacement
The A.C. electrical conductivity is determined by [20,21]

$$\sigma_{AC} = \omega \acute{\varepsilon}\, \varepsilon_o, \tag{3}$$

where w angular frequency

3. RESULTS AND DISCUSSION

The scanning electron microscope (SEM) is used to study the morphological of PMMA/Si_3N_4/TaC nanocomposites. The SEM images of pure PMMA and PMMA/Si_3N_4/TaC nanocomposites are revealed in Fig. (1) with various concentration 0, 2, 4, 6 and 8 wt.% of Si_3N_4 and TaC NPs with a magnification 50 KX and scale 200 nm. From this figure in image A, it is observed that the pure PMMA was homogenous and smooth this indicates a good method for prepared films. Also, it is observed that in images (B, C, D and E) the homogeneous dispersed of Si_3N_4 and TaC NPs inside the PMMA matrix respectively primarily due to strong covalent interaction between the Si_3N_4 and TaC NPs in the PMMA matrix [22,23].

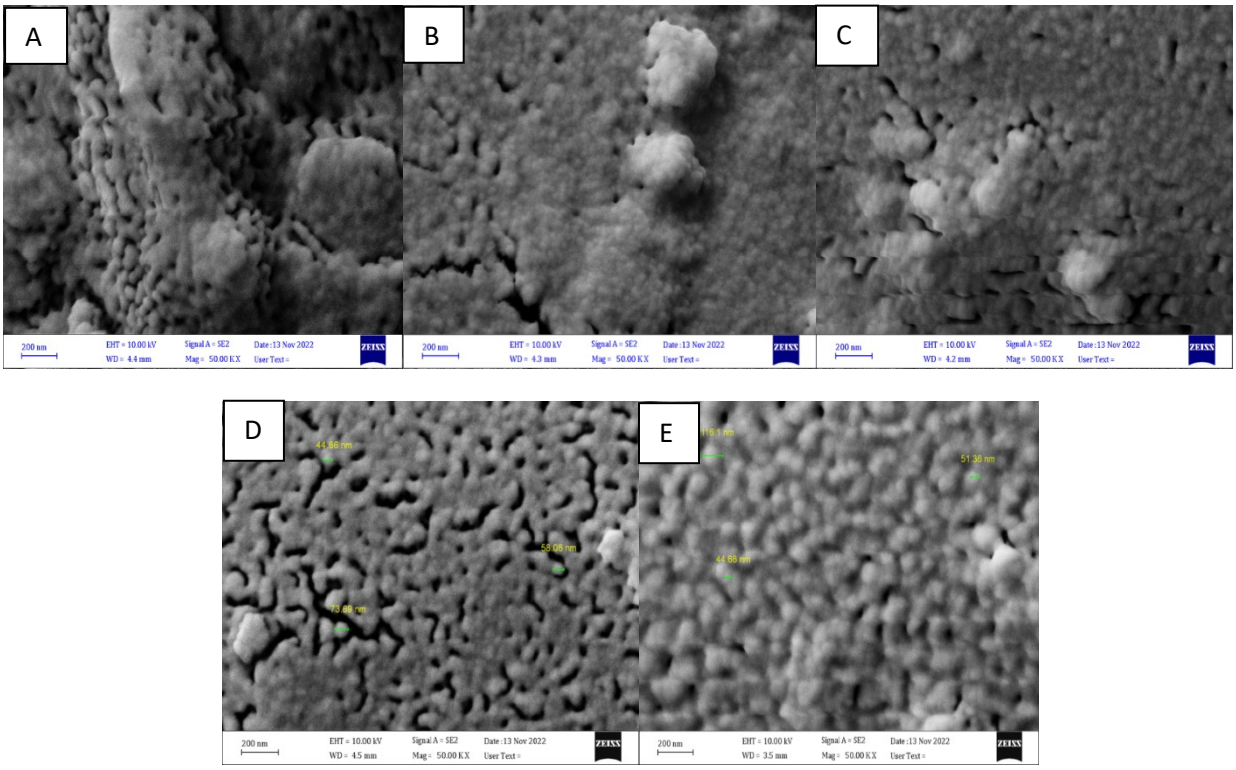

Figure 1. SEM images of (PMMA/Si_3N_4/TaC) nanocomposites, (A) for (PMMA), (B) 2 wt.% Si_3N_4 / TaC NPs, (C) 4 wt.% Si_3N_4 / TaC NPs, (D) 6 wt.% Si_3N_4 / TaC NPs, (E) 8 wt.% Si_3N_4 / TaC NPs

The optical microscope gives the change of surface morphology of PMMA/Si_3N_4/TaC nanocomposites. Figure (2) displays the PMMA/Si_3N_4/TaC nanocomposites optical microscope (OM) at magnification power (10x) for all specimens. The polymer blend film surface image(A) shows an uniform phase without phase separation; in the other hand, it has a finer morphology and smooth surface, demonstrating at this successful polymer ratio of PMMA. While image (B-E), it can be seen, that Si_3N_4/TaC NPs are well dispersed on the surface of the PMMA polymer films and this apparent more evident with the increase in the wt.% of Si_3N_4/TaC. The nanocomposite shows nearly elliptical structure of particles of uniform shape. This is because the NPs have a large surface area while the polymeric solution containing different polar groups has a high affinity for Si_3N_4/TaC which leads to the orientation of the nanoparticles within the polymer chain and thus the Si_3N_4/TaC structure becomes more compact and thus the consistency of the material increases. This provided a suitable preparation method for preparing nanocomposite films [24-26].

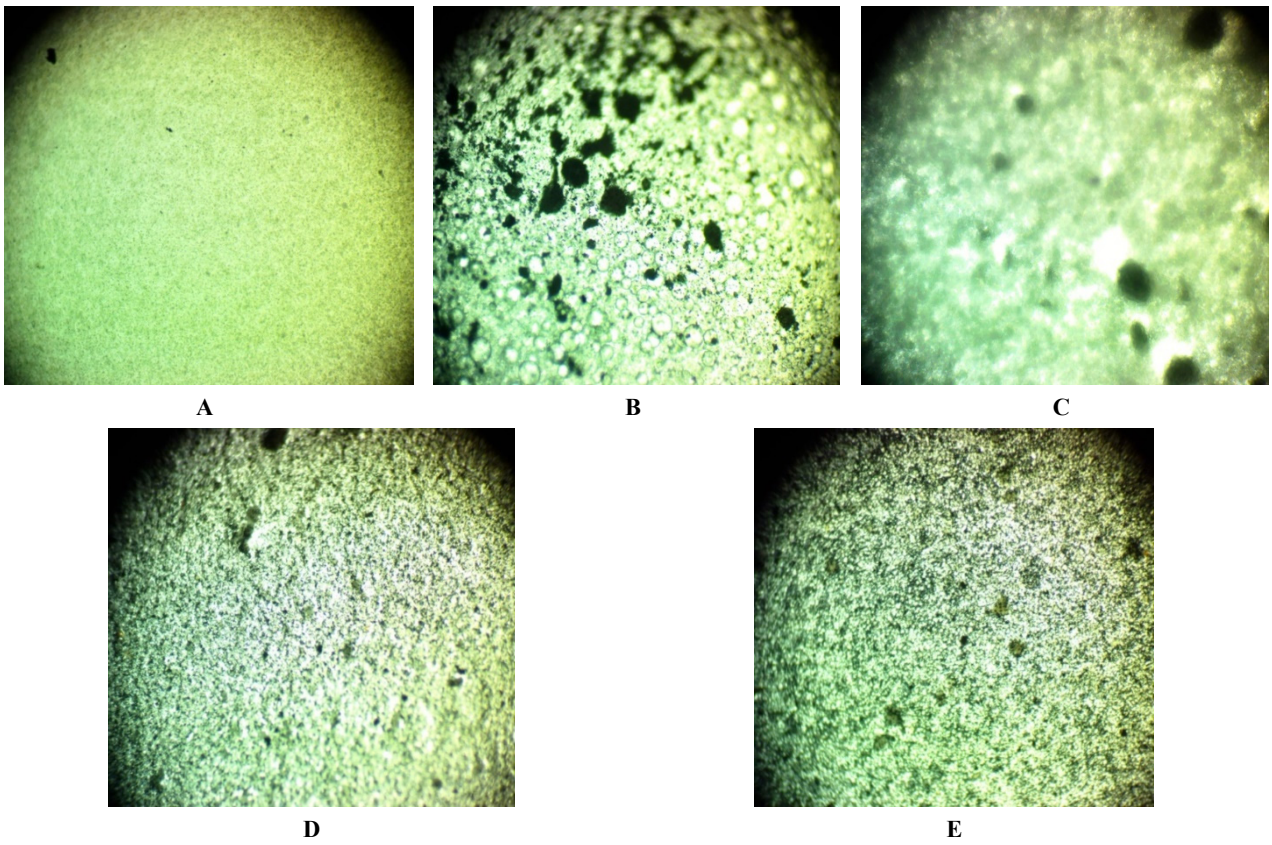

Figure 2. OM images (10×) for (PMMA/Si_3N_4/TaC) nanocomposites A. pure polymer, B. 2 wt.% of Si_3N_4/TaC NPs, C. 4 wt.% of Si_3N_4/TaC NPs, D. 6 wt.% of Si_3N_4/TaC NPs and E. 8 wt.% of Si_3N_4/TaC NPs

FTIR spectra provide information on the vibration and rotation of molecular groups in a material. Figure (3) displays the FTIR spectra of (PMMA/Si_3N_4/TaC) nanocomposites in the range wave number (500–4000) cm^{-1}. In the image (A), FTIR spectra of (PMMA) polymer reveal an absorption band at 2950.45 cm^{-1} corresponding to the CH_3 bending vibration and the band 1723.22 cm-1 attributed to the C=O stretching vibration. CH_3 stretching vibration indicate to the band 1434.60 cm^{-1}. The absorption band at 1142.95 attribute to the symmetric stretching vibration of C-O. The bands 985.57 cm^{-1}, 698.13 cm^{-1} and 750.29 cm^{-1} corresponding to the C−C bending and stretching vibration respectively. The spectra of PMMA with variant concentration of Si_3N_4 and TaC NPs in images B, C, D and E respectively. In image B where the additive 2 wt.% Si_3N_4 and TaC NPs caused change shift in some bands and intensities at low wavenumber (1434.63, 1142.83) cm^{-1} and high wave number at bands (1723.30, 995.01, 750.01) cm^{-1}but bands 2950.45 cm^{-1} and 698.13 cm^{1} there is not affected on this band while, the image C which additive concentration of 4 wt.% from Si_3N_4 and TaC NPs, affected change shift in some bands and intensities at low wavenumber (995.34, 750.45) cm^{-1} and high wave number at bands (1723.45, 1434, 1143) cm^{-1}but bands 2950.45 cm^{-1} and 698.13 cm^{1} there is not affected on this band. the image D which additive concentration of 6 wt.% from Si_3N_4 and TaC NPs, shifted and changed several bands' intensity at low wavenumber (995.42, 750.02) cm^{-1} and high wave number at bands (1723.54, 1434.64, 1143) cm^{-1}but bands 2950.45 cm^{-1} and 698.13 cm^{1} has not been impacted and added concentration of 8 wt.% from Si_3N_4 and TaC NPs in image E, caused shifts in certain bands and intensities at low wavenumber (994.90, 749.93) cm^{-1} and high wave number at bands (1724.04, 1434.68, 1143) cm^{-1}but bands 2950.45 cm^{-1} and 698.13 cm^{1} has not been impacted. The FTIR studies show that adding different concentration of Si_3N_4/TaC in images B, C,D and E leads to the displacement of some of the bonds and not emergence of new peaks therefore, there is no chemical interaction between Si_3N_4/TaC nanoparticle and the PMMA polymer matrix [27-29].

Equation (1) was used to calculate the dielectric constant (ε') of (PMMA/Si_3N_4/TaC) nanocomposites. Figure (4) explain the dielectric constant of (PMMA/Si_3N_4/TaC) nanocomposites with frequency. It is note that dielectric constant decrease with rising of frequency for all the sample prepare which, as a result of the capabilities of dipoles in nano-composites samples to transform in the direction of the applying electric current and the reduction of space charge polarization [30-32]. Figure (5) explain the dielectric constant of (PMMA/Si_3N_4/TaC) nanocomposite with the content of nanoparticle at 100 Hz. It is observed that dielectric constant rise with rising of concentration nanoparticle for all illustrations of nano-composites. Interfacial polarization in the nanocomposites' internal alternating electric field and an increase in the charge carriers are responsible for these processes of (PMMA/Si_3N_4/TaC) nanocomposite [33-35].

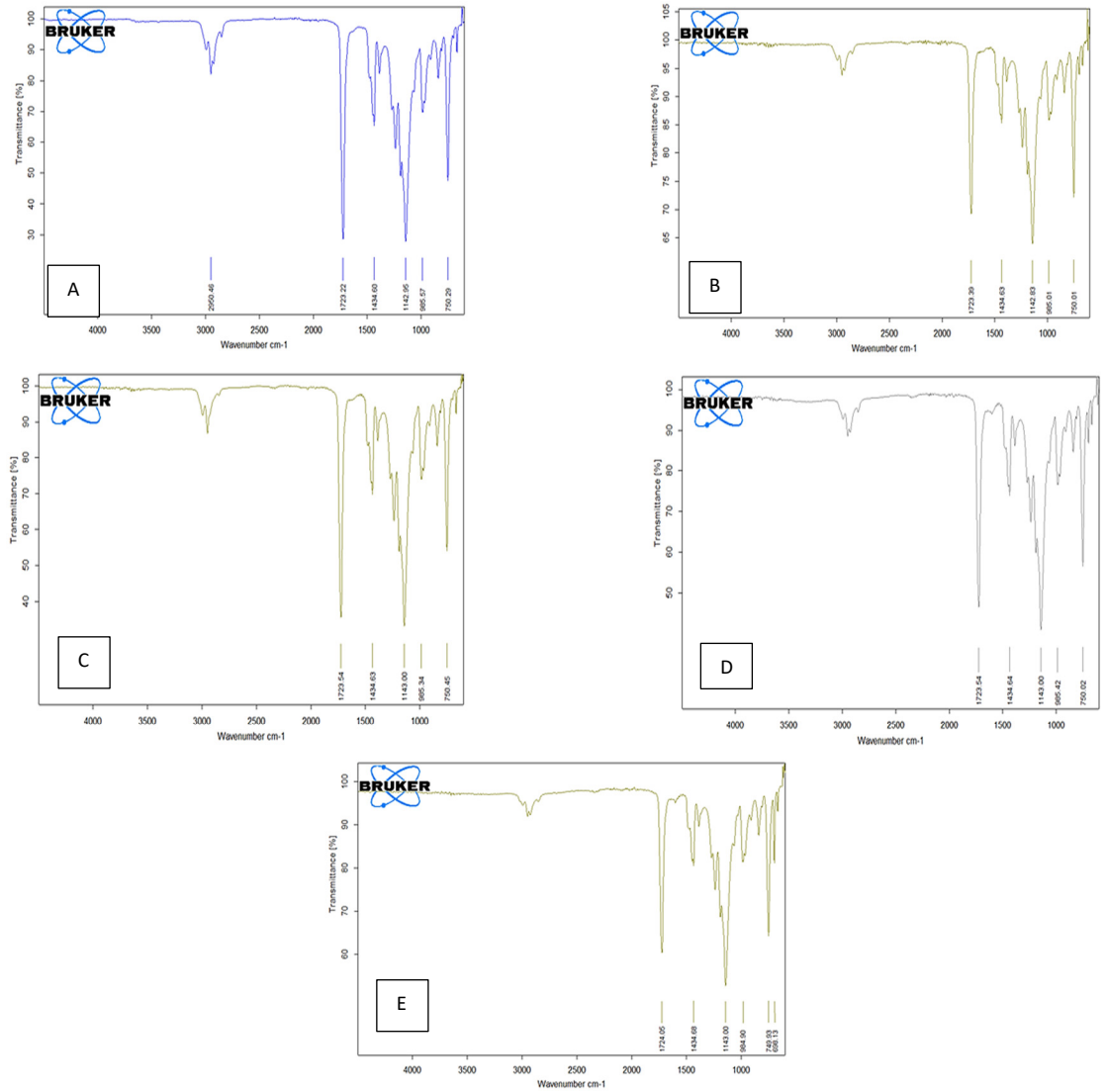

Figure 3. FTIR spectra of PMMA/Si_3N_4/TaC nanocomposites A. pure polymer, B. 2 wt.% of Si_3N_4/TaC NPs, C. 4 wt.% of Si_3N_4/TaC NPs, D. 6 wt.% of Si_3N_4/TaC NPs and E. 8 wt.% of Si_3N_4/TaC NPs

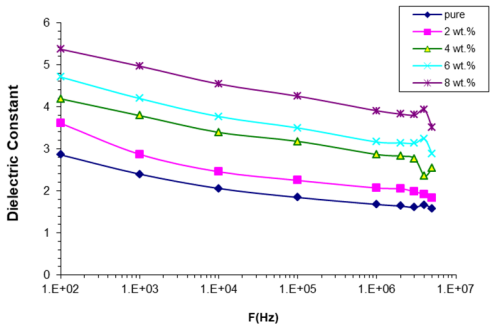

Figure 4. Variation of dielectric constant with frequency of PMMA/Si3N4/TaC nanocomposite

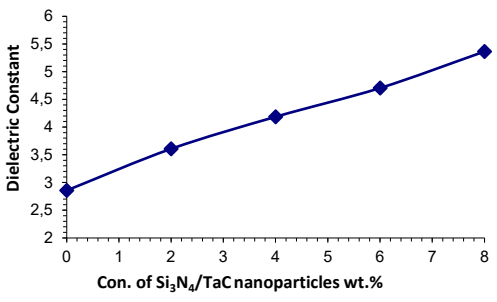

Figure 5. Effect of Si_3N_4/TaC nanoparticles concentrations on dielectric constant for PMMA/ Si_3N_4/TaC nanocomposite at 100 Hz

Equation (2) was used to calculate the dielectric loss (ε″) of the nanocompsites. Figure (6) shows the relation between dielectric loss of PMMA/Si_3N_4/TaC nanocomposites and frequency. The dielectric losses for nanocomposites reduce as the frequency increases for all samples. This phenomenon was linked to a reduction in the contributions of polarization of space charges. According to the data, nanocomposites have a substantial dielectric loss at low frequencies. Due to the reduced time available for the dipoles to align at high frequencies, the dielectric loss decreases [36-38]. The dielectric loss of PMMA/Si_3N_4/TaC nanocomposites as a function of Si_3N_4/TaC NPs are shown in Figure (7). From this figure, it is note that the dielectric loss rises with rising content of the Si_3N_4/TaC nanoparticle. This result attributed to increased charge on the dipole [39-41].

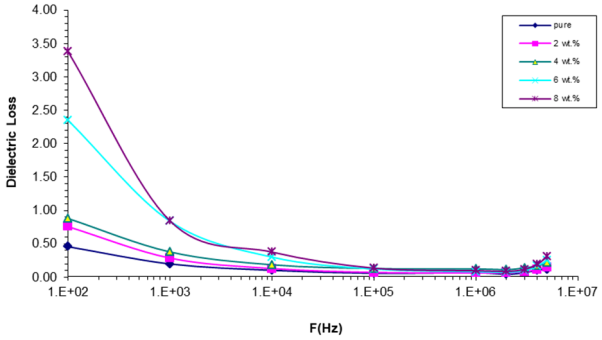

Figure 6. Variation of dielectric loss with frequency of PMMA/Si3N4/TaC nanocomposite

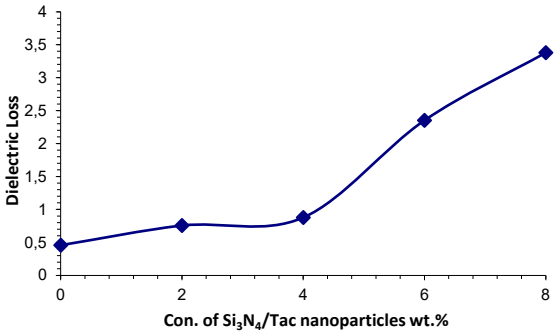

Figure 7. Influence of Si_3N_4/TaC NPs concentrations on dielectric loss for (PMMA/ Si_3N_4/TaC) nanocomposite at 100 Hz

Equation (3) was used to calculate the A.C electrical conductivity. Figure (8) shows the relationship between A.C. electrical conductivity of PMMA/Si3N4/TaC nanocomposites and frequency. The mobility of charge carriers and the hopping of ions from the cluster cause the A.C electrical conductivity of all specimens to increase as the frequency of the electric field increases. At low frequencies, the amount of mobile ions and electrical conductivity decreased due to increased charge accumulation at the electrode and electrolyte interface [42-44]. Because charge carriers moved more easily at high frequencies, the electrical conductivity of PMMA/Si_3N_4/TaC nanocomposites increases with frequency [45-47]. Figure (9) reveals that the electrical conductivity of nanocomposites rises with the rising of Si_3N_4/TaC nanoparticle content due to rise in the ionic charge carriers and the formation of a continuous network of Si_3N_4/TaC nanoparticles inside polymer matrix [48-50]. The results of the ε', ε″, and A.C. conductivity are shown in Table (1).

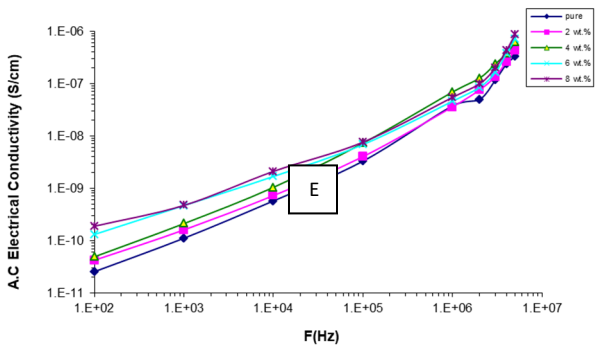

Figure 8. Variation of AC electrical conductivity with frequency of PMMA/Si3N4/TaC nanocomposite

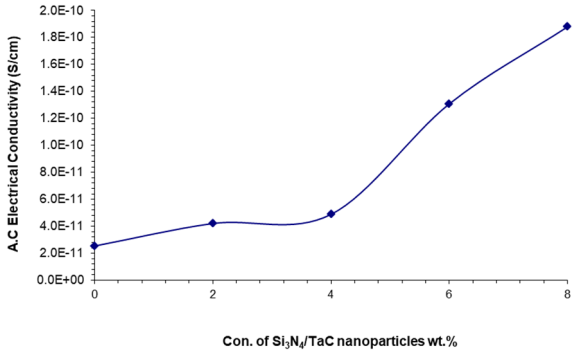

Figure 9. Effect of Si_3N_4/TaC NPs content on AC electrical conductivity for PMMA/Si_3N_4/TaC nanocomposite at 100Hz

Table 1. The values of the Dielectric constant, Dielectric loss and AC electrical conductivity at 100 Hz of (PMMA/Si_3N_4TaC) nanocomposites

Concentration of Si_3N_4/TaC (wt.%)	Dielectric constant	Dielectric loss	A.C. Conductivity (S/cm)
0	2.86	0.46	2.54E-11
2	3.61	0.76	4.21E-11
4	4.19	0.88	4.89E-11
6	4.71	2.35	1.31E-10
8	5.37	3.38	1.88E-10

4. CONCLUSION

This work summaries that the scanning electron microscope (SEM) indicate that the homogenous, smooth and dispersed of Si_3N_4 and TaC NPs inside the PMMA matrix due to strong covalent interaction between the Si_3N_4 and TaC NPs in the PMMA matrix which mean a good method for prepared films. Optical microscope (OM) images explained

that when increasing in content of nanoparticles that forming network paths inside the polymeric matrix that act as charge carriers. FTIR exhibited that when combined, the polymer and the nanoparticles exist in a physical superposition. at the concentrations of Si_3N_4 and TaC nanoparticles rise, the (PMMA/Si_3N_4/TaC) nanocomposites ε', ε″, and A.C conductivity also rise. When the frequency is raised, the A.C. electrical conductivity raised while the ε' and ε″ of nanocomposites decreased. This behavior make it may be considered as excellent electronics materials for electrical applications.

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ВПЛИВ НАНОМАТЕРІАЛІВ Si_3N_4/TaC НА СТРУКТУРНІ ТА ЕЛЕКТРИЧНІ ХАРАКТЕРИСТИКИ ПОЛІМЕТИЛМЕТАКРИЛАТУ ДЛЯ ЕЛЕКТРОТЕХНІКИ ТА ЕЛЕКТРОНІКИ

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У цьому дослідженні використовувався метод лиття для отримання нанокомпозитів PMMA/ Si_3N_4/TaC з різним вмістом (0,2,4,6,8) % мас. наночастинок (НЧ) Si_3N_4/TaC. Досліджено структурні та електричні властивості. Дослідження на скануючому електронному мікроскопі (SEM) вказують на однорідність, гладкість і дисперсію НЧ Si_3N_4 і TaC всередині матриці ПММА через сильну ковалентну взаємодію між НЧ Si_3N_4 і TaC в матриці ПММА, що означає хороший метод для підготовлених плівок. Зображення з оптичного мікроскопа пояснюють, що збільшення вмісту наночастинок утворює мережеві шляхи всередині полімерної матриці, які діють як носії заряду. Спектри FTIR вказують на фізичну інтерференцію між полімерною матрицею та наночастинками. Електричні властивості змінного струму нанокомпозитів показали, що діелектрична проникність і діелектричні втрати зростають зі збільшенням вмісту наночастинок і зменшуються зі збільшенням частоти прикладеного електричного поля. Тоді як електропровідність змінного струму зростає зі збільшенням частоти та ваги наночастинок Si_3N_4/TaC. Ці результати показали, що наноструктури PMMA/Si_3N_4/TaC можна вважати перспективними матеріалами для електроніки та електричних нанопристроїв.

Ключові слова: *нанокомпозити; ПММА; Si_3N_4; TaC; електричні властивості змінного струму*