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ENHANCEMENT OF STRUCTURAL AND OPTICAL PROPERTIES OF CMC/PAA BLEND BY ADDITION OF ZIRCONIUM CARBIDE NANOPARTICLES FOR OPTICS AND PHOTONICS APPLICATIONS†

Majeed Ali Habeeb*, Zainab Sabry Jaber
University of Babylon, College of Education for Pure Sciences, Department of Physics, Iraq
**Corresponding Author: pure.majeed.ali@uobabylon.edu.iq*
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Nanocomposites of (CMC-PAA-ZrC) made with different nano zirconium carbide percentages by casting method (0, 1.5, 3, 4.5, and 6 wt%). The results showed that FTIR spectra shift in peak position and change in shape and intensity, compared with pure (CMC-PAA) blend. Microscopic photographs show a clear difference in the samples when increasing proportions of zirconium carbide nanoparticles, when the concentration of zirconium carbide NP reached 6% wt, the nanoparticles make up a continuous network inside (CMC-PAA) blend. Structural and optical characteristics have investigated the findings showed that the absorption of (CMC-PAA-ZrC) nanocomposites increases with increasing of ZrC NPs, while transmission decrease. The absorption coefficient, extinction coefficient, refractive index, real and imaginary parts of dielectric and optical conductivity are increasing with rises concentration of ZrC. Also optical energy gap decreased from 4.9 eV to 4.05 eV and from 4.5 eV to 3.65 eV for allowed and forbidden indirect transition respectively with increasing ZrC NPs. The results indicate that the (CMC-PAA-ZrC) nanostructures can be considered as promising materials for optoelectronics applications.
Keywords: Nanocomposites, structural Properties, Zirconium Carbide NPs, Optical Properties
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1. INTRODUCTION

In order to change mechanical, electrical, optical and thermal properties, to meet the required characteristics, new polymers, blends, combinations and advanced materials must be made. This development is parallel to comprehensive studies aimed at clarifying the relationship between the modified materials and the structure and the property [1]. New polymer films must be optically, electrically and thermally characterized in order to manufacture them, optical films, multipliers, full reflectors, narrow pass band filters, etc. may be used as transparent films [2]. The CMC is a major industrial polymer with numerous uses, detergents, textiles, paper, foods, medical products and well-boiled oil, including flocculation, drag reduction. CMC is a cellulose derivative that results from the reaction of sodium and chloroacetic acid. A number of cellulose molecules that have been introduced to promote water solubility have been contained in sodium carboxymethyl groups (CH2COONa). Three factors depend on the diverse properties of CMC the molecular weight of the polymer, the average carboxyl content of the hydroglucose unit, and the distribution of carboxyl substitutes along the polymer chain [3,4]. Poly acrylic acid (PAA) has been gaining considerable interest due to its tremendous application in a variety of fields, including electrochemical, mechanical, biomedical, etc. It was widely used in various optical products, such as an effective corrosion inhibitor and eco-friendly platform, a solid lithium-ion electrolyte battery, super capacitors, ion replacements. PAA coatings can, in electronic and electrochemical applications, further improve the chemical stability of metal oxide nanoparticles. Thus, PAA related compounds are increasingly developed in the PAA matrix containing various oxide nanoparticles. Installations for physiotherapy [5,6]. For advanced temperature reactor fuels, zirconium carbide (ZrC) is an oxygen-based or inert-containing possible coating medium. ZrC has demonstrated desirable properties including excellent corrosion resistance and retention capabilities for fission product, for these fuel applications. However, ZrC processing results in stable sub stoic and carbon-rich compositions with and without major microstructural inhomogeneity and textural anisotropy, as well as phase separation, leading to physical, chemical, therapeutic and mechanical variations. Zirconium carbide (ZrC) is part of a class of materials known as ultra-high temperature ceramics (UHTCs) because of their high melting points (3550°C), low densities (6.7 g/cm^3), and strong thermal conductivity. The results of high-temperature neutron irradiation remain poorly understood at present [7]. Each bulk material, however, has low fracture strength, low resistance to oxidation and sintering potential, which limits its use in harsh environments. Zirconium carbide (ZrC) has both ceramic and metallic properties, such as one of the highest very high hardness (30-35 GPa) melting point, good wear resistance [8].

2. EXPERIMENTAL PART

In this work, 30% poly acrylic acids (PAA) and 70% carboxy methylcellulose (CMC) were prepared in 90 ml of distilled water with magnetic stirrer to achieve more homogeneous solution at temperature 80 C for 30 minutes. ZrC nanoparticles were added with concentrations (0, 1.5, 3, 4.5 and 6) wt% by using casting method. Using the microwave instead of traditional heat devices, as the use of a microwave oven reduces the reaction time and leads to obtaining a

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homogeneous solution and nanoparticles uniform in size and shape. Absorption spectra were recorded for (CMC-PAA-ZrC) nanocomposites in the wavelength range (200–800) nm, double beam spectrophotometer (UV/1800/Shimadzu) at room temperature the absorption spectrum was registered and used to obtain optical constants, absorption coefficient, extinction coefficient, refractive index and energy gap. To make additional remarks about the homogeneity in distributing zirconium carbide, the effect of the filler and the existence of impurities, a test for samples was conducted in different concentrations using an Olympus type Nikon-73346 optical microscope which has a magnifying power of (100×) and equipped with a camera used in the microscopic photography. FTIR spectra have been investigated by FTIR in the range of wave numbers (500-4000) cm^{-1} (Bruker company, German origin, type vertex-70). The following equation is calculated absorbance (A) [9]:

$$A = \frac{\mathrm{IA}}{\mathrm{I_o}} \tag{1}$$

where: I_A is the intensity of light absorbed by material and Io is the intensity of light incident.

Transmittance (T) is computed as the following equation: [19]

$$\mathrm{T} = \exp(-2.303\ \mathrm{A}) \tag{2}$$

The following equation is calculated for absorption coefficient (α): [11]

$$\alpha = 2.303\ \mathrm{A/t} \tag{3}$$

where t is the thickness of the sample

The indirect transition calculated by using relation: [12]

$$\alpha h\upsilon = \mathrm{B}(h\nu - \mathrm{Eg})^{r} \tag{4}$$

where: B is fixed, hυ is photon energy, Eg is optical band gap, r = 2 is permitted indirect, and r = 3 is prohibited indirectly.

The coefficient of extinction (k) was determined using the equation below: [13]

$$\mathrm{K} = \frac{\alpha\lambda}{4\pi} \tag{5}$$

where λ is the wavelength

Calculated refractive index (n) from equation: [14]

$$n = \sqrt{\frac{4R - k^2}{(R-1)^2}} - \frac{(R+1)}{(R-1)} \tag{6}$$

where: R is a reflection

Dielectric constant (real ε_1 and imaginary ε_2) parts are calculated by: [15]

$$\varepsilon_1 = n^2 - k^2 \tag{7}$$

$$\varepsilon_2 = 2nk \tag{8}$$

The optical conductivity (σ) is obtained by using the relation: [16]

$$\sigma = \frac{\alpha n c}{4\pi} \tag{9}$$

where: c is the velocity of light

3. RESULTS AND DISCUSSION

Figure 1 shows FTIR has been used to analyze the interactions among it interacts with the molecule because it has a dipole electric in (CMC-PAA-ZrC) nanocomposites. These interactions can include changes in the vibrational modes of the nanocomposites. The (FTIR) transmittance spectra of (CMC-PAA-ZrC) nanocomposites films with the different ratio of ZrC nanoparticles are shown in figure 1(A)–(E) are recorded at room temperature in the range (500–4000) cm^{-1}. It can be seen from the figure that the wide bands are observed at about (3566-3902) cm^{-1} due to OH groups. The band corresponding to CH2 asymmetric stretching vibration occurs at about 2360 cm^{-1}. The peaks at 1541 and 1698 cm^{-1} have been attributed to the CO, CC stretching mode. The absorbent peak at around (1338–1868) cm^{-1} belongs to CO group. The band relating to the poly acrylic acid CO group is located at 1698 cm^{-1}. In case of (CMC-PAA-ZrC) with different ZrC ratio, compared to pure (CMC-PAA) blend, FTIR spectra display variations in peak position as well as changes in form and strength. This shows the decoupling of two polymers and zirconium carbide nanoparticles between the related vibrations, the deviation towards greater or smaller wavelengths occurs as a result of compensation or the effect of solvents, as for the vertical displacement, it indicates an increase or decrease in the absorption value [17,18].

Figure 2 shows the optical microscope images of (CMC-PAA-ZrC) NCs taken at a magnification power of 10× for specimens with different concentrates of ZrC nanoparticles. When the proportion of zirconium carbide nanoparticles (shown in the images B, C, D, and E) is increased by microscopic photos, after the addition of the nanoparticulate content of ZrC a network is formed in the main phase of the (CMC-PAA) blend. This network has paths along which carriers will travel the paths that have a declining resistivity of the (CMC-PAA) blend [19].

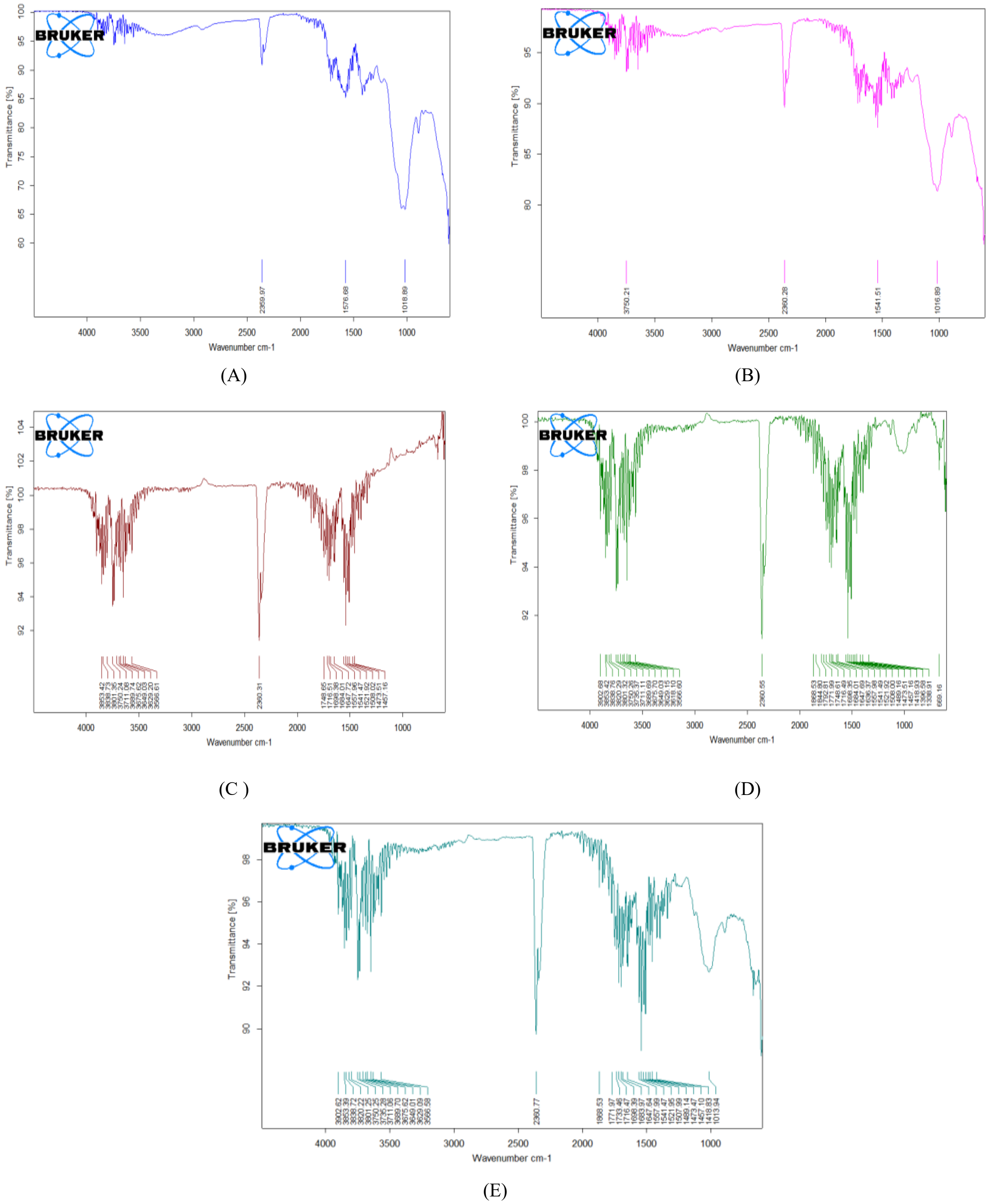
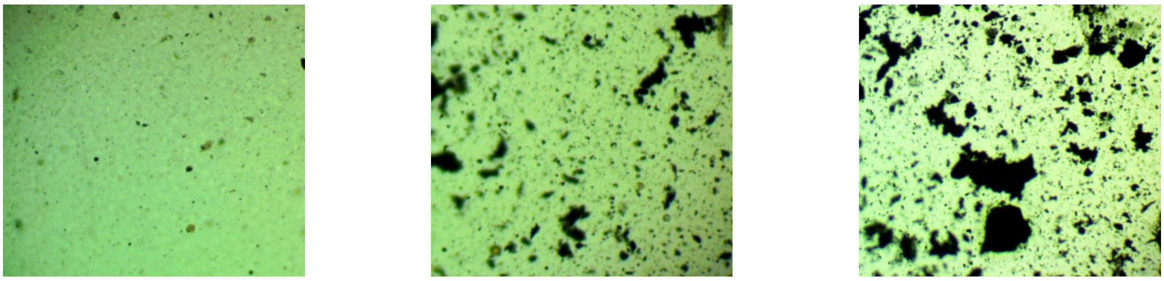


Figure 1. FTIR spectra for (PAA-CMC-ZrC) nanocomposites. (A) (PAA-CMC) blend, (B) 1.5 wt.% ZrC nanoparticles, (C) 3 wt.% ZrC nanoparticulates,(D) 4.5 wt.% ZrC nanoparticulates, (E) 6 wt.% ZrC nanoparticulates

Figure (3) shows variation of optical absorbance with wavelength for (CMC-PAA-ZrC) nanocomposites. From this figure the absorption is increased in the ultraviolet region and decreases in visible and infrared region, since the incident photon has a high wavelength and the photon is not transmitted because there is not enough energy to communicate with atoms. The photon-material interaction occurs and the photon absorbs as the wavelength decreases. The absorbance increases with increasing of ZrC concentration, this is due to free electrons absorbing the incident light [20,21].

Figure (4) shows the transmittance spectrum for (PAA-CMC-ZrC) nanocomposites as a function of wavelength. The transmission decreases with increase of the concentration of ZrC nanoparticles. The method is not followed by emission from the radiation because the transferred electrons in its outer orbits have occupied vacancy positions of the energy bands, absorb part of the light incident that does not exceed a material dosage which is induced by ZrC electrons on their outer orbits and transmits them to higher energy levels [22,23].



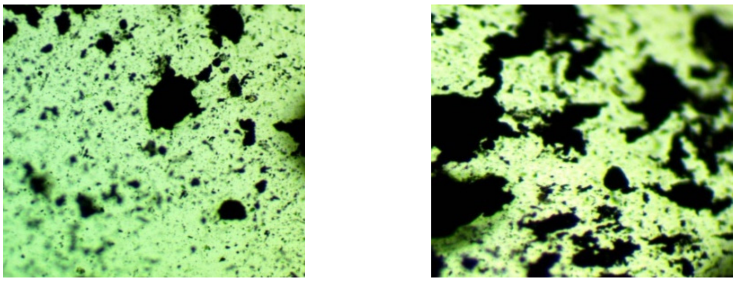


Figure 2. Photomicrographs (10x) for (PAA-CMC-ZrC) nanocomposites: (A) (PAA-CMC) blend, (B) 1.5 wt.% ZrC nanoparticles, (C) 3 wt.% ZrC nanoparticulates, (D) 4.5 wt.% ZrC nanoparticulates, (E) 6 wt.% ZrC nanoparticulates

The relation between the absorption coefficient and wavelength of (PAA-CMC-ZrC) nanocomposites shown in Figure 5. The absorption coefficient helps to illustrate the nature of electron transfer. When the absorption coefficient values of nanocomposites (CMC-PAA-ZrC) are strong $\alpha > 10^4$ cm^{-1}, the electron is passed directly. Although the absorption factor values of nanocomposites are tiny at $\alpha < 10^4$ cm^{-1}, the electron transfer is indirectly. We can see that with the rise in the concentrations of ZrC nanoparticles, the absorption coefficient is increased, due to increased carriers of charge [24], the absorption and absorption coefficient of (CMC-PAA-ZrC) NCs are also enhanced.

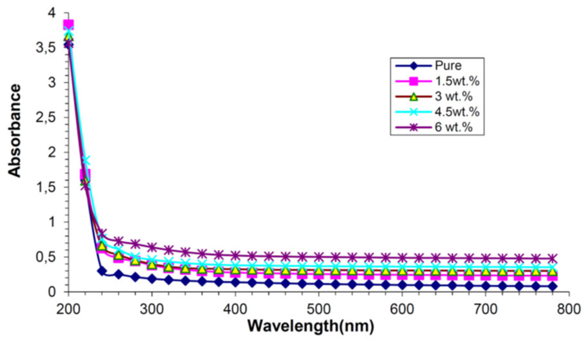


Figure 3. Absorbance spectra with photon wavelength for (PAA-CMC-ZrC) nanocomposites

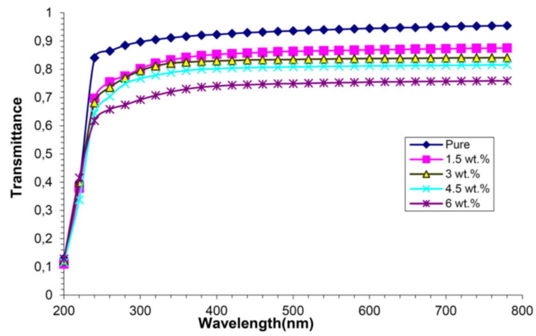


Figure 4. Transmittance spectra of (CMC-PAA-ZrC) nanocomposites as a function of wavelength

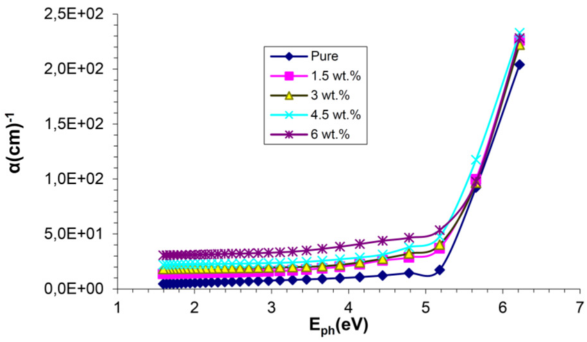


Figure 5. Variation of absorption coefficient for (CMC-PAA-ZrC) NCs with photon energy

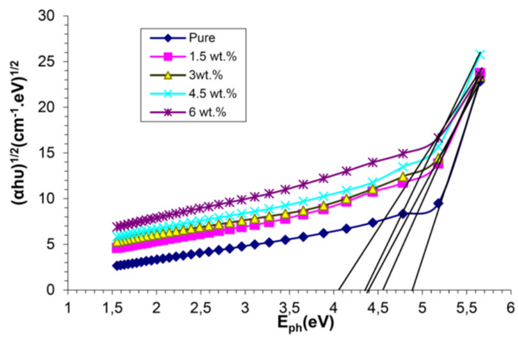
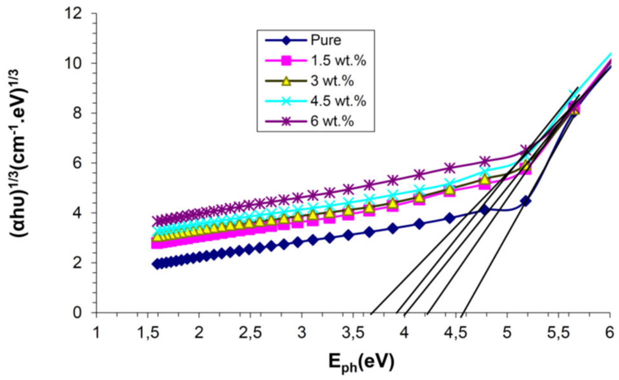


Figure 6. The relationship between $(\alpha h\nu)^{1/2}$$(\mathrm{cm}^{-1}\cdot\mathrm{eV})^{1/2}$ and photon energy of (CMC-PAA-ZrC) nanocomposites

Figure 6 demonstrates the relationship between absorption edge $(\alpha h\nu)^{1/2}$ and photons energy. This figure indicates an energy gap decreased by an increase in ZrC concentration for (CMC-PAA-ZrC) nanocomposites, this is because the material has risen in disruption, which means that the secondary excitation within the band can be made possible and the width of these levels increases with increasing concentrations of ZrC NPs, that minimize the energy gap [25,26.

The relation between $(\alpha h\nu)^{1/3}(cm^{-1}\cdot eV)^{1/3}$ and photon energy of **(CMC-PAA-ZrC)** nanocomposite is shown in Figure 7. This figure obvious that the energy gap values for prohibited indirect transition decline, this result is explained because the density of localized states increased with a rise in the concentration of ZrC nanoparticles [27]. As well as values of prohibited indirect transition are lower than the permitted indirect transition.

The change in the extinction coefficient is seen in Figure 8 as a function of wavelength. With an increase in ZrC nanoparticles we have noticed that the extinction coefficient increases. This is because the absorption value is enhanced and the ZrC nanoparticles weight percentage is increased. Variation the extinction coffiecient with wavelength of (PAA-CMC-ZrC) nanocomposites there is an effect of the geometric structure of the material on the tops extinction coefficient, when the percentage of nanomaterial increases, the proportion of geometric deformation increases in the crystal lattice [28-30].



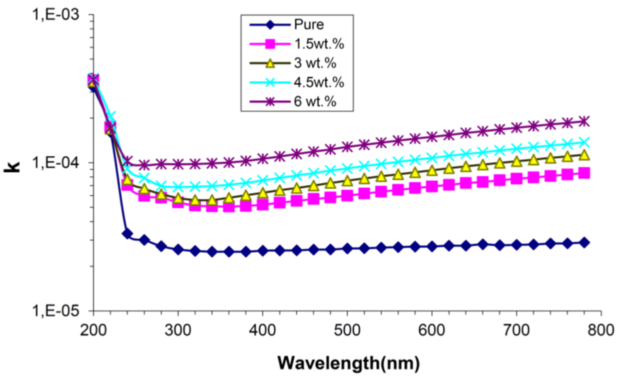
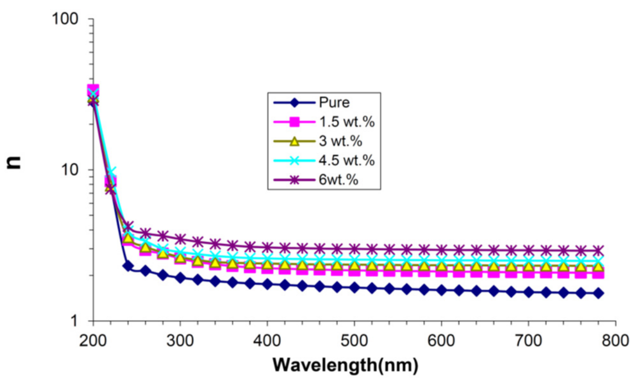


Figure 7. The relationship between $(\alpha h\nu)^{1/3}(cm^{-1}\cdot eV)^{1/3}$ and photon energy for (CMC-PAA-ZrC) NCs

Figure 8. Variation of extinction coefficient with wavelength of (PAA-CMC-ZrC) nanocomposites

Figure 9 shows the relationship between the refractive index and wavelength for (CMC-PAA-ZrC) nanocomposites. From this figure it can be seen by increases the concentration of ZrC nanoparticles the refractive index increases for all samples of (CMC-PAA-ZrC) nanocomposites. The ultraviolet region shows high refractive index values due to low transmission in this region however, due to the high transmission in this region, the visual and near IR regions notice low values [31,32].



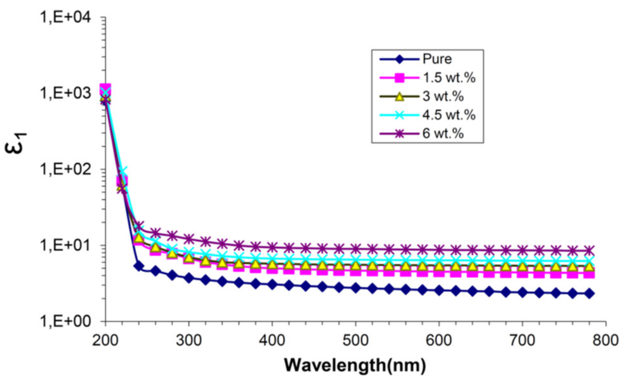


Figure 9. As a function of wavelength, refractive index for (PAA-CMC-ZrC) nanocomposites

Figure 10. As a function of wavelength the actual dielectric constant for (PAA-CMC-ZrC) nanocomposites

Figure 10 indicates the difference between the actual part of the dielectric constant with wavelength. These graphs demonstrate how raising the concentration ratio of ZrC NPs enhanced real part of dielectric constant. The rise in electrical polarization in the nanocomposites is responsible for this. On the other hand variation of ε_1 is primarily, depends on the n^2 because of little k^2 values, whereas ε_2 is mainly dependent on k values, the variations in the absorption coefficient are related to [33,34], as shown in Figure 11.

The relationship between optical conductivity and wavelength for (PAA-CMC-ZrC) nanocomposites is shown in Figure 12. It is noted that with increased zirconium carbide nanoparticles, optical conductivity increases. This behavior is attributed to the formation of localized states in the energy gap; increasing the concentrations of ZrC nanoparticles causes a rise in the density of localized levels in the energy gap, which raises the absorption coefficient and, as a result, the optical conductivity of the nancomposites [35,36].

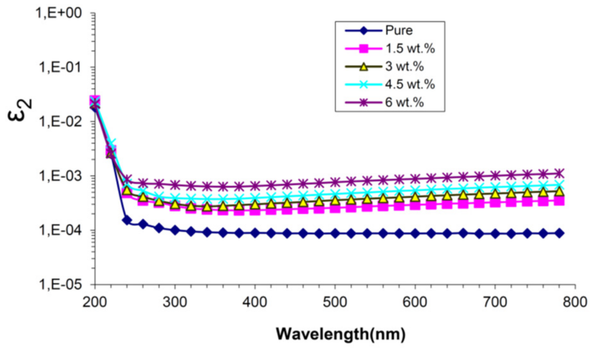


Figure 11. Change imaginary dielectric constant with wavelength for nanocomposites of (PAA-CMC-ZrC)

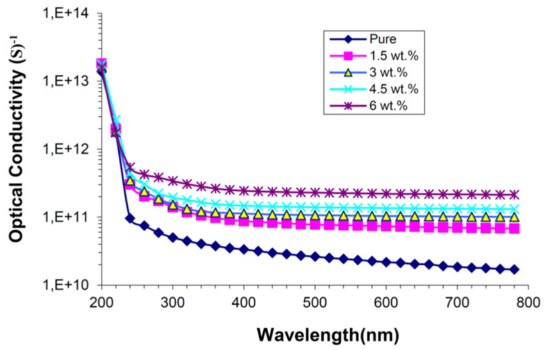


Figure 12. Optical conductivity of (PAA-CMC-ZrC) NCs as a function of wavelength

4. CONCLUSION

1 - FTIR spectrum shows both a difference in peak position, form and strength in contrast to the pure (CMC-PAA) blend. This suggests the disassociation between the vibrations in the two polymers and the zirconium carbide nanoparticles.
2 - Optical microscope appear forming a continuous network within polymers when the ratio of zirconium carbide nanoparticles reached (6 wt%).
3 - Absorbance of (CMC-PAA-ZrC) NCs increase with increasing of concentration of ZrC nanoparticles, while transmittance and energy gap of this nanocomposites decreases.
4 - The absorption coefficient, extinction coefficient, refractive index, real and imaginary part of the dielectric constants and the optical conductivity rise with an increase in the zirconium carbide weight percentage.

This finding suggests that (CMC-PAA-ZrC) nanocomposites are a good choice for use in different electronic and photonics applications.

ORCID IDs
Majeed Ali Habeeb, https://orcid.org/0000-0001-5064-2835

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ПОЛІПШЕННЯ СТРУКТУРНИХ ТА ОПТИЧНИХ ВЛАСТИВОСТЕЙ СУМІШІ CMC/PAA ШЛЯХОМ ДОДАВАННЯ НАНОЧАСТИНОК КАРБІДУ ЦИРКОНІЮ ДЛЯ ОПТИКИ ТА ФОТОНІКИ

Маджід Алі Хабіб, Зейнаб Сабрі Джабер
Вавилонський університет, Коледж освіти для чистих наук, Фізичний факультет, Ірак

Нанокомпозити (CMC-PAA-ZrC), виготовлені з різним процентним вмістом нанокарбіду цирконію методом лиття (0, 1,5, 3, 4,5 і 6 мас.%). Результати показали, що спектри FTIR зміщуються в положенні піку та змінюють форму та інтенсивність порівняно з чистою сумішшю (CMC-PAA). Мікроскопічні фотографії показують чітку різницю в зразках при збільшенні частки наночастинок карбіду цирконію, коли концентрація NP карбіду цирконію досягла 6% мас., наночастинки складають безперервну мережу всередині суміші (CMC-PAA). Досліджено структурні та оптичні характеристики. Результати показали, що поглинання нанокомпозитів (CMC-PAA-ZrC) збільшується зі збільшенням кількості наночастинок ZrC, а пропускання зменшується. Коефіцієнт поглинання, коефіцієнт екстинкції, показник заломлення дійсної та уявної частини діелектричної та оптичної провідності зростають із збільшенням концентрації ZrC. Крім того, розрив оптичної енергії зменшується з 4,9 еВ до 4,05 еВ і з 4,5 еВ до 3,65 еВ для дозволеного та забороненого непрямого переходу відповідно зі збільшенням наночастинок ZrC. Результати показують, що наноструктури (CMC-PAA-ZrC) можна розглядати як перспективні матеріали для застосування в оптоелектроніці.

Ключові слова: нанокомпозити, структурні властивості, НЧ карбіду цирконію, оптичні властивості