Original Article

One-Pot Pulsed Laser Ablation route assisted copper oxide nanoparticles doped in PEO/PVP blend for the electrical conductivity enhancement

A.A. Menazea\textsuperscript{a,b}

\textsuperscript{a} Laser Technology Unit, National Research Centre, Dokki, 12311, Giza, Egypt
\textsuperscript{b} Spectroscopy Department, Physics Division, National Research Centre, Dokki, 12311, Giza, Egypt

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\section*{ABSTRACT}

Polyethylene Oxide (PEO)/Polyvinyl pyrrolidone (PVP)/Copper Oxide Nanoparticles (CuONPs) were synthesized by one-step Pulsed Laser Ablation of pure Copper plate immersed in PEO/PVP (70/30 wt.\%) solution. PEO/PVP/CuONPs composite films were prepared using casting method. The role of copper nanoparticles on modifying PEO/PVP was demonstrated. The effect of different laser ablation times on the characterization of the prepared PEO/PVP/CuONPs composite films have been studied via XRD, UV-vis, PL, SEM, and Ac conductivity. XRD and SEM confirm the complexation between CuONPs and PEO/PVP matrix. The appearance of the characteristic absorption peak at 275 nm in UV-vis spectrum was attributed to the surface plasmon resonance (SPR) of CuONPs. The values of the direct and indirect optical band gap show a decreasing after doping PEO/PVP matrix by CuONPs. PL analysis confirmed that the existence of the CuO nanoparticles comprehensively reorders the delocalized n-electron system of PEO/PVP blend matrix. The behavior of dielectric constant and dielectric loss are progressively decreased as the frequency increased. The values of AC conductivity are increased as the laser ablation time increased. $M'$ and $M''$ values are increased by increasing the concentration of CuONPs in the PEO/PVP blend matrix, which induces an increase in the ion conduction of the prepared samples.

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1. Introduction

Nanomaterials have attractive properties, which are fundamentally very different with their mass state. A significant growth in the generation and manufacture of one-dimensional material has been achieved over the past decade. Metal nanoparticles have amazing nano-structural optical, electrical, magnetic, antibacterial, mechanical, and thermal properties. Particularly, nanowires and nanorods were used in the generation of nanocomposite and nano-devices [1–6]. Through the metal nanoparticles, Copper Oxide Nanoparticles (CuONPs) have a high importance according to their ability to effectively interact with light through dint of surface plasmon resonance (SPR) [7,8]. CuONPs are now very involved in electrical, catalytic, optical antibacterial and mechanical characterization because of their wide range of applications [9–12].

Pulsed Laser Ablation in Liquids (PLAL) method of synthesizing metal nanoparticles is one of promising new physical
methods for obtaining metal colloids. FLAL process considers a new fantastic route have simple experimental setup to preparation of high quality nanoparticles from bulk metal [13–17]. FLAL is a Top-Down approach gets nanoparticles having desired shape, size, chemical composition, non-toxic, and free chemical contamination, comparing by conventional routes [18–20]. Abd El-kader et al. and Menazea et al. prepared AgNPs by FLAL and doped in PVK [21,22]. Menazea et al. prepared PVA doped with different metal nanoparticles by FLAL [23]. Abdelghany et al. prepared zeolite nanoparticles by FLAL in catalytic for 1, 8-dioxa-octahydroxanthene and N-aryl-1, 8-dioxodecachloroacridine with molecular docking validation [24].

Polyethylene oxide (PEO) is a high thermal and chemical strength linear semi-crystalline polymer [25]. A large ordering of the phase of crystalline structure within PEO polymer confirms its conductivity, so it must be combined with amorphous polymer. Polyvinyl pyrrolidone (PVP) has an attractive characterization as a good thermal stability and mechanical strength [26]. PVP is an effective capping compound for nanoparticles (NPs) because of the functional group have an ion pair of electrons that help to stabilize metal nanoparticles inside its composite owing to the presence of both carbonyl groups [27].

Polymer blending is considered one of the best favorable routes for designing new polymeric materials and for the development of materials with a huge range of characterization [28]. The final result from blending system can be adapted to the specifications of applications that cannot be accomplished by one polymer alone. Polymer/Nanoparticles composite has a great interest in the present time to introduce a new opportunities for getting a new material have attractive properties that shows advantages in electrical, optical, and mechanical properties [29,30].

PVP was considered to be an excellent collaborator for PEO in polymer blend preparation due to its simple process capability and moderate electrical conductivity that related to its individual characteristics, in addition to high solubility of PVP in water, which is a typical PEO solvent, and also besides PVP is also an amorphous polymer and reduces the crystallinity of PEO blending process [31,32].

This work has been performed to obtain the synthesis of copper oxide nanoparticles in PEO/PVP solution blend via one-step pulsed laser ablation process via fundamental wavelength 1064 nm Nd:YAG nanosecond laser to enhancement the electrical properties of PEO/PVP blend at different laser ablation time. The structural, optical, and morphological characteristics of the prepared PEO/PVP/CuONPs samples were investigated by using: XRD, UV–vis, PL, and FE-SEM. Ac conductivity of the prepared sample was conducted as a function of frequency using A.C. measurements at room temperature.

2. Materials and method

2.1. Materials

PEO with molecular weight M.W. ≈ 40,000 g/mol was purchased from Sigma Aldrich in powder form and PVP with M.W. ≈ 72,000 g/mol was purchased from ACROS in powder form. Both PEO and PVP polymers were used as basic components of the polymeric blend. Deionized Water (DIW) was used a common solvent for each PEO and PVP. Copper plate was purchased with high purity of (99.999%).

2.2. Preparation of PEO/PVP blend solution

An appropriate weight percentages of PEO/PVP (70/30 wt.%) powders was added and dissolved in 100 ml DIW with continuous stirring for 21 h at 50 °C to complete the dissolution process until a homogenous viscous liquid slurry was established.

2.3. Preparation of PEO/PVP/CuONPs composite by FLAL

Copper oxide nanoparticles were synthesized and doped in the PEO/PVP blend by one-step FLAL process. The experiment setup of generation of nanoparticles by FLAL was described previously in details [19,33,34]. Nd:YAG pulsed nanosecond laser that emits its fundamental wavelength (λ) at 1064 nm was utilized as source of ablation process. The laser source has 3.6 W powers, 10 Hz pulse repetition rate, 7 nm pulse duration. Copper metal plate (4 mm radius × 3 mm thickness) was polished with emery paper and washed several times in DIW. The copper plate was immersed in a vessel’s bottom containing 15 ml of the previously prepared PEO/PVP (70/30 wt.%) solution. The thickness of PEO/PVP solution above the target was about 7 mm. The laser beam was focused perpendicularly on the copper plate by a lens with a focal length of 10 cm to enable the laser ablation on the surface. The laser ablation process was repeated different times by different laser ablation time; (5, 7.5, 10, 12.5, and 15 min).

The prepared films of PEO/PVP/CuONPs nanocomposite were formed via solution casting technique; the solution of each laser ablation time was poured into polypropylene dishes and dried in the furnace at 40 °C for 5 days. After drying process, the prepared films at the bottom of dishes were stripped off from the dishes and placed in heavily evacuated desiccators to avoid the absorption of moisture. The thickness of prepared films was in the range of 0.2 mm.

2.4. Characterization techniques

Several spectroscopic techniques were performed to the prepared PEO/PVP/CuONPs films to study its physical characterization. XRD diffractograms were obtained via (Schimadzu 7000, Japan) occupied with Cu-Kα radiation (λ = 0.154060 nm) generated at 30 kV and 30 mA, diffraction patterns were collected within the Bragg’s angle (2θ) ranging between 5° and 80°. UV–vis spectroscopy analysis was performed via JASCO (V-570) double beam spectrophotometer in the wavelength region of 200–1000 nm at room temperature. Photoluminescence spectra of the prepared samples were recorded at room temperature using spectrophotometer (Jasco, FP-6500, Japan). Surface morphology of the samples was examined via a Field Emission-Scanning Electron Microscope (FE-SEM) type (Quanta FEG 250, USA). AC conductivity measurements were carried out by using The Broadband Dielectric Spectroscopy (BDS) type (concept 40) Novocontrol High Resolution Alpha
Analyzer assisted by Quatro Temperature Controllers using pure nitrogen as the heating agent. The samples were measured at room temperature in frequency range from 0.1 Hz to 20 MHz.

3. Results and discussion

3.1. X-ray diffraction analysis (XRD)

Fig. 1a shows the X-ray spectra of pure PEO and PVP. The spectrum of PEO supports the semi-crystalline structure of PEO and shows peaks at 2θ; 19.26°, 23.65° and 26.39° which are assigned to (112), (120) and (222) respectively [31,35]. Several low intensity peaks have also been observed in PEO spectrum around 2θ; 12.90°, 14.69°, 28.1°, 30.75°, 36.12°, 39.59°, and 44.96°. The PVP spectrum supports the amorphous structure of PVP and shows two large peaks at 10.74° and 21.46° [36]. Fig. 1b displays the X-ray spectra of pure PEO/PVP blend and the blend doped by Copper nanoparticles at different laser ablation times. The pure blend film spectrum reveals two well-defined diffraction peaks at 2θ; 19.26° and 23.65° followed by low strong peaks at 2θ; 14.74°, 27.8°, 30.75°, and 36.27° which attributed to existence of PEO and confirms the semi-crystalline structure of PEO/PVP matrix. The XRD spectrum of PEO/PVP/CuONPs nanocomposite films obtains that the intensity of the main two diffraction peaks 19.26° and 23.65° are increased in 5 min laser ablation time then its intensity decreased from 7.5 min with increasing the laser ablation time until 15 min. Also, there are decreasing in the intensity of peaks at 14.74°, 27.8° with increasing the laser ablation time. This decrease indicates an increase in amorphous regions suggesting that the arrangement of CuONPs in the polymeric matrix PEO/PVP has now became unpredictable and induces a decrease in crystallinity degree. This finding suggests a complexation between the blend components and CuONPs, resulting in a decrease in intermolecular interaction between the polymeric chains PEO/PVP. Also, XRD spectrum of PEO/PVP/CuONPs nanocomposite films obtains appearance of the characteristic sharp diffraction peak of CuONPs at 2θ = 38.32° which corresponding to the plane (111) that indexed for single-phase with a monoclinic structure of CuONPs (JCPDS File No. 05-661) [37]. The appearance of this peak implies the existence of CuONPs within the PEO/PVP matrix. The absence of the most peaks in CuONPs may be due to the low loading of CuONPs in PEO/PVP system. This result confirms that further laser ablation of CuONPs can cause structural changes in PEO/PVP matrix. Fig. 1b shows that the intensity of this peak is increasing dramatically with increasing the laser ablation time. This increasing in this peak proved that the concentration of CuONPs was increased in PEO/PVP blend by increasing the laser ablation time.

The average crystalline size of CuONPs is calculated using Debye–Scherrer’s formula [38]:

\[ \text{Size} = \frac{K\lambda}{\beta \cos \theta} \]  

Where K is a dimensionless constant close to unity (shape factor), \( \lambda \) is the X-ray wavelength, \( \beta \) is the line broadening full width at half maximum (FWHM) and \( \theta \) is the Bragg angle. It was found that the average size of embedded CuONPs in PEO/PVP at 2θ = 38.32° is ranged between 18.32–34.25 nm. These values suggest that the size of CuONPs increased after embedding process within the PEO/PVP matrix which is verified by TEM micrographs and UV–vis analysis.

3.2. Optical properties (UV–vis)

Fig. 2a shows the absorption spectrum of pure PEO/PVP blend, PEO/PVP blend doped by different concentrations of CuONPs via one-step laser ablation at the spectrum range from 200 to 700 nm. The spectrum of pure blend PEO/PVP showed the only absorption peak that appeared at 207 nm within the UV region, that mainly can be attributed to \( n \rightarrow \pi^* \) electronic transitions [39] that confirms its semi-crystalline structure where it has almost zero absorption in the wavelength region (300–700 nm). After the doping with copper oxide nanoparticles, UV–vis spectra of PEO/PVP/CuONPs composite films at
different laser ablation time show a red shift in the peak position at about 207 nm to higher wavelength. This shift indicates that within the PEO/PVP there is a change in optical band gap and crystallinity that confirmed the complexation and interaction between CuONPs and PEO/PVP matrix. In addition, the UV–vis absorption spectra of PEO/PVP/CuONPs composite films in Fig. 2a achieve an apparent characteristic absorption peak at 275 nm. The existence of this peak is due to the band transition of copper electron from deep level valance band and also named the surface plasmon resonance (SPR) of CuONPs [40] that confirmed the successful formation of CuONPs in the PEO/PVP matrix. As the laser ablation time increases from 5 min to 15 min, the doping of CuONPs in the PEO/PVP matrix is increasing, leading to an increase in the SPR peak intensity suggesting the effect of quantum confinement [41]. CuONPs size is an indicator associated with the amount of surrounding stable polymer chains that prevent CuONPs from agglomerating. As the laser ablation time continues to increase, it is believed that the nanocomposite sample chains are cross-linked with each other, resulting in an increase in the amorphous region content as further confirmed previously by the XRD. Furthermore, the decrease in SPR peak broadness suggests that the narrow size distribution of CuONPs at higher laser ablation time and this observation is further asserted by the results of TEM.

The Optical absorption coefficients $\alpha$ of the prepared PEO/PVP blend and PEO/PVP/CuONPs samples at different ablation time as a function of the wavelength $\lambda$ was revealed in Fig. 2b and can be determined via Beer–Lambert’s formula [42]:

$$\alpha(\lambda) = \frac{2.303}{d} \frac{A}{\lambda}$$  \hspace{2cm} (2)

Where; $A$ is the absorbance and $d$ is the thickness of the films, and $\lambda$ is the film thickness. As the laser ablation time increases, it can be seen that optical absorption coefficients $\alpha$ is increased with increasing $\lambda$. Moreover, as the laser ablation time increases, the absorption edge shifted to lower photon energy, suggesting that the PEO/PVP optical band gap decreased with the introduction of CuONPs. The absorption edge values are tabulated in Table 1. The pure PEO/PVP blend absorption edge is 4.56 eV, this value decreased significantly by increasing the laser ablation time until reached 3.39 eV in PEO/PVP/CuONPs films (15 min).

The optical band gap $E_g$ for the prepared PEO/PVP blend and PEO/PVP/CuONPs samples at different ablation time can be directly measure via Tauc’s formula [43]:

$$\alpha \lambda^m = A (\lambda E_g)^{m}$$  \hspace{2cm} (3)

Where $h\nu$ is the photon energy, $A$ is the band tailing parameter and $m$ is the power, that describes the transition process in the K-space (m equal ½ for direct energy gap, and m equal 2 for indirect energy gap).

The direct and indirect optical band gap energy of PEO/PVP blend and PEO/PVP/CuONPs samples at different ablation time
Table 1 – Values of absorption edge, direct, indirect optical energy gap and refractive index of pure PEO/PVP and PEO/PVP/CuONPs composite.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Laser Ablation Time (min)</th>
<th>Absorption Edge (eV)</th>
<th>Direct (eV)</th>
<th>Indirect (eV)</th>
<th>Refractive index</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pure PEO/PVP</td>
<td>0.0</td>
<td>4.56</td>
<td>4.66</td>
<td>4.94</td>
<td>2.71</td>
</tr>
<tr>
<td>PEO/PVP/CuONPs</td>
<td>5.0</td>
<td>4.39</td>
<td>4.48</td>
<td>4.86</td>
<td>2.73</td>
</tr>
<tr>
<td>PEO/PVP/CuONPs</td>
<td>7.5</td>
<td>4.24</td>
<td>4.39</td>
<td>4.82</td>
<td>2.77</td>
</tr>
<tr>
<td>PEO/PVP/CuONPs</td>
<td>10.0</td>
<td>4.06</td>
<td>4.28</td>
<td>4.78</td>
<td>2.79</td>
</tr>
<tr>
<td>PEO/PVP/CuONPs</td>
<td>12.5</td>
<td>3.86</td>
<td>4.18</td>
<td>4.71</td>
<td>2.81</td>
</tr>
<tr>
<td>PEO/PVP/CuONPs</td>
<td>15.0</td>
<td>3.39</td>
<td>4.07</td>
<td>4.68</td>
<td>2.86</td>
</tr>
</tbody>
</table>

were performed by plotting \((\alpha\nu)^{1/2}\) and \((\alpha\nu)^2\) against \((\nu)\) which has been represented in Fig. 2c and d, respectively. According to the intercept of the best fit of the linear portion of \((\alpha\nu)^{1/2}\) and \((\alpha\nu)^2\) with \(\nu\) on x-axis, the values of direct and indirect energy band gaps had been calculated and recorded in Table 1.

From this table, it was obtained that the values of the direct and indirect optical energy band gap of pure PEO/PVP blend were 4.66 eV and 4.94 eV, respectively. Through increasing the concentration of CuONPs in PEO / PVP, these values are decreased through increasing the laser ablation time to 15 min reaching 4.07 eV and 4.68 eV, respectively. This decrease can be due to the existence of chemical bonding between the of PEO/PVP blend chains and CuONPs indicating that CuONPs will increase the number of energy-localized electronic states between the PEO/PVP valence and conduction bands, so that the electronic structure of the PEO/PVP matrix has been modified. In other words, this decreasing in direct and indirect optical energy band gap values induce lattice defect and increases the electronic disorder degree in PEO/PVP leading reduces in crystallinity, as confirmed in XRD analysis.

The refractive index \((n)\) was determined according to Dimitrov and Sakka equation in terms of indirect energy band gap [30]:

\[
n^2 - 1 = \frac{1}{n^2} + \frac{E_g}{20} \tag{4}
\]

The values of \((n)\) are calculated and listed in Table 1. These values indicate an increasing trend for the PEO/PVP/CuONPs samples by an increasing the laser ablation time. This, in addition, leads to an increase in the average number of coordination of the studied PEO/PVP/CuONPs films due to an increase in the number of non-bridging oxygen bonds (NBO) bonds between CuONPs and the OH group adjacent to of PEO/PVP blend [44], which increases the refractive index. The increase in CuONPs within PEO/PVP blend led to an increase in the dispersion of the incident light, as well as a decrease in the speed of light passing and thus increased the refractive index. Such improvement in \(n\) improves the usefulness of these components in optical and optoelectronics applications.

3.3. Photoluminescence studies (PL)

A Photoluminescence (PL) analysis is an important tool for evaluating the optical characterization of materials and obtaining the strong interaction between the prepared composite samples component; PEO/PVP and CuONPs.

PL emission is largely owing to the existence of defects like impurities or vacancies of materials which results in the recombination of electron bound to donor and free holes and its intensity is directly proportional to the rate of electron hole pair recombination. PL spectra in room temperature of the prepared PEO/PVP blend and PEO/PVP/CuONPs samples at different ablation time are shown in Fig. 3. PL spectra were performed using a Xenon laser as light source at 350 nm excitation wavelength. The PL spectrum of the pure PEO/PVP blend shows strong photoluminescence emission and violet band peak centered at 413 nm nearly falls in the visible region [45]. Existing of PL for given wavelength is related to optical transitions between valence and conduction bands.

PL measurements of PEO/PVP/CuONPs samples show a drastically reducing in the intensity of the PL band, which could be due to the concentration-quenching effect [46]. Also, PL of PEO/PVP/CuONPs samples shows the peak centered at 413 nm with more broadness, the broad emission peaks occurred in the visible region can be attributed to the deep-level structural defects and vacancies singly ionized oxygen present in CuONPs [47]. The low intensity of PL band was at 5 min laser ablation time. After that, the intensity of the
PL band starts to enhance by increases the laser ablation time. In other words, we can say that the PL intensity of PEO/PVP/CuONPs samples was increases as the concentration of CuONPs increases.

A significant red shifting in PL peak position from 413 nm to 419 nm was obtained by increasing CuONPs concentration respectively as related the laser ablation time from 5 min to 15 min in PEO/PVP matrix. Therefore, the shift in the PL emission band position may be the result of the quantum confinement effect [48] due to the size of nano-crystallites controlled by ablation parameters. These results indicating that the existence of the CuO nanoparticles comprehensively reorders the delocalized n-electron system of PEO/PVP blend matrix. Results of optical measurements (UV–vis and PL) indicate great promise of the CuONPs doped in PEO/PVP blend films prepared in the present study are expected to be more useful in photonics and optoelectronics applications.

3.4. Scanning electron microscope (SEM)

Scanning electron microscopy (SEM) studies were carried to illustrate the changes resulting for the addition of CuONPs on surface morphological of PEO/PVP blend. Fig. 4 depicts FE-SEM photos of pure PEO/PVP and PEO/PVP blend doped by Copper nanoparticles at different laser ablation times for 5 min laser ablation time (a), 10 min (b), and 15 min (c).

In Fig. 4a, FE-SEM photos of the surface of pure PEO/PVP blend shows a smooth homogeneous and coherent morphology structure of the prepared blend and confirms the compatible between the two polymers PEO and PVP. In Fig. 4 (a–d), FE-SEM photos of the surface of pure PEO/PVP blend doped by CuONPs was obtained. These figures show an increase of the doping CuONPs on the surface of the samples by increasing laser ablation time. For laser ablation time 5 min, the Fig. 4b shows a small number of bright copper nanoparticles with the most beautiful homogeneous distribution on the surface of the sample. By increasing the laser ablation time at 10 min in Fig. 4c, we saw that the increasing of laser ablation time was followed by some agglomerates that start to obtain in certain parts of the sample. In Fig. 4d for 15 min laser ablation time, the CuONPs were aggregates in the surface and obtained as big granules and obtained as bright spot. It was obtained from Fig. 4 (c,d) that the CuONPs was imbedded step by step inside the PEO/PVP blend with increasing the laser ablation time from 5 min to 15 min. These observations indicate the complexation process between CuONPs and PEO/PVP matrix.

3.5. AC electrical conductivity

In this section, the AC electrical conductivity and the dielectric properties of the prepared PEO/PVP blend and PEO/PVP/CuONPs samples at different ablation time (5, 7.5, 10, 15 min) as a function of the frequency in the range (0.1 Hz–20 MHz) at room temperature were investigated according the given data that represents in Figs. 5, 6, 7 and 8.

3.5.1. Dielectric analysis

Fig. 5 (a and b) represent the dependence of frequency on the dielectric constant ε’ and the dielectric loss ε’’ for the prepared samples, respectively. The complex dielectric of a material is given by to Debye according the following equation [49]:

$$\varepsilon = \varepsilon' + j \varepsilon''$$

(5)
3.5.2. Conductivity analysis

Fig. 6 (a and b) obtains the real $\sigma_{ac}'$ and imaginary $\sigma_{ac}''$ part of conductivity for the prepared samples in dependence of frequency (log $f$). The values of $\sigma_{ac}'$ (Fig. 6 a) are very small at low frequencies for the sample then these values start to increase by increasing the frequency. This increasing can be related to space-charge polarization [51]. We observed that the values of $\sigma_{ac}'$ are increased as the laser ablation time increased at higher frequencies and also observed that the values at 15 min laser ablation time have a high value compared by the sample at other concentration.

The values of $\sigma_{ac}''$ (Fig. 6 b) are very small at low frequencies for all prepared samples then these values begin to decrease by increasing the frequency. It can be obtained that the values of $\sigma_{ac}''$ are decreased as the CuONPs concentration increased at high frequencies.

3.5.3. Electric modulus analysis

The complex modulus ($M'$) can be evaluated by the following mathematical formula [52]:

$$M' = M'' \cdot M''^* = \frac{1}{\varepsilon''} = \frac{\varepsilon' + j\varepsilon''}{(\varepsilon')^2 + (\varepsilon'')^2}$$  \hspace{1cm} (7)

An indication of electrical properties is given to the real modulus ($M'$) and imaginary modulus ($M''$) according to the relaxation of the electrical field. The values of $M'$ and $M''$ are determined using the complex modulus data as [53]:

$$M' = \frac{\varepsilon'}{(\varepsilon' + \varepsilon'')^2}$$  \hspace{1cm} (8)

$$M'' = \frac{\varepsilon''}{(\varepsilon' + \varepsilon'')^2}$$  \hspace{1cm} (9)

Fig. 7 (a and b) obtains the real $M'$ and imaginary $M''$ part of electrical modulus for the prepared samples in dependence of frequency (log $f$). It was observed that $M'$ is increase with the increase of frequency for all prepared PEO/PVP blend and PEO/PVP/CuONPs samples. Also, $M''$ values are increase with increasing the concentration of CuONPs in the PEO/PVP blend matrix causing an increase of the ionic conduction of the samples.

On the other hand, it is observed in Fig. 8a that the imaginary part $M''$ of pure PEO/PVP and PEO/PVP/CuONPs is characterized by two different relaxation peaks in lower and higher frequency regimes. The relaxation peak at low frequency was related to the interfacial polarization or MWS polarization process [54], whereas, the peak at high frequency was related to the cooperative chain segmental motion. The broadness of $M'$ for the prepared samples is observed due to the distribution of relaxation time because of the inhomogeneous behavior of the relaxing dipoles of the samples. This broadness is also increased as the frequency increase implies an evidence of the nature of the relaxation processes. There is a clear increase in $M'$ peak with CuONPs increase and shift to the lower frequencies. In addition, the maximum $M'$ peak value for the doped samples increases compared to the pure PEO/PVP blend, implying that the CuONPs relate to the relaxation process.
3.5.4. Complex impedance analysis

Fig. 8 (a and b) obtains the real $Z'$ and imaginary $Z''$ part of impedance for all prepared PEO/PVP blend and PEO/PVP/CuONPs samples in dependence of frequency (log $f$) in the range between 0.1 Hz and 10 MHz at room temperature. Ac-impedance complex $Z$ can be obtained by the following equation in terms of their real $Z'$ and imaginary parts $Z''$ of the ac impedance according to the equation [56]:

$$Z = Z' + jZ''$$

The both values of $Z'$ and $Z''$ are very high at low frequencies for the prepared pure PEO/PVP and PEO/PVP/CuONPs sample.
Then these values start to decease as the frequency increasing. This increasing is continuous until it reached constant values at the higher frequencies for all the prepared samples. We can observe that the values of $Z'$ and $Z''$ at low frequencies are increased as the concentration of CuONPs increased in the PEO/PVP blend matrix.

4. Conclusion

In this study, Nd:YAG was utilized to synthesis of PEO/PVP/CuONPs via one-step PLAL method. PEO/PVP/CuONPs films with different ablation time were prepared by traditional casting technique. The role of CuONPs in enhancement the electrical properties of PEO/PVP was discussed. XRD obtains the characteristic diffraction peak of CuONPs at 2θ = 38.32° which corresponding to the plane (111) plane that corresponds to CuONPs (JCPDS File No. 05-661). Results of optical measurements (UV-vis and PL) indicate great promise of the CuONPs doped in PEO/PVP blend films.

prepared in the present study are expected to be more useful in photonics and optoelectronics applications. FE-SEM scans show a clear change in the morphology of the prepared samples confirming the doping of CuONPs on the surface. Both $\varepsilon'$ and $\varepsilon''$ values were increased with increase the laser ablation time but also decreasing its values reaching the constant values at higher frequencies. The values of $\sigma_{ac}'$ are increased as the laser ablation time increased at higher frequencies. The values of $Z'$ and $Z''$ at low frequencies are increased as the concentration of CuONPs increased in the PEO/PVP blend matrix.

Conflicts of interest

The authors declare that they have no conflict of interest.

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