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Mechanical and electrical properties evaluation of PVA-carbon dot polymer nanocomposites

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The main objective of this study is to investigate the effect of incorporationC-dots within polyvinyl alcohol (PVA) matrix for improving the mechanical and electrical properties. The thin films of these composites are prepared by the polymer intercalation solution casting method with various C-dot nanoparticles loading (0.5, 1.0, 1.5 and 2.0wt%) and compared properties with the pristine PVA. The fabricated nanomaterial composite films are characterized using well-known techniques such as Fourier transform infrared spectroscopy (FTIR) and X-ray diffractometer (XRD). From the analysis of FTIR and XRD results, it was estimated that the C-dot nanofiller has strong interaction with PVA polymer matrix. Further, from the mechanical properties estimation, it is observed the elastic modulus of 37 MPa increased by 98.05% when compared to pristine PVA. Tensile strength is found to increase with increase in filler concentration, 300% increase in tensile strength was observed for 2wt% filler loading compared to the pristine PVA. Electrical properties are strongly dependent on C-dot nanofiller loading, and the same was analyzed using LCR meter. It has been observed that the dielectric constant decreased with an increase in frequency. AC conductivity was estimated with respect to the frequency, the highest conductivity (7×10^{-5} Sm^{-1}) was observed for 1.5wt% of C-dot nanofiller PVA composite.

Keywords: Carbon dots, Conductivity, Fluorescence spectra, Mechanical properties, PVA composite.

Wide range of applications of nanomaterial polymer composites has continued to attract the researchers' attention because of their various unique structural, mechanical, electrical, and optical properties. Polymer nanocomposites exhibit enhanced characteristics than those of the pure polymers, without substantial alteration in polymer processing. These composite materials with metal nanoparticles incorporated into a host polymer matrix shown to have a larger matrix-nanofiller interfacial area that is the key research area of interest in these polymer-metal nanocomposites. In addition, they have improved mechanical and electrical properties. These unique characteristics of nanocomposites offer exceptional research opportunities for exciting technological advances in the field of material science. The characteristics of these nanocomposites were analyzed by the organic and inorganic ingredients reinforced in the host matrix of polymers[1–6]. Due to their enhanced properties such as stiffness, high tensile strength and other relevant mechanical properties, an essential usage of nanomaterials today is in reinforcing polymer composites[7].Composite materials containing nanomaterials have gained their importance in advanced technological materials for use in photoelectric conversion, transistors because of good electrical conductivity[8,9]. In general, the efficiency of reinforcing nanoparticles in the hostmatrix is inversely proportional to the size of nanomaterial and directly proportional to the surface-to-volume ratio of nanomaterial dispersed in the host matrix. Thus, the geometrical arrangement of nanomaterial is one of the vital factors that affect the performance of reinforcement of the nanoparticles dispersed composites. It is reported that the blending result can be effectively influenced by increasing the surface-to-volume ratio of the nanofiller[4–7].

Although considerable research is being carried on various polymer nanocomposites, the effect of the addition of C-dot nanoparticles as nanofiller materials on material and mechanical properties of composites is an unexplored field of research. The exceptional properties of these composite materials such as excellent biocompatibility and low toxicity offered favourable C-dot materials for plenty of applications in bio-imaging, biosensor, and drug delivery[10]. Further, based on the excellent properties such as optical and electronic, C-dot materials can also be adopted in applications of catalysis, sensors, and optronics[11,12].

Research work on the synthesis of C-dots from various organic sources has been explored in the past[10-14]. For example, researchers have reported the luminescent mechanism, fluorescent and electrochemical properties of various composites based on C-dots. Thus in the present work, C-dots were prepared using lactose as a precursor material and used for the thin film composite studies. Synthesis of composite films by reinforcing the C-dots in polyvinyl alcohol and evaluation of the effect of carbon dot concentration on mechanical and electrical properties were studied. For this research, PVA (polyvinyl alcohol) was used as a host matrix material for reinforcement of nanofillers as it has compelling properties such as the ability to form a good film, good chemical compatibility, and excellent mechanical properties[14,15]. In addition, this PVA as a host matrix facilitates the preparation of nanocomposites with uniform distribution of particles inside the host matrix due to its hydrophilic nature[16-19]. C-dots as filler in the synthesis of polymer composite, has not been explored. Therefore, it is decided to synthesis PVA-C-dots nanocomposites and to study its mechanical and electrical properties. Thus, in the present study, C-dots reinforced polymer matrices synthesized by the C-dots reinforced polymer matrices were characterized using UV-Vis spectra and fluorescence spectra. The casted films were analyzed using XRD and FTIR for surface morphology and polymer matrix interaction with the filler. The synthesized polymer matrix was analyzed for enhancement in the mechanical and electrical properties.

Experimental Section

Reagents and materials

All chemicals used in the study were used as procured without further purification. Lactose ($C_{12}H_{22}O_{11}$) AR grade was procured from Merck. NaOH extra pure and PVA $(C_2H_4O)_n$ (AR grade) having molecular weight 85000 to 124000 (86-89%) were procured from SDFCL (SD Fine Chemicals Ltd.), Mumbai, India. Double distilled water was used for the synthesis of the films.

Synthesis and characterization of C-dot nanoparticles

Fluorescent C-dots were synthesized by the dehydration of lactose using a domestic microwave. A 20 mL of an aqueous mixture with 1:1 volume ratio of lactose and NaOH solution was taken in a glass beaker. Then the mixture is heated in a microwave oven. Two mL polyethylene glycol was added to stabilize the C-dots. The solution of C-dot was obtained, and it was purified using dialysis for about 24 h. Later, the sp^2 hybridized C-dots were obtained within 30 seconds and dispersed uniformly in the aqueous solution, followed by a change in colour from colourless to yellow. The synthesized C-dots were then characterized using the UV-visible and fluorescence techniques. The UV-visible spectrum for the synthesized C-dots is shown in Fig. 1. A high absorption spectrum due to the conjugated C bonds (absorption band at 250-300 nm and 400 to 420 nm) which is due to the C=C, π-π*, C=O and n-π* bonds.

The excitation edge for C-dots and an emission peak were observed at around 320-400 and 450-600 nm, respectively in the fluorescence spectra, as shown in Fig. 2. Further, it can be observed that the bright

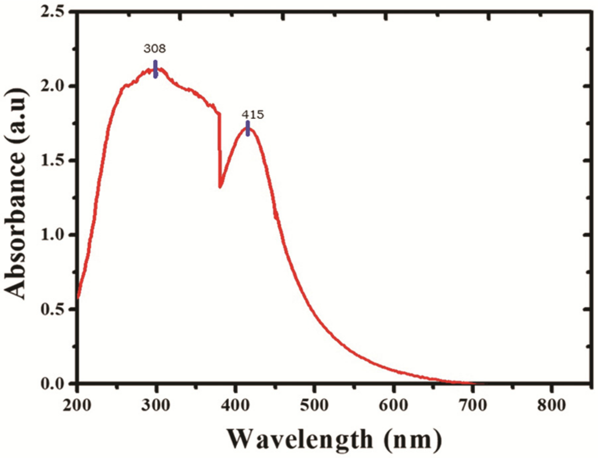

Fig. 1 — UV-Visible spectrum of the C-dot nanofillers.

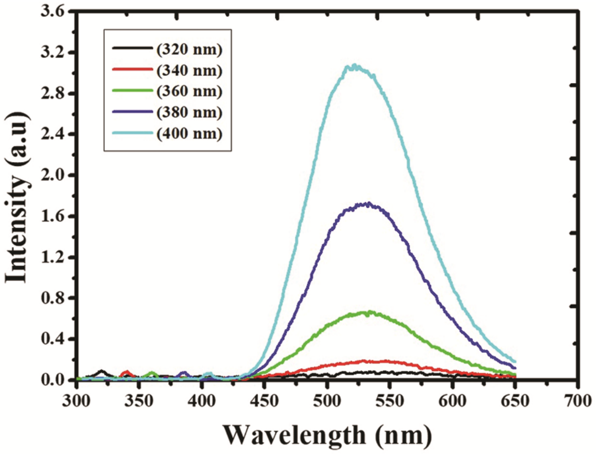

Fig. 2 — Fluorescence spectra (excited at 320 to 400 nm) of C-dot nanofillers.

fluorescence that was originating from the synthesized C-dots. The emission spectra recorded for excitation wavelengths between 320 and 400 nm showed that the fluorescence emission increases as an increase in the excitation wavelength. It is further observed that the maximum emission fluorescence centred at 550 nm due to the excitation at 400 nm wavelength. Furthermore, a strong fluorescence exhibited by the C-dots may be responsible for the quantum confinement of the passivity surface energy traps[20].

Fabrication of PVA-C-dot nanocomposite films

PVA–C-dot nanofiller composites were synthesized by the solution casting technique. Films of these composites were cast from a C-dot water suspension, in which PVA matrix was dissolved by heating at 65 ± 5 °C with constant stirring on a water bath for 4 h. However, a solid content in the PVA solution was maintained at 7.5 wt%. Then this solution was ultrasonicated at 65 ± 5°C for 45 min to ensure uniform mixing. The solution was casted on a clean glass-mold and allowed to dry at ambient temperature for 48 hr. Nano filler PVA films were fabricated by varying amounts of stabilized C-dot nanoparticles viz. 0.5, 1, 1.5 and 2 wt% and ensured that the casted films were free from air bubbles. C-dot nanoparticles were uniformly distributed in the PVA host matrix. The digital Vernier calliper was used to measure the thickness of the films fabricated and found to be between 0.19 and 0.21mm.

Measurements

FTIR spectra of the films were captured by plotting a graph of % transmittance versus wave number (cm^{-1}) in the spectral range of 4000-500 cm^{-1} using Bruker Alpha FTIR with the resolution of 4 cm^{-1}. The powder X-ray diffraction (XRD) analysis was carried out using a Brucker D-8 advanced wide-angle diffractometer. X-Ray patterns were recorded in the range of $2\theta = 0–60°$. LCR meter (Agilent 4294A) was employed for electrical characterization in the frequency range of 4 kHz to 1MHz. These tests were performed at a constant potential of 1V at room temperature. The mechanical properties of PVA–C-dot nanofiller composite films and pristine PVA were measured using Universal Testing Machine (KIPL-PC 2000 UTM). The tensile tests were carried out in accordance with the ASTM standards (D638). The load cell was set at 0.2kN and UTM had an error range of ± 0.2 MPa. The thickness and width of the characterized thin films were in the range of 0.19-0.21 mm and 17-20 mm, respectively.

Results and Discussion

Understanding the interaction of C-dot nanofiller with the PVA host matrix as well as some insights on the surface morphology of PVA–C-dot nanocomposite films is essential to know the effect of the addition of the filler in PVA nanocomposites. Sophisticated material characterizations such as FTIR and XRD were performed on the synthesized thin-film composite materials.

Fourier transform infrared spectroscopy (FTIR)

Figures 3(a) and (b) indicate the relationship between the % transmittance versus wave number (cm^{-1}). PVA–C-dot nanocomposite films were analyzed using FTIR spectra to understand the functional groups between the nanomaterial and PVA matrix. The spectral range (wave number) was maintained for all samples from 4000 to 400 cm^{-1} with a resolution of 4 cm^{-1}. The FTIR spectra of PVA and C-dot filled PVA at different percentages are shown in Figures 3(a) and (b). Figure 3(a) represents transmittance from wavelength 400 to 750 cm^{-1} and Figure 3(b) from 750 to 4000 cm^{-1}. From the spectra, it can be observed a strong absorption band at 3000–3300 cm^{-1} for the O–H stretching vibration of PVA. The group equivalent to the C–H asymmetric stretching vibration followed at 2914-2925 cm^{-1}. The band corresponding to C=O stretching vibration (vinyl acetate group of PVA) occurred at the peak of 1713-1736 cm^{-1}. The bands at 1416-1431 and 1300-1332 cm^{-1}, respectively are corresponding to the C–H flexible of $–CH_2$ in PVA backbone and $–CH_2$ wagging. Another stronger group detected at 840-845 cm^{-1} has also been recognized for $–CH_2$ in an enlarging manner. A peak corresponding at 1080-1090 cm^{-1} is credited to C–O–C enlarging of $–CO–CH_3$ (acetyl group) existing on PVA strength. Supplementary peaks in FTIR spectra was not recognized; on the other hand, a significant change in the peak position of the group corresponding to C–O–C and C=O stretch vibrations, respectively, it has specified a physical interface among the hydroxyl groups of C-dot nanofiller and PVA. The probable mechanism or interaction between CD's and PVA is due to the formation of hydrogen bonding between –OH group of PVA and –COOH or $–NH_2$ group present in C-dot's[21]. The charge-transfer occurred due to the O-H group of PVA have a strong tendency with C-dot nanoparticles through chelation.

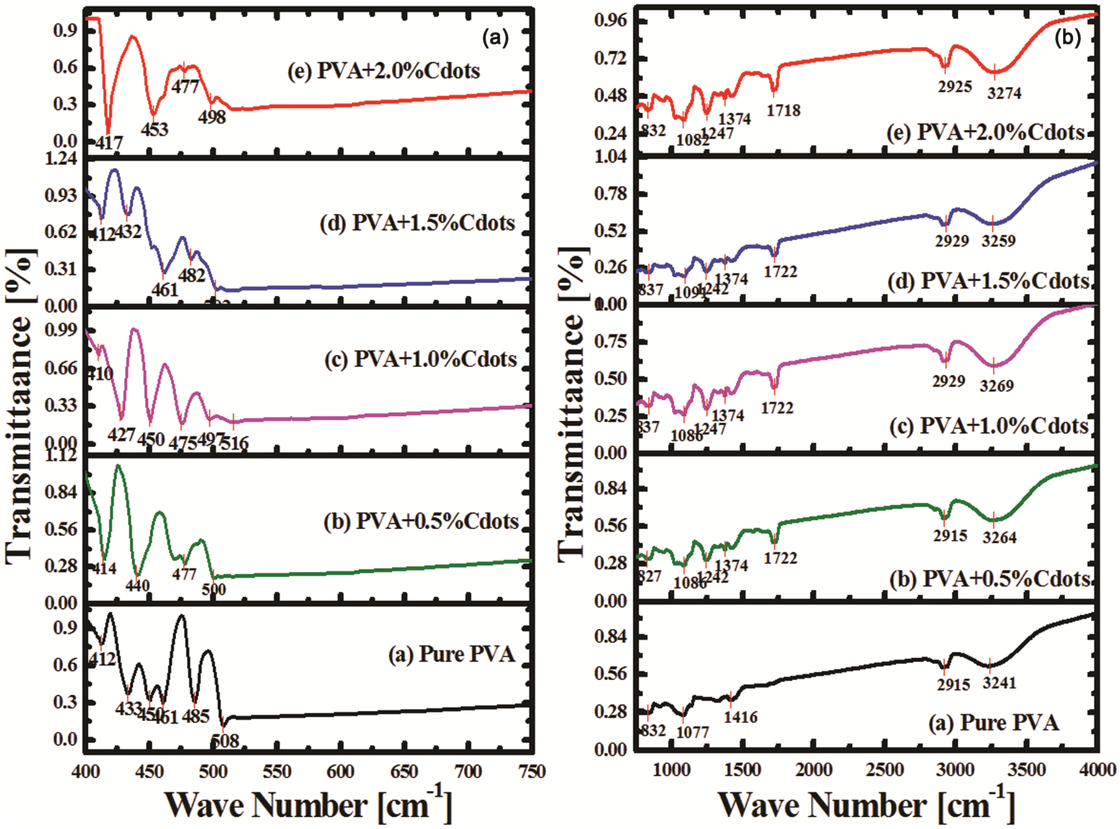

Fig. 3 — FTIR spectra for C-dot nanofiller PVA composite films from wavelength (a) 400 to 750cm^{-1} (b) 750 to 4000cm^{-1}

X-Ray diffraction analysis

XRD patterns of the pristine PVA and composite PVA–C-dot nanofiller films having 0.5, 1, 1.5 and 2wt% are presented individually in Fig. 4. The nanofiller composite films of 0.19-0.21mm thickness were mounted on a sample holder, and the X-Ray patterns were recorded from 0 to 60° at a speed of 8° per min. C-dot nanofiller composite films exhibited a peak at around $2\theta = 19.4^{\circ}$ that corresponds to (101) crystal plane for pure PVA and carbon that shows the presence of the semi-crystalline nature of PVA. The intermolecular interaction is strong between PVA networks. XRD patterns of the C-dot nanofiller composite filmsdisplayedno enlargement of the peak at $2\theta = 19.4^0$ or shifting. Meanwhile, PVA encloses the highest number of hydroxyl (O-H) groups that can successfully prevent theaccumulationof C-dot nanofillers and support to have the nanofillers well distributed at the nanoscale level in the aqueous solution PVA.

Mechanical characterization

Stress/strain relationship of PVA nanofiller composite films

Figure 5 represents the stress/strain relation for both pristine PVA and nanofiller composite films. Stress/strain values are tabulated in Table 1 and presented in Fig. 5. From the figure, it can be inferred that higher levels of stress are achieved by increasing C-dot fillers content compared to that of pristine PVA. From the results, it is observed that C-dot nanoparticles had properly reinforced in PVA matrix. The plots of stress versus strain show that the increase in the percentage of elongation at break the C-dot nanoparticle filler PVA composite films in PVA. The addition of C-dot nanomaterial enhances the interaction of molecules in the PVA host matrix. Enhanced bonding occurred between the hydroxyl groups and PVA–C-dot nanofiller composites, and higher filler content loading, which leading to the elongation at break values and also at ultimate stress values.

Effect of filler concentration on the elastic modulus

The effect of C-dot filler concentration on elastic modulus is presented in Table 1. From Table 1, the effect of filler concentration on elastic modulus is found to be linear. However, a slight deviation in linearity is observed at higher filler concentrations (1.5 and 2.0wt%). Elastic modulus values for all the weight percentages of filler concentration are presented in Table 1. This shows that the elastic modulus increased with an increase in the filler content, and the highest

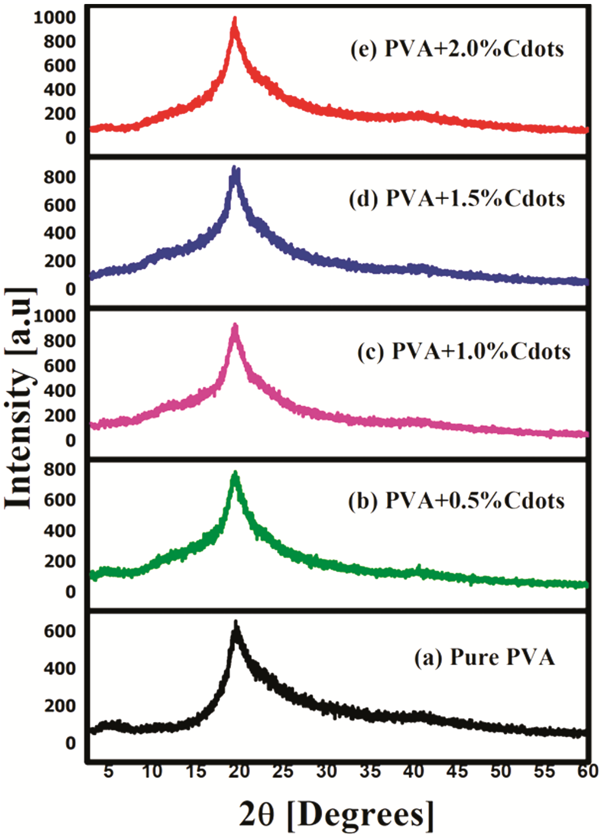

Fig. 4 — XRD profiles for a) 0wt % b) 0.5wt% c) 1wt% d) 1.5wt% e) 2wt% C-dot nanofiller loaded PVA composites

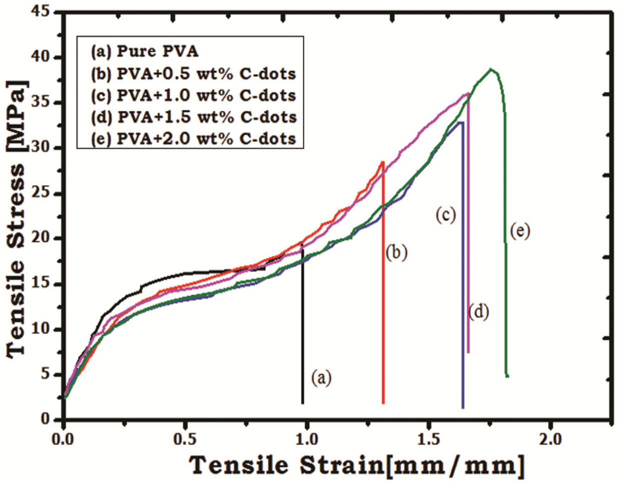

Fig. 5 — Stress-Strain diagram for (a) pristine PVA (b) 0.5wt% (c) 1wt% (d) 1.5wt% (e) 2wt% of C-dot nanofiller loaded in PVA matrix.

value of 37 MPa was found to be at 2wt% C-dot content. The strong interfacial strength between the nanofiller and PVA matrix improved the stiffness and also enabled to transfer the load towards the filler polymer nanoparticle interface.

Table1— Mechanical properties of PVA with different concentration of C-dots.

C-dot content (wt%)	Elastic modulus (Mpa)	Increase in elastic modulus (%)	Toughness (Mpa)	Increase in toughness (%)	Tensile strength (Mpa)
0	19.01	0	14.21	0	19.61
0.5	24.10	26.77	21.20	49.19	28.44
1	28.97	52.39	30.04	111.40	32.80
1.5	36.00	89.37	31.65	122.73	36.11
2	37.65	98.05	34.84	145.18	38.74

Tensile strength

The variation in ultimate tensile strength in C-dot nanofiller PVA composites is shown in Table 1. There is a significant increase in the ultimate tensile strength with increase in filler loading, nearly 300% increase in tensile strength for 2wt% PVA-C-dot nanocomposites when compared to pristine PVA. The strong reinforcement between nanofiller and PVA matrix is due to effective load transfer from filler to the polymer matrix interphase.

Toughness properties

The toughness is defined as the area under a stress-strain plot, which is the measure of the energy required for breaking the films. A significant improvement in the toughness for nanofiller composite films was observed, which is tabulated in Table 1. The increase in toughness for 2wt% PVA-C-dot-film was much higher than pristine PVA. This increase in toughness may be attributed to the change in structure induced by the addition of nanofiller as well as the increase in mobility of filler. It may be noted that the maximum toughness was recorded for 2.0 wt% C-dot filler containing composite film.

Increasing the C-dot loading in PVA–C-dot nanocomposites, the tensile strength, toughness and elastic modulus are increased. Good mechanical properties are observed at a higher concentration (2wt%). Increasing the C-dot concentration, result in a significant shift in peaks for C-H bending and C-O-C stretching as shown in Figure 3, which signifies very a strong interaction between the hydroxyl(O-H) group of PVA–C-dot nanocomposites. The similar observation is made from the estimation of mechanical properties, i.e. increase in C-dot concentration increases the mechanical properties.

Electrical characterization

Dielectric permittivity

The permittivity of the nanocomposites films was determined using dielectric polarization. These polarizations are associated with C-dot nanofiller and PVA matrix and interfacial polarizations at interfaces of filler and PVA matrix. Films of the synthesized nanocomposite will have a large volume fraction. Interfaces, where polarization process occurs, depends on the reaction kinetics. Many researchers have estimated the dielectric properties of the polymer composites[8,9,22,23] and observed that when oxides are used as fillers in the composites, exhibits lower permittivity values[24,25].

The complex permittivity is represented in the following equation (1):

$$\varepsilon''(\omega) = \varepsilon'(\omega) - i\varepsilon''(\omega) \qquad \ldots (1)$$

Where ε' (ω) and ε'' (ω) are the real permittivity and imaginary permittivity with respect to the energy storage and energy loss inthe electric field.

From the capacitance (C), the dielectric constant ε' (ω) was calculated from the following equation (2):

$$\varepsilon'(\omega) = \frac{C \times d}{A \times \varepsilon_o} \qquad \ldots (2)$$

where*d* is the C-dot filler polymer composites film thickness (in m), A is the surface area of the composite film (in m^2), ε_o is the air permittivity = 8.854×10^{-12} F/m.

The following equation (3) was used to estimate the dielectric loss:

$$\varepsilon''(\omega) = \frac{\sigma_{ac}}{\omega \times \varepsilon_O} \qquad \ldots (3)$$

where σ_{ac} and ω the AC conductivity (in S/m) and the angular frequency (ω = 2πf; radians/s), respectively.

In Fig. 6, it can be observed the dielectric constant,ε' (ω), for C-dot PVA polymer composites decreased with an increase in the frequency. The decrease in ε' (ω) may be attributed to the presence of the dipoles in the sample, which rearranges themselves in the direction of the applied field[26]. The decrease in the $\varepsilon'(\omega)$ is insignificant compared to the lower frequency which may be attributed to difficultly in self-orientation of $\varepsilon'(\omega)$ at higher frequencies. The dielectric permittivity of material depends on size, density, and dipole number, and material interaction. The higher values of $\varepsilon'(\omega)$ at lower frequency are owing to the interfacial and electrode[23]. When polar materials used as filler in PVA resulted in higher values of $\varepsilon'(\omega)$, which tend to decrease with an increase in the AC field.The inherent permittivity of C-dot nanoparticles also decreases with an increase in the frequency of the applied field[26,27]. The decrease in permittivity of C-dot nanoparticles and PVA resulted in a decrease of the dielectric constant of the composites by increasing the frequency of the applied field.

AC conductivity

The AC conductivity(σ_{ac}) of the polymer that arises from the motion of charge carriers is measured using the equation(4)[28,29]:

$$\sigma_{ac} = \omega \times \varepsilon_O \times \varepsilon' \times \tan\delta \qquad \ldots (4)$$

whereε_o is an air dielectric permittivity,ω is the angular frequency, tan δ is tangent delta and ε' is the dielectric constant. From Fig. 7, it can be observed that the highest conductivity was 7×10^{-5} Sm^{-1} for

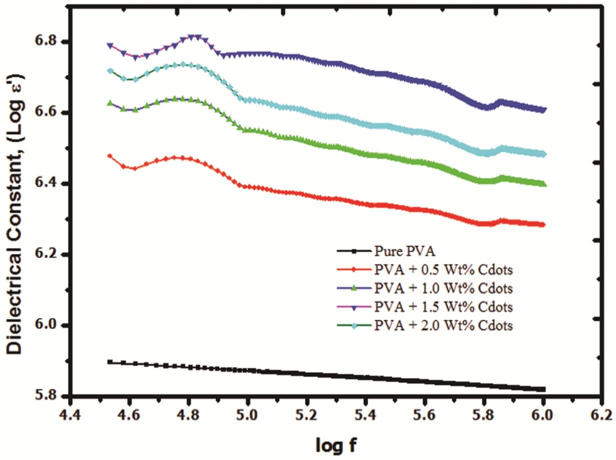

Fig. 6 — Dielectric constant of PVA–C-dot nanocomposites as a function of log frequency

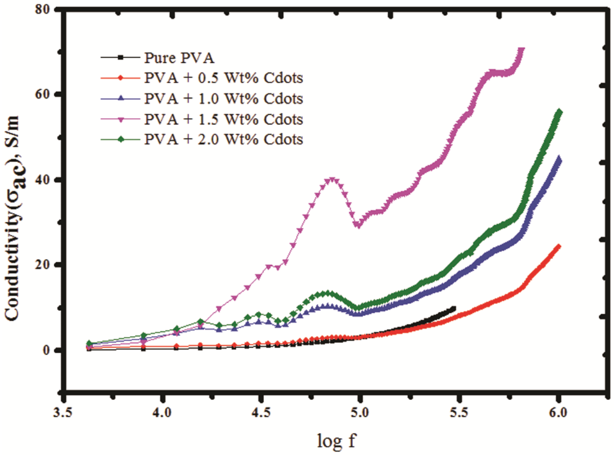

Fig. 7 — AC Conductivity of C-dot nanofiller PVA composite films as a function of log frequency

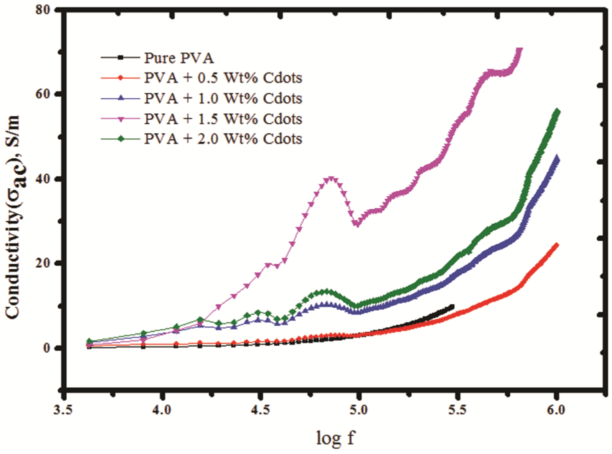

Fig. 8 — Dielectric loss of C-dot nanofiller PVA composite films as a function of log frequency

1.5% C-dot composites. At low frequency, there is negligible change in conductivity. Up to 5000 Hz frequency, the AC conductivity (σ_{ac}) is the same for all nanofiller loading. A sharp increase in the values of conductivity at a higher frequency may be attributed to the electronic interaction within the composites as resulted in conductive material at a higher frequency.

Dielectric loss

Variation of dielectric loss (ε'') with frequency for PVA–C-dot composites is presented in Fig. 8. This can be described on the basis of the ε'', the decrease in the dielectric loss with frequency can be related to a delay in a quick rotation of dipoles, leading to a delay between the applied electric field and the frequency of oscillating dipole. At lower frequencies the dielectric loss(ε'') is large because of free charge motion inside the material. On the basis of the Maxwell–Wagner interfacial model, the dielectric medium contains double-layers with fine conducting particles, which are divided by the conducting particle boundaries. By applying the external electric field, the charge transporters can be easily migrated with particles which were later gathered at particle limits, result in great dielectric constant and significant polarization. The large dielectric constant value due to the non-uniformity of dielectric structure (this is due to particle structure and porosity). The dielectric properties of PVA–C-dot nanocomposites like dielectric losses decrease with an increase in the frequency. That indicates that beyond a definite frequency of externally applied field and the hopping between metal ions cannot pursue alternating field[30-32].

Conclusion

C-dot filler polyvinyl alcohol composite films were synthesized using the casting method. The C-dot filler concentration was varied from 0 to 2 weight% with increments of 0.5 weight%. The structural properties and interactions of C-dot nanofiller with PVA were studied using X-Ray diffraction and FTIR. Mechanical properties such as toughness, elastic modulus, % elongation, and tensile strength, and observed to have significant improvements with reinforcement of C-dot nanomaterials in PVA matrix. The enhanced mechanical properties were found at 2.0wt% filler loading. Almost 300% enhancement in ultimate tensile strength was observed in comparison with the pristine PVA. 2.0wt% of C-dotfiller loading had shown a 275% enhancement in toughness compared to filler loading. From the results, we can conclude that the enhanced mechanical, structural and electrical properties of PVA–C-dot nanofiller composites are the promising candidates for nanotechnology in future.

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