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Research Article

Influence Of Polarization and Temperature on The Dielectric Properties of ZnO/Polypyrrole Nanocomposite Films

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Abstract

The dielectric properties of zinc oxide (ZnO) doped polypyrrole (PPy) nanocomposites were investigated with emphasis on the effect of thermal polarisation on dielectric constant behaviour. ZnO nanoparticles with an average size of 41.37 nm were synthesised by a precipitation method. The FWHM were obtained from the peaks and the size of the nano particles were calculated by the Debye-Scherrer formula, and the JCPDS data gives the hexagonal nanostructure.PPy was prepared through chemical oxidative polymerisation. ZnO/PPy nanocomposite films were cast by dispersing the constituents in a polyvinyl alcohol matrix. Dielectric measurements were carried out on unpolarized and thermally polarised films at 60°C. The results reveal that thermal polarisation markedly enhances the dielectric constant by improving dipole alignment and reducing structural disorder. An optimal ZnO concentration of 3% and 4% yielded maximum dielectric enhancement, whereas higher loading caused interfacial trapping and reduced dielectric performance.

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INTRODUCTION

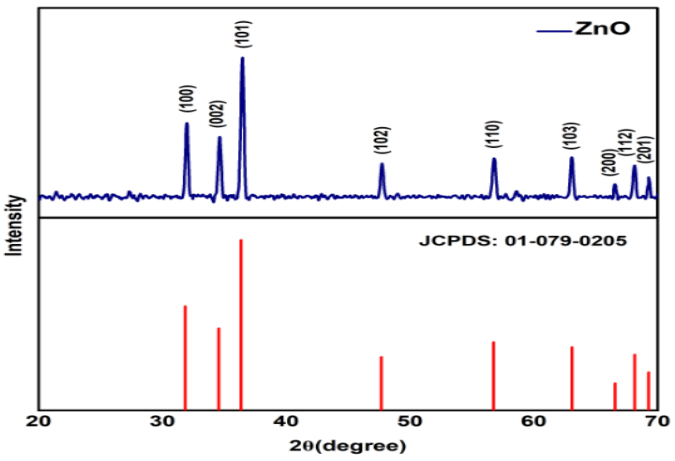
The utilisation of conductive nanocomposites consisting of polymer has received significant attention in the past several years since the conductive nanocomposites possess adjustable electrical, dielectric and mechanical properties applicable in sensors, energy storage, electromagnetic shielding and bendable electronics [4,7]. Among the conducting polymers, Polypyrrole (PPy), which is a high electrical conductivity, stability in the environment, as well as the ease with which it can be produced and the relatively low cost [3,6]. However, clean PPy is generally susceptible to such failures as low mechanical functionality, low thermal stability and reduced dielectric efficiency at high temperatures, which restrict its practical use [15].

To overcome these drawbacks, the inorganic semiconductor nanoparticles have been entrained into conducting polymer matrices to form polymer-nanoparticle composites, which have better multifunctional properties [2,9]. Zinc oxide (ZnO) is one of the best studied examples of metal oxide semiconductors with a large band gap (3.37 eV), a large exciton binding energy,

non-toxic, and with good thermal and chemical stability [13, 20]. It has also been discovered that the incorporation of ZnO nanoparticles in PPy matrices can improve the charge transport, dielectric response and thermal stability by interfacial polarisation and charge carrier mobility [17,14].

The degree of concentration of fillers, the particle size, the interfaces between the filler and the polymer, the temperatures and the polarisation conditions are found to strongly determine the strong dependence of the dielectric behaviour of the polymer nanocomposites [10]. The most common interfacial (Maxwell-Wagner-Sillars) polarisation occurs in heterogeneous materials, e.g. ZnO/PPy composites, especially in low frequencies due to the concentration of charges at the interface of the polymer filler [11,19]. This is further enhanced by the availability of fillers in nanoscale, which add to the interfacial area and hence the dielectric constant with loss properties as well as significantly changed [4].

Temperature is also another parameter that defines electrical and dielectric properties in conducting polymer composites. Homogeneous temperature enhances charge carrier hopping, dipole orientation and segmental movement of polymer chains and results in the change of conductivity, as well as dielectric relaxation behaviour [12]. It has been established that thermal polarization schemes can increase the alignment of dipoles and lower structural disorder, leading to reduced activation energy for charge transportation and increased dielectric performance [8].



XRD Plot and JCPDS of Zinc Oxide Nanoparticles

Sulphate impurities were then removed by extensive washing of the gel using distilled water following the ageing process. The rest was then put on a Petri dish evenly and left to dry.

The dried (annealed) sample was exposed to a temperature of less than 150℃ in a muffle furnace. ZnO fine powder was received after allowing it to cool to room temperature [20].

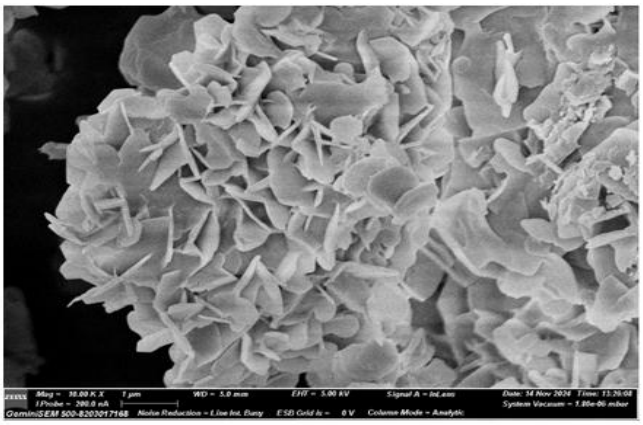
The crystallite size of the particle was calculated as per the Debye-Scherrer equation by the X-ray diffraction (XRD) analysis of ZnO nanoparticles [13].

Though several studies have already been done on the PPy-based nanocomposites, there was no systematic investigation on the interactive influence of temperature and thermal polarisation on the dielectric characteristic of the ZnO-doped PPy films [16]. In particular, one needs to be aware of the optimum filler content and polarisation parameters to attain the required materials to be used on a specific electronic application [14]. The present paper fills this gap as the behaviour of the two-film consisting of ZnO/PPy, both in dielectric behaviour, electrical behaviour, the unpolarized state and the thermally polarised state are explored, especially the relationship between charge conduction at different temperatures.

Experimental Procedure:

Synthesis of Zinc Oxide (ZnO) Nanoparticles

Zinc oxide (ZnO) nanoparticles were synthesised by a simple chemical precipitation method [20]. The 2.0g of sodium hydroxide (NaOH) in 40mL of distilled water and 10g of zinc sulphate ($ZnSO_4$) in 120mL of distilled water were dissolved separately. The magnetic stirrer was turned on so that there was continuous stirring of the mixture of the two solutions for 1h. The pH of the solution was maintained at approximately 7.5. The obtained gel was allowed to age and it turned out 48h.



FESEM Image of Zinc Oxide

$$D = \frac{K\lambda}{\beta cos\theta}$$

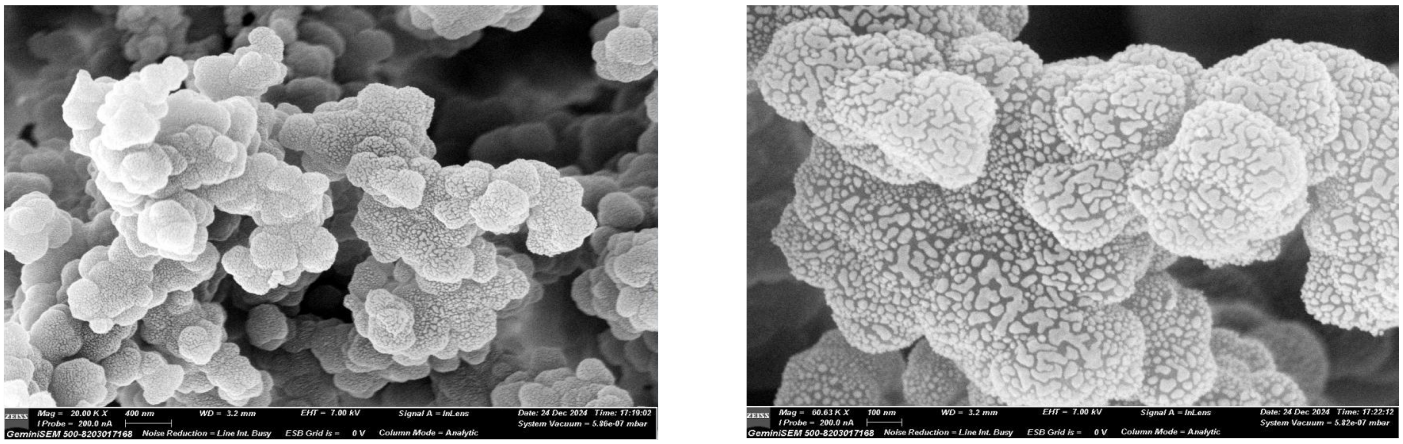
where D is the crystallite size, K is the shape factor, λ is the wavelength of radiation, β is the full width at half maximum (FWHM) of the diffraction peak, and θ is the Bragg angle. The average crystallite size was calculated to be 41.37 nm for the (002) plane with a d-spacing of 2.5938 Å and lattice parameter.

$$a = 3.2417 \text{ Å}.$$

Synthesis of Polypyrrole (PPy):

Polypyrrole (PPy) synthesis was carried out under the chemical oxidative polymerisation method in the presence of ammonium persulfate (APS) as an oxidiser [6]. 1.0 mL of pyrrole monomer was dissolved in 70 mL of 1.5M solution of hydrochloric acid (HCl). The 2.04 g of APS in 20 mL of the deionised water was dissolved in individual test tubes.

The pyrrole solution, which was constantly stirred at room temperature, was added with drops of the APS solution. The reaction was left to stirred 5h, until it was completely polymerised [18]. The precipitated black was filtered and washed to remove any unreacted monomer and oxidant by bathing in ethanol and deionised water, and dried in a hot air oven at 60℃ of 12h.



FESEM Image of Polypyrrole(PPy)

Casting of ZnO/Polypyrrole Nanocomposite Films:

These nanocomposite films of ZnO/Polypyrrole (ZnO/Ppy) were prepared by the casting technique [16]. The desired quantity of ZnO/PPy nanocomposite powder was synthesised and dispersed in an appropriate amount of polyvinyl alcohol (PVA) and left to mix under a magnetic stirred with 5 h. Once again, the solution was ultrasonicated (30 min) to ensure that the nanoparticles of zinc oxide are evenly distributed within the polymer matrix.

This was then poured on a clean and level glass surface and allowed to spread. The drying was at room temperature, whereby the films were dried to remove the moisture and subsequently dried at 60℃ in a hot air oven, several hours to remove the solvent residues. The free-standing film of ZnO/PPy nanocomposites was dried and peeled off the substrate.

The thickness of the films was measured using a digital micrometre screw gauge, and the films were used for dielectric and electrical characterisation.

Measurement of Dielectric Constant:

The dielectrics of the nanocomposite films of ZnO/PPy were measured using an LCR meter. The cast samples were turned into circular samples with equal thickness. Silver paste was then applied to the films on the top and the bottom to form an electrode plate capacitor arrangement (parallel). The dielectric constant (ε′) was calculated using the relation:

$$\varepsilon' = \frac{(C \times d)}{(\varepsilon_0 \times A)}$$

where C is the capacitance found, d is the thickness of the film, A is the electrode area, and ε_0 is the permittivity of free space. To study the frequency-dependent dielectric behaviour of the films of the ZnO/PPy nanocomposites, the measurements were carried out at the frequency of various frequencies at the room temperature.

RESULT AND DISCUSSION

Materials have four polarisation mechanisms, and they include electronic, ionic, orientation and interfacial polarisation. ZnO has all four mechanisms that do not play any significant role in terms of the contribution it gives to the overall product, but the frequency of operation. A combination of these mechanisms at low frequencies gives a large dielectric constant, and this portion of the dielectric constant is gradually decreasing as the frequency increases. Temperature too is important, as the temperature rises, frozen dipoles are liberated and raise permittivity and overall polarisation.

A capacitor is formed when a dielectric material is located between two conductive plates and is exposed to alternate electric field. The capacitance is given by

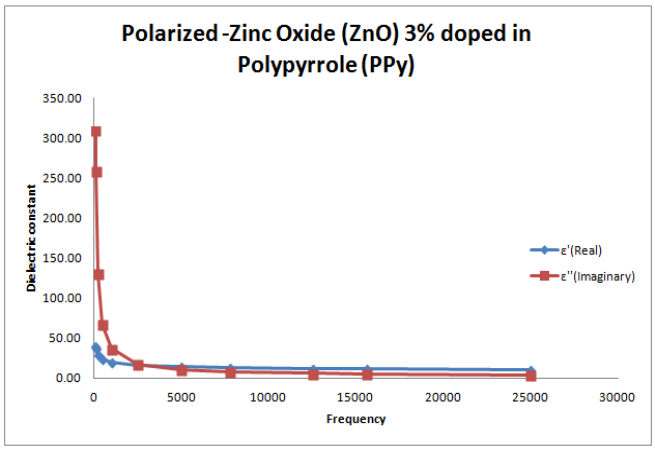
$$C = \frac{(\varepsilon_0 \varepsilon_r A)}{d}$$

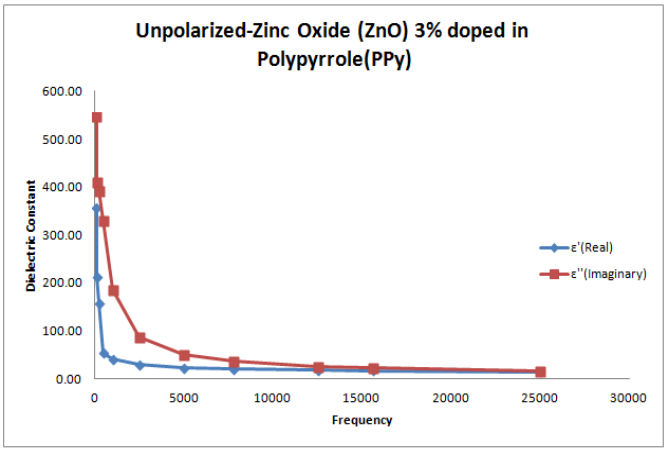
Capacitor (C) is based on four factors: area of the electrodes A, gap between the electrodes d, and permittivity of free space. $\varepsilon_0 = 8.854 \times 10^{-14}$ F/cm, and the relative permittivity or dielectric constant of the material ε_r . All the dipoles within the dielectric material get aligned in the same direction as those of the applied electric field. The alignment of dipoles give lead to an induced electric field which is in the opposite direction to the utilized electric field. The method of alignment of dipoles is called polarisation, as a result of which the material can store charge in it.

The ratio of absolute permittivity (ε) of the material and permittivity of free space (ε_0) is called relative permittivity (ε_r) or dielectric constant (k), which is given as:

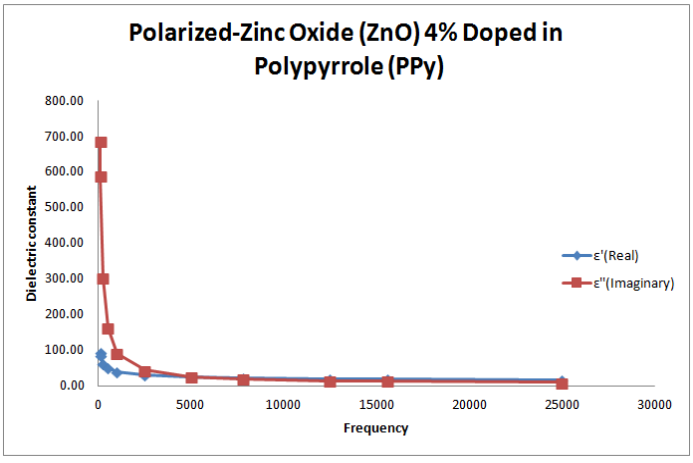
$$\varepsilon_r = \frac{\varepsilon}{\varepsilon_0}$$

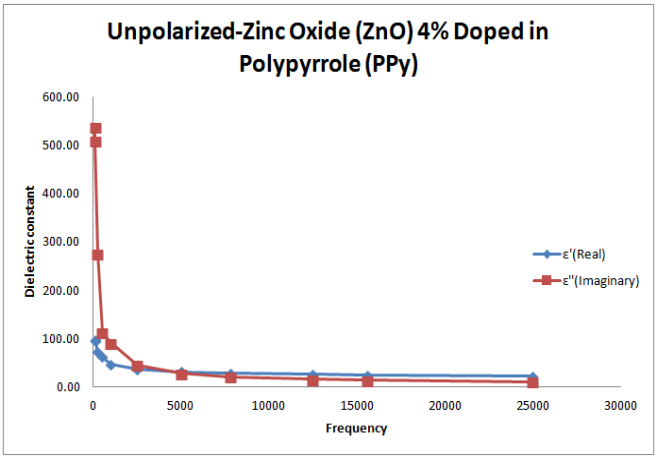
When a thick film of dielectric material is subjected to AC voltage, depending upon the statictaticε_r of the material, it permits or allows the electric charge storage phenomena. The electric energy that does not get absorbed in the material and is dissipated in the form of heat is called dielectric loss.





According to the graphs, polarised and unpolarized action of the 3% ZnO doped polypyrrole (PPy) exhibit a distinct difference in the dielectric response in the two states. The tendencies of both the real (ε') and imaginary (ε''The components of the dielectric constant of the two samples are similar in that there is a strong tendency at higher frequencies, which is characteristic of dipolar relaxation and Maxwell-Wagner interfacial polarisation. However, this decrease is compounded by the state of polarisation. The sample of the unpolarizing sample exhibits very high values of dielectric constant of 500 and below at low frequencies, suggesting that there is a lot of dielectric loss and a high concentration of charge carriers at the interface. This is the sign of an extremely messy system where trapped charges will assist in the loss of energy. On the contrary, the polarised sample has significantly smaller. ε' and ε'' values where the latter is highest at very low frequencies of about 320. Polarisation also enhances dipole alignment as well as charge carrier mobility, reducing interfacial charge trapping and dielectric loss. The two samples exhibit a low and constant dielectric at frequencies above 5000 Hz, that is, they exhibit low dipole response. Overall, polarisation enhances the dielectric stability of ZnO/PPy nanocomposites.





Since 4% ZnO-doped polypyrrole (PPy) exhibits dielectric characteristics in the polarised and unpolarized state, it is seen that frequency and polarisation treatment strongly affect the material. In both cases, the actual (ε') and imaginary (ε'') parts of the dielectric constant are the largest when we are at the lower frequencies and later diminish swiftly as we advance in frequency, and lastly they tend to take up nearly constant values as we proceed in higher frequency. This is typical of dielectric relaxation, which is space-charge and interfacial polarisation dominated at low frequencies. The unpolarized sample exhibits large ε′ and ε″ values, with ε″ exceeding 550 at low frequencies, indicating high dielectric loss and significant charge trapping at interfaces. On the other hand, polarized sample exhibits even greater values of dielectric with ε' approaching a value of 650,

showing the enhancement of alignment of dipoles and polarisation of an interface following a polarisation treatment. Although the samples tend to have similar low dielectric values at high frequencies, the polarised sample has low values of dielectric losses at high frequencies. Overall, ZnO/PPy nanocomposites can be improved in terms of storing and maintaining polarisation in terms of dielectric.

At Temperature 333K

Freq (Hz)	3%Polarized Real (ϵ′)	3%Polarized Imaginary (ϵ″)	3%Unpolarized Real (ϵ′)	3%Unpolarized Imaginary (ϵ″)	4% Polarized Real (ϵ′)	4% Polarized Imaginary (ϵ″)	4% Unpolarized Real (ϵ′)	4% Unpolarized Imaginary (ϵ″)
100	38.88	309.08	356.15	547.24	93.79	685.54	97.66	537.41
120	36.22	258.04	213.21	409.65	85.68	588.05	96.54	508.51
250	28.63	130.77	158.41	392.23	63.65	301.26	73.52	276.31
500	23.53	67.46	53.76	330.48	49.96	162.71	62.23	112.19
1000	19.78	36.65	40.7	186.67	39.93	89.93	47.92	91.47
2500	16.07	17.39	28.89	86.44	30.22	43.4	37.62	45.86
5000	13.95	10.4	23.11	49.86	24.99	25.81	31.91	28.33
7800	12.82	7.67	20.24	35.87	22.33	18.92	28.99	21.37
12500	11.81	5.69	17.72	25.54	19.97	13.93	26.39	16.19
15600	11.38	5.01	16.69	22.15	19.02	12.2	25.38	14.4
25000	10.58	3.8	14.77	15.33	17.21	9.16	23.31	11.18

The frequency-dependent dielectric constant (real ε' and imaginary ε'') of ZnO-doped polypyrrole (PPy) in the polarities of 333 K and 4% of polarised and unpolarized types are displayed in this table". The values of ε' and ε'' are very large at low frequencies (100-250Hz) and decrease and decrease respectively at the high frequencies, and at the very high frequencies ~ 5000Hz are small and nearly constant. This is characteristic of the Maxwell-Wagner effects of interfacial polarisation and space-charge effects at low frequencies, and dipolar relaxation at high frequencies.

Unpolarized samples possess by far the highest values of ε ′ and ε ″ as opposed to polarised, especially at low frequency, and consequently, the dielectric loss is large due to the build-up and trapping of charges at interfaces. The 4% ZnO doped PPy possesses higher dielectric constants as compared to the 3%, which means that it has more interfacial polarisation when there is more filler. This is the effect of polarisation treatment, which reduces ε' and ε'' and suggests superior polarisation of the dipole, a reduced trapping of space charges and reduced dielectric loss. Overall, polarisation enhances dielectric stability, but the percentage of ZnO enhances dielectric performance at low frequencies.

CONCLUSION

The dielectric studies of ZnO-doped polypyrrole nanocomposites reveal strong frequency- and polarization-dependent behavior. High dielectric constants at low frequencies arise from Maxwell–Wagner interfacial polarisation and space-charge effects, while values stabilise at higher frequencies due to limited dipole response. Increasing ZnO content enhances interfacial polarisation and dielectric. activity. Polarisation treatment significantly improves dielectric stability by promoting better dipole alignment, reducing charge trapping and lowering dielectric loss. Overall, polarised ZnO/PPy nanocomposites exhibit improved dielectric performance, making them promising candidates for energy storage and electronic applications.

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