*Review Article*

# Dielectric Properties of ZnO-Based Nanocomposites and Their Potential Applications

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Energy storage devices constitute one of the research areas in recent years. Capacitors are commonly used for the storage of electrical energy. The current research is focusing on not only the improvement in energy density but also the materials which are environment friendly. Polymer composites are known to be technically essential materials owing to their wide range of applications. Enormous research has been devoted to zinc oxide- (ZnO-) based polymer nanocomposites, due to their extraordinary dielectric properties. This review article presents a detailed study of the dielectric properties of ZnO-based nanocomposites. The dielectric constant study includes the effect of transition metals and rare earth metals as a dopant in ZnO. This review gives an insight into the mechanism responsible for the variation of dielectric constant in ZnO nanocomposites due to various factors like size of nanoparticles, thickness of the thin film, operating frequency, doping concentration, and atomic number. The observations have been summarized to convey the mechanism and structural changes involved in the ZnO nanocomposites to the researchers. The deployment of biodegradable nanocomposite materials is expected to open an innovative way for their outstanding electronic applications as storage materials.

## 1. Introduction

The dielectric properties of material define its characteristic role in electronics. To study the dielectric properties of any material, dielectric constant ($k$) is a crucial factor that determines the charge storage capability of the material [1]. High-$k$ materials are used as gate dielectrics in MOS transistors, memory cells, capacitors, supercapacitors [2, 3], and so forth, whereas low-$k$ materials find their applications in electrical insulation and high-speed integrated circuits [4]. The semiconductor industry is greatly influenced by dielectric materials and emerges out as a revolutionary stream of microelectronics [5]. Metal oxide dielectric materials are considered to be a key element for diverse thin-film electronic systems owing to their superior dielectric and mechanical properties [6]. Titanium dioxide ($TiO_2$) in rutile form exhibits a high dielectric constant and can be used as a filler in hybrid (i.e., organic-inorganic) composites for application in modern electronics [7, 8]. Tin dioxide ($SnO_2$) is an n-type semiconductor possessing a wide direct bandgap of 3.37 eV at room temperature [9]. Due to low electrical resistivity, high chemical, mechanical, and thermal stability [10], it is very useful for applications in spintronics [11], lithium-ion batteries (LIBs), and supercapacitors [12, 13]. Due to its high dielectric constant [14], $SnO_2$ can be used for applications in thin-film electrodes, sensors, batteries, and solar cells [15].

ZnO is a semiconductor of groups II-VI. It is actively used due to its very attractive properties and nontoxic nature. Among metal oxides, nanostructured zinc oxide (ZnO) exhibits exceptional electronic, optical, and electrochemical properties. In addition to this, the wide bandgap (3.37 eV) and massive exciton binding energy (60 meV) permit the material to operate at much higher voltage, frequency, and

temperature than conventional semiconductors [16]. Moreover, ZnO-based composites have shown up potential applications in the field of industrial electronic devices like transducers, electrochemical sensors, solar cells, supercapacitors, and lithium-ion batteries [17–19]. ZnO is the most widely used inorganic compound because of its unique electronic, physical, and chemical properties [20]. This review article deals with the fundamental aspects behind the dielectrics along with the effective role of particle size and metal doping in ZnO to study its dielectric behaviour. Furthermore, the relative effect of transition metal and rare-earth-metal are also discussed in detail towards the dielectric origin in ZnO. Lastly, ZnO doped biodegradable polymer nanocomposites and their applications have been discussed thoroughly in this study.

## 2. Dielectric Materials

Dielectric materials are insulating materials. However, dielectric materials have some additional properties as compared to insulators. As they do not allow the flow of charge through them, they permit exerting electrostatic field and hence storing charge [21]. The thin films made up of dielectric materials extend enormous applications in the electronic industry like barrier layers, gate oxide, varactor dielectrics, and electrical isolation between conductive regions [22]. Numerous transition metal oxides demonstrate colossal dielectric constant and therefore have enormous potential for applications in modern microelectronics for the development of new capacitors for energy storage [23]. Lunkenheimer et al. observed a larger dielectric constant in various transition metal oxide materials, for example, $CaCu_3Ti_4O_{12}$ has a $k$ value of 17000 and $La_2SrNiO_4$ has a $k$ value of 10000 for their application in capacitive circuits [23]. Meher et al. suggested that $Sr_2TiMnO_6$ can be used as dielectric material in capacitors, due to its large dielectric constant of the order of $10^5$ [24]. The value of $k$ is larger due to the high degree of polarization.

*2.1. Polarization.* When an atom is subjected to an electric field, it gives rise to the formation of a dipole [25]. If the dielectric material is subjected to an alternating field by placing it between two conducting plates acting as electrodes, it gives rise to the formation of a capacitor [26, 27] as shown in Figure 1. According to the equation of the capacitor,

$$C = \frac{(\varepsilon_0 \varepsilon_r A)}{d}. \tag{1}$$

Capacitance ($C$) depends upon four factors: area of the electrodes $A$, spacing between the electrodes $d$, permittivity of free space $\varepsilon_0 = 8.854 \times 10^{-14}$ F/cm, and relative permittivity or dielectric constant of the material $\varepsilon_r$.

All the dipoles within the dielectric material get aligned in the same direction as those of the applied electric field. The alignment of dipoles gives rise to an induced electric field which is in a reverse direction to the applied electric field
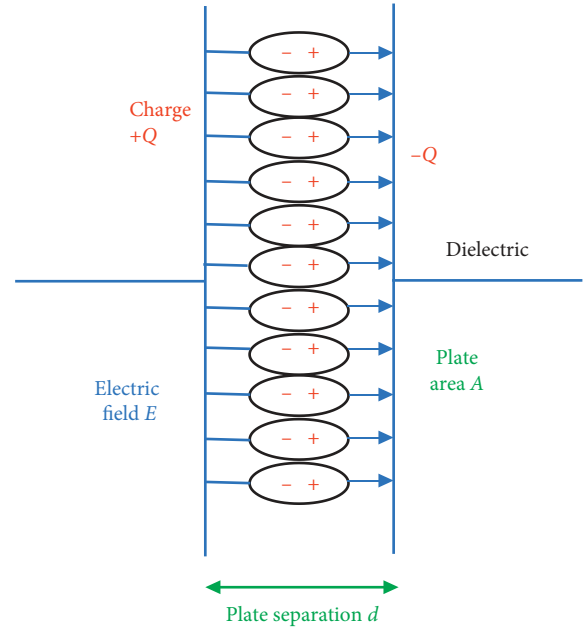


FIGURE 1: Structure of the dielectric capacitor.

[28, 29]. This process of alignment of dipoles is called polarization, due to which the material can store charge in it [30].

*2.2. Source of Polarization.* There are several aspects that give rise to polarization in the materials. Polarization is responsible for the relative permittivity of a material, subjected to an electric field [31]. It is originated from four different roots, namely, electronic, ionic, orientation, and interfacial polarization. Polarization is a frequency-dependent phenomenon [25, 32] as shown in Figure 2.

In a material, there may be single or multiple polarization sources active at a time. In a material having high relative permittivity, a greater number of sources or all these sources are active [31]. On the other hand, a few will be active in a material having low relative permittivity. In ZnO, all the four types of polarizations exist but the active one is dependent on the frequency of operation [33–37] The origin of high $k$ in a material at low frequencies is the presence of all four sources of polarization, and it starts decreasing with the increase in frequency since dipoles in the material cannot follow the high frequency of applied field [38]. The value of $k$ in a material increases with the increase in temperature which is attributed to the fact that a greater number of frozen dipoles are liberated due to thermal energy [39].

*2.3. Dielectric Constant and Dielectric Loss.* The ratio of absolute permittivity ($\varepsilon$) of the material and permittivity of free space ($\varepsilon_0$) is called relative permittivity ($\varepsilon_r$) or dielectric constant ($k$), which is represented as follows:

$$\varepsilon_r = \frac{\varepsilon}{\varepsilon_0}. \tag{2}$$

When a thick film of dielectric material is subjected to AC voltage, depending upon $\varepsilon_r$ of the material, it permits or
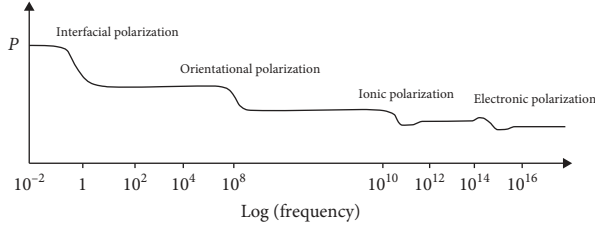
FIGURE 2: Frequency dependence of different dielectric polarizations.

allows the electric lines of force to get absorbed in it and gives rise to 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. The complex permittivity of a material, which is subjected to an alternating field, is defined by the Debye expression [40–42]:

$$\varepsilon^*(\omega) = \varepsilon_\infty + \left( \frac{\varepsilon_s - \varepsilon_\infty}{1 + J\omega\tau} \right), \tag{3}$$

where $\varepsilon_\infty$ = dielectric constant at high frequency, $\varepsilon_s$ = dielectric constant at low frequency, $\omega = 2\pi f$, $\tau$ = relaxation time, and $\varepsilon^*(\omega)$ is complex permittivity and is given by

$$\varepsilon^* = \varepsilon' - J\varepsilon'', \tag{4}$$

where $\varepsilon'$ = real part of permittivity and is given by

$$\varepsilon' = \varepsilon_\infty + \left( \frac{\varepsilon_s - \varepsilon_\infty}{1 + \omega^2\tau^2} \right), \tag{5}$$

which is also called the real part of the complex dielectric constant ($k$). $\varepsilon''$ = imaginary part of permittivity and is given by

$$\varepsilon'' = \left( \frac{(\varepsilon_s - \varepsilon_\infty)\omega\tau}{1 + \omega^2\tau^2} \right), \tag{6}$$

which is also called the imaginary part of the complex dielectric constant ($k$). $\varepsilon'$ and $\varepsilon''$ both are frequency-dependent.

The dielectric loss factor is given by

$$\tan\delta = \frac{\varepsilon''}{\varepsilon'}. \tag{8}$$

Metal oxides possess a high dielectric constant along with low dielectric loss, thus having huge potential to be used in microelectronics industry [22, 23].

## 3. Classification of Dielectric Materials

The dielectric constant ($k$) of a material is basically the charge storage capability of that material. Materials can be classified into two categories: high-$k$ materials and low-$k$ materials [5]. Materials that have a value of dielectric constant more than silicon nitride, $k > 7$, are called high-$k$ materials and the materials that have a dielectric constant value less than silicon dioxide, $k < 3.9$, are called low-$k$ materials. The lowest value of $k$ is of air, which is equal to

unity. And the highest value of $k$ is of relaxor ferroelectric, which is ~24000 at 1 KHz [4].

3.1. Applications of High-k and Low-k Materials. High-$k$ materials can be used to develop efficient memory cells for high storage memory chips [43], to develop passive components like capacitors and supercapacitors [44]. They can be used for gate dielectrics in MOS transistors and TFTs [45]. Low-$k$ materials find their applications in EMI Shielding [46], in the field of satellite communication, telecommunication in space [35], and high-speed IC packaging [47].

3.2. Demand for High-k Materials. Nowadays, electronic gadgets have become the fundamental need of daily life and with continuous thought of green evolution to minimize the use of paper, an efficient memory storage device plays a significant role [48]. These memory chips are made up of small capacitors and transistors as supporting circuitry [4]. The continuous efforts in the research and development in the field of material science make it possible to decrease the size of transistors and to accommodate a larger array on a single chip resulting in enhanced performance and storage. From equation (1), there are three approaches through which the storage capacity of a capacitor can be improved. First, the capacitance can be increased by increasing the area $A$, of capacitor's plates, but it will increase the device size also, which is not acceptable [26]. The second is to decrease the thickness of dielectric $d$ or gap between capacitor's plates [27]. But dielectric breakdown strength of a material imparts constrain over a certain limit on the dielectric thickness, beyond that the material will either breakdown or the leakage current will start to flow through it. Therefore, the only possibility to enhance the performance and storage capacity of transistors/capacitors is to increase the value of the dielectric constant or to use a high-$k$ material [28].

Thin-film transistors (TFTs) are the basic building block of every electronic gadget. TFTs are used in almost every flat panel display as a switch to drive the pixels. The resolution and clarity of the display depend upon the switching speed and performance of the TFTs [49–51]. The performance of a TFT is greatly influenced by the properties of its dielectric layer. The dielectric layer is responsible for the accumulation of charge in the channel and leakage current in the device [52, 53]. There is a fundamental limitation on the thickness of a silicon-based gate dielectric that the oxides having a thickness less than 2 nm suffer from direct tunnelling current or leakage current [54]. Therefore, high-$k$ materials are required to overcome this problem. Due to these reasons, high-$k$ materials are high in demand in the electronic industry and gain the attention of researchers [27]. Several materials like $Ta_2O_5$, $Nb_2O_5$, $Y_2O_3$, $TiO_2$, $Al_2O_3$, $HfO_2$, and $ZrO_2$ are available which have a value of dielectric constant varying from 10 to 100 [22, 55]. Water ($H_2O$) has $k = 80$ but it has very low dielectric strength. It cannot withstand high voltage. Hence, it cannot be used for practical applications [56]. So, there is a need to search new materials or synthesize new materials and tailor their properties as required. ZnO is one of the materials which is nontoxic and is safe for the

environment. It can offer greater dielectric constant after doping it with some other metals; hence, it can satisfy the need of energy storage devices.

## 4. Factors Affecting the Value of Dielectric Constant of a Material

Apart from frequency and temperature dependence, $k$ of a material is also affected by its structural properties or microstructure:

(1) Crystallite size: if the size of the crystal of a material is large, then the value of $k$ is also high. This is because a larger particle contains a greater number of dipoles which get aligned under the influence of the electric field and results in greater permittivity, thus a higher value of $k$ [57, 58].

(2) Porosity: if the material is porous or has low density, then the value of $k$ will be low [59]. A porous material is a dense material which is filled with random air pores. The value of permittivity or $k$ of the material will decrease because the relative permittivity of air is much lower than the material itself; hence, with the increase in porosity, the overall value of $k$ decreases [60].

(3) Composition and doping concentration: the composite materials consist of more than one material having different values of $k$. So, the resulting dielectric constant of the composite material will depend upon all the constituents. A dopant of low $k$ will reduce the overall effective value of $k$ of the composite material depending upon its percentage of concentration. A higher concentration of low-$k$ dopant will result in more dip in the overall effective value of $k$ of the material as compared to the lower concentration of dopant [61, 62].

(4) Defects: above room temperature, every material fundamentally has point defects to establish thermal equilibrium in the system:

(a) Surface defects: the surface of every material possesses inherent defects as it has a smaller number of bonds than the atoms in the bulk portion of the material. These loose bonds of the surface are called dangling bonds. If a material has more numbers of dangling bonds, that means it has a good density of charge carriers that will lead to large polarization [63].

(b) Vacancies: they exist in the system when there are some missing atoms in the crystal called vacancies. The existence of vacancies has a direct impact on the dielectric constant of the material [64]. The presence of vacancies in a material helps in trapping the charge carriers which generates large polarization, hence resulting in a high dielectric constant at low frequencies [65].

(c) Substitution defects: when some impurity atom or doping atom substitutes in the place of a host atom, it gives rise to substitution defects. These defects trap the charge carriers and produce large polarization, hence increasing the value of the dielectric constant at low frequencies [66].

(d) Grain boundaries: grain boundary is an interface that separates two grains in a polycrystalline material. It arises due to inhomogeneity in the surface of the material. If a material has a greater number of grain boundaries, then the value of $k$ becomes high in the range of frequency, where interfacial polarization is active, as grain boundaries trap the charge carriers at the interfaces and do not allow them to move. Hence, they enhance the polarization and ultimately give rise to an increased value of dielectric constant at low frequencies [67].

## 5. Zinc Oxide (ZnO)

Zinc oxide (ZnO) is an n-type semiconductor that belongs to the II-VI group of the periodic table. It has been extensively studied and analysed by numerous researchers due to its outstanding properties, which include cost-effectiveness, wide availability, and nontoxic nature [68–70]. ZnO exhibits a wide bandgap of 3.37 eV. It also possesses a high exciton binding energy of 60 meV, which permits the device to operate at much higher voltage, frequency, and temperature than conventional semiconductors [71]. It shows a great performance of photocatalytic activities for the degradation of organic pollutants present in water, due to its ability to absorb almost the entire UV spectrum [72–74]. ZnO exhibits high quantum efficiency due to greater order of electron mobility 200–300 $cm^2 \cdot V^{-1} \cdot s^{-1}$ [75–77]. It has already been discussed that the dielectric properties of a material depend upon its structure. ZnO has structural defects like oxygen vacancies and zinc interstitials which are intrinsic to the synthesis process [66]. Huang et al. demonstrated that the number of oxygen vacancies that exist in the ZnO lattice exhibit a great impact on the relative permittivity and hence its dielectric properties [64]. ZnO is a polar molecule, hence possessing a permanent dipole moment, due to whichthe electronic polarization is high at low frequencies and decreases as the frequency of the applied field is increased [78]. Due to exceptional properties like great thermal stability, antimicrobial features, and biocompatibility, ZnO has proved to be a promising antibacterial agent [79–82]. ZnO in the food packaging industry has proved to improve the shelf life of food. It shows superb antibacterial properties for almost the entire range of bacteria including *Staphylococcus aureus* and *Escherichia coli* which is responsible for spoiling the food [83–86]. Due to the nontoxic nature of ZnO nanoparticles, these are biocompatible and hence safe for human beings [87–89].

Along with all these properties, it is ecofriendly, cheap, and widely available [90–92]. ZnO finds its application in drug delivery [93], targeted gene delivery, and tumour imaging [94]. It is used as an additive in ointments for various skin problems [95], as an antiseptic [96]. In the textile industry for developing a fabric which is UV radiation protected due to its UV absorption properties [97]. For food

packaging application, its antimicrobial properties help it to prevent the growth of bacteria which spoils the food and thus develop diseases [98, 99]. It also finds applications in devices like LED [100–104], TFTs [105, 106], memory cells [107], solar cells [108], photocatalyst [109], gas sensors [110], lasers [111], storage devices [112], and photodetectors [113].

*5.1. Dielectric Constant of Bulk ZnO and ZnO Nanoparticles.* The value of $k$ of any material is a very important characteristic for various electronic applications like capacitors, transistors, dry electrodes, memory cells, passive components, and gate dielectrics. The $k$ value for bulk ZnO ranges from 6 to 10 approximately at 1 MHz frequency and at room temperature [114]. Dielectric properties of ZnO have been examined by various researchers to find out the value of $k$ of pure ZnO. Look et al. [115] found the value of the dielectric constant of bulk ZnO to be $\approx 8$ at 1 MHz and at room temperature. Langton and Matthews [116] performed the dielectric studies of pure ZnO in the frequency range of 50 KHz to 25 MHz at 25°C and found the value of $k$ for ZnO is 10.4 in the frequency range of 1 MHz to 10 MHz. As the frequency is increased above 10 MHz the value of $k$ decreases and becomes 9.1 at 25 MHz.

It is well known that, with the increase in the ZnO layer thickness, the value of $k$ increases at room temperature. At a fixed frequency, the $k$ of ZnO varies with the variation of its thickness and temperature of operation [105]. It has been observed by Shashikant Sharma et al. that the dielectric constant increases with the increase in the film thickness of ZnO thin films, due to the quantum confinement effect [117]. The reduction in the value of dielectric constant of the thinner film is attributed to the powerful captivity of the smaller ZnO grains [118]. Table 1 and Figure 3 show the variation of dielectric constant as a function of the thickness of ZnO thin films at a fixed wavelength of 600 nm. The grade of crystallinity of ZnO thin films refines with the increase in thickness of films. With the increase in thickness of deposited films, the disorders like dislocation density, stress, and strain reduces. The dielectric behaviour of ZnO is ascribed to the defects like oxygen vacancies along with zinc interstitials [78]. In the optical properties, blue shift is observed with the decrease in thickness [119].

Lanje et al. [33] studied the dielectric properties of ZnO nanoparticles at room temperature and found the value of $k$ to be 10.26 at 1 MHz. Literature disclosed that the value of $k$ of a synthesized material depends upon various factors. The first factor is the size of nanoparticles of the synthesized material. Size dependence of the dielectric constant of ZnO nanoparticles has been studied in this paper. As shown in Table 2 and Figure 4, an increase in the value of $k$ is observed with the increasing size of nanoparticles. It has been observed that the larger is the crystallite size of the particle, the higher is the value of $k$ because the larger particle contains a greater number of dipoles which get aligned under the influence of the electric field, hence resulting in greater permittivity and higher value of $k$ [57].

The dielectric constant of bulk zinc oxide is observed to be less than ZnO nanoparticles due to the fact that

TABLE 1: Variation of the dielectric constant of ZnO thin films with the variation of film thickness at a fixed wavelength of 600 nm [118].

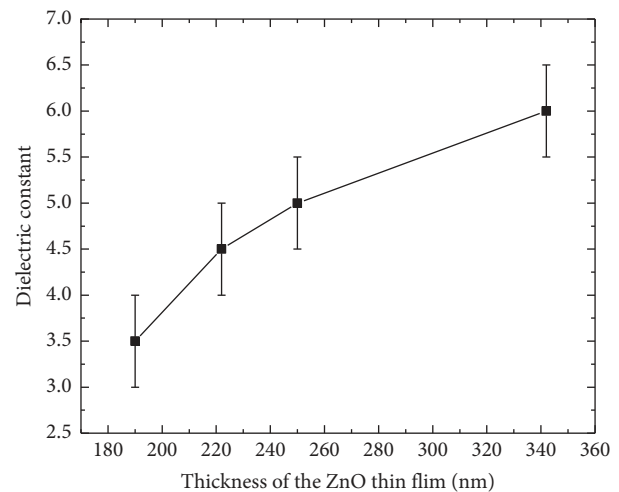| Thickness of the ZnO thin film (nm) | Value of dielectric constant |
|---|---|
| 190 | 3.5 |
| 222 | 4.5 |
| 250 | 5 |
| 342 | 6 |



FIGURE 3: Variation of the dielectric constant of ZnO thin films with respect to film thickness.

TABLE 2: Variation of dielectric constant of ZnO nanoparticles with respect to the particle size.

| Crystallite size of pure ZnO | Dielectric constant | References |
|---|---|---|
| 15 | 5 | [65] |
| 19 | 14.78 | [120] |
| 19.97 | 9.7 | [78] |
| 22.84 | 18.5 | [121] |
| 33.03 | 17 | [122] |
| 55 | 45 | [123] |

semiconductor nanoparticles exhibit a quantum confinement effect. As an outcome of their size in the nanometric range, the widening of the bandgap is caused when the spatial dimension is reduced. Quantum confinement regulates the band structure of the nanoparticles; hence, dielectric properties also change as the size of the nanoparticles is changed [124].

Pure ZnO has limited electronic and optical applications as pure ZnO has blue light emission which is the least eye sensitive colour and shows more scattering [125]. Pure ZnO has fixed and specific electric properties like bandgap, electron mobility, thermal conductivity, and exciton binding energy. The bandgap of ZnO can be tuned to a high or low value by adding some dopant. The addition of magnesium (Mg) into ZnO will increase the bandgap from 3.37 eV to 4 eV and the addition of cadmium (Cd) will decrease the bandgap to 3 eV [36]. Electrical and dielectric properties of ZnO depend upon the concentration of dopant. So, these
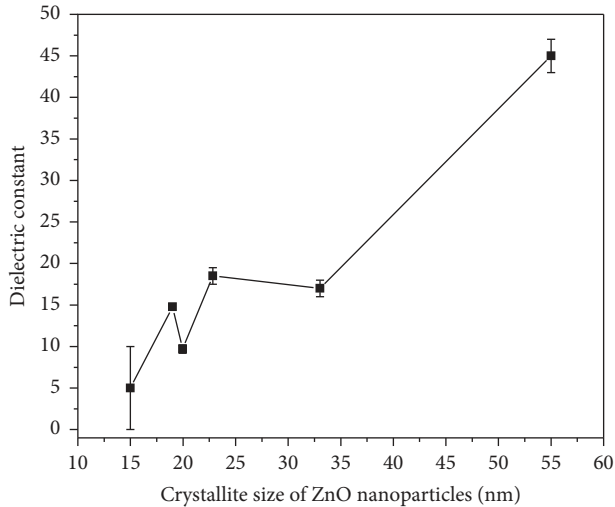
FIGURE 4: Variation of the dielectric constant of ZnO nanoparticles with respect to the crystallite size of ZnO nanoparticles.

properties can be tailored by adding some other materials in it and creating the composite.

## 6. Composites

When two or more materials which have quite different physical and chemical characteristics are combined in a manner that their properties remain distinct at the microscopic level, this type of material is called a composite material. Due to good physical strength and high melting point, high thermal conductivity composites find their applications in various industries [39].

*6.1. Metal-Doped ZnO Nanocomposites and Their Applications.* Two groups of metals, transition metal and rare earth metal, doped ZnO nanoparticles have been chosen for this review article. Ferromagnetism in transition metal (TM) doped ZnO is theoretically investigated by Sato and Katayama-Yoshida [126]. It turned out that most of the incomplete 3d shell metal ions can be used to produce room-temperature magnetism in ZnO, doped with transition metal [127]. In the case of rare earth (RE) doping, the 4f shell electrons are localized, exchange interactions are indirect as they occur via the 5d or 6s conduction electrons. However, the high orbital momentum leads to a high total magnetic moment per atom [128]. Other than introducing ferromagnetism transition metals, rare earth metal ions have been used by many researchers to dope ZnO. The doping thus modifies its optical, electrical, and dielectric properties by changing the bandgap and electronic structure of the composite material. Due to enhanced properties, it turned perfect for different technological applications like LED, TFTs, memory cells, solar cells, photocatalyst, gas sensors, lasing, storage devices, and photodetectors [129, 130]. Various researchers have synthesized different composites and have checked their dielectric properties.

*6.1.1. Transition Metal-Doped ZnO.* In this article transition metal-, Cr [121], Mn [57], Fe [78], Co [131, 132], Ni [122], and Cu [123], doped ZnO nanoparticles have been discussed for their dielectric studies. There are various factors that affect the value of the dielectric constant of a material like operating frequency, percentage of doping concentration, and atomic number of the dopant metal.

The first factor is the operating frequency: at low frequency, the value of $k$ is high, and at high frequencies, the value becomes low for every material due to polarization phenomena [41, 133]. Figure 5 shows the graph which shows the variation of the dielectric constant of various transition metal-doped ZnO nanoparticles with respect to frequency in the range of 0 Hz to 10 MHz. Figure 5 shows that the value of the dielectric constant of every metal-doped ZnO is high at low frequencies since all four sources of polarization are active at low frequencies. ZnO is a polar molecule, so all the dipoles get aligned in the direction of the applied electric field and give rise to a higher value of $k$. As the frequency increases the value of $k$ starts decreasing and becomes constant at a higher frequency because the dipoles are unable to align themselves with the changing direction of the applied field. Hence, the value of $k$ decreases at high frequencies [120–123]. The phenomena of dielectric dispersion in such heterogeneous structures can be simply explained by the Maxwell–Wagner model [134–136]. According to this model, a dielectric medium is composed of conducting grains which are separated by resistive grain boundaries. The grains are effective at high frequencies and the grain boundaries are effective at low frequencies. Under the influence of the external field, the charge carriers easily transfer through grains but get trapped into the grain boundaries at low frequencies; this produces large polarization and high $k$. Among all the transition metals discussed in this article, Mn is showing a significant dielectric dispersion. The higher value of $k$ at low frequencies can be explained based on interfacial polarization due to inhomogeneous structure, defects like oxygen vacancies, and grain boundary defects. Due to this reason, the value of $k$ in Mn-doped ZnO is quite high at low frequencies; as the frequency increases, the value of $k$ starts decreasing; and after a particular frequency, it becomes constant due to the fact that hopping between metal ions cannot follow the alternating field and are unable to align themselves in the direction of the electric field.

*(1) Effect of Transition Metal Doping Concentration on Dielectric Constant.* The second factor that affects the value of $k$ is the composition and doping concentration. When ZnO nanoparticles are doped with metal ions, the crystal structure and microstructure of the nanoparticles change drastically which affects its chemical, electrical, mechanical, optical, and dielectric properties. This gives rise to various applications. The variations of the dielectric constant of different transition metal-doped ZnO nanoparticles have been discussed with respect to the percentage of doping concentration. As shown in Figure 6, some of the doped materials have a linearly decreasing trend with the increase in doping percentage and others have an increasing trend.
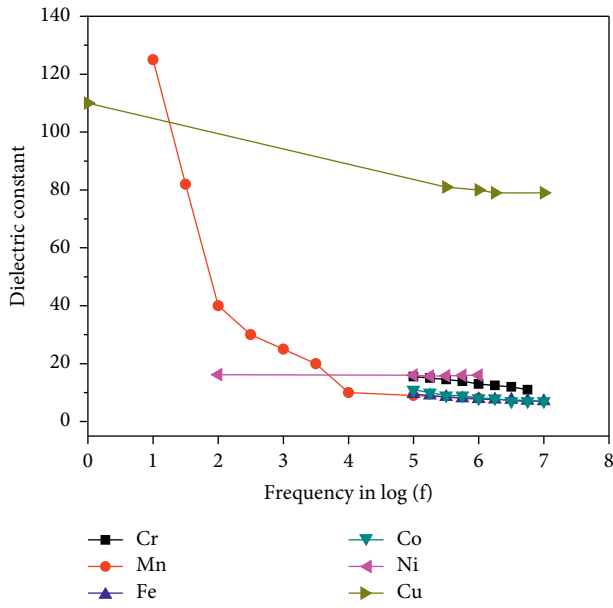
FIGURE 5: Variation of the dielectric constant of various transition metal-doped ZnO nanoparticles with respect to frequency.
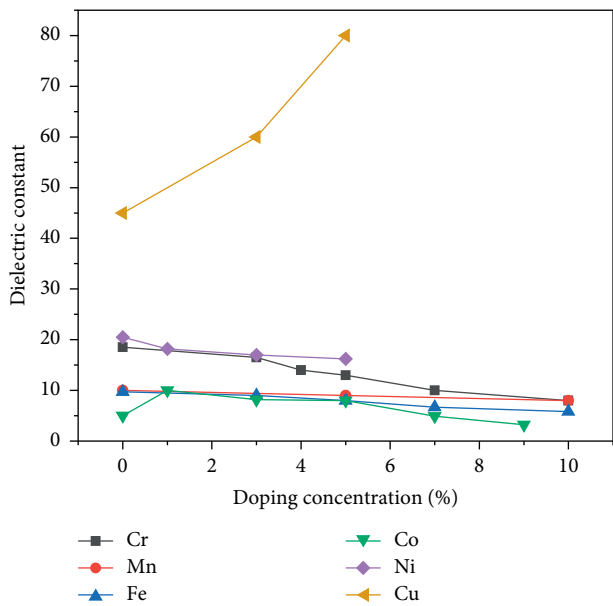


FIGURE 6: Variation of dielectric constant of transition metal-doped ZnO nanoparticles with respect to doping concentration at 1 MHz frequency.

Figure 6 shows the variation of $k$ of transition metal-doped ZnO nanoparticles with different doping concentrations at 1 MHz frequency. The value of $k$ decreases with the increase in the concentration of Cr, Mn, Fe, Co [132], and Ni into the ZnO nanoparticles. The doping of these metals prevents the growth of crystal grains and slows down the motion of a grain boundary. This reduction in motion of grain boundaries can be well explained with the Zener pinning effect [122]. As the ionic radius of these metals is smaller than Zn, it reduces the lattice parameters and

crystallite size of the doped material [137, 138]. It has already been discussed that, with the reduction in crystallite size of nanoparticles the value of $k$ decreases [57, 58]. Hence, a reduction in the size of nanoparticles leads to a reduction in the $k$ value of doped material. When these metal ions are doped into ZnO at a lower concentration, the hopping mechanism between the metal ion and $Zn^{2+}$ is predominant which results in a higher value of $k$. As the concentration is increased the metal ion in ZnO acts as a deep donor and decreases the concentration of intrinsic donors. It substitutes $Zn^{2+}$ ions and produces charge imbalance around the dopants, which reduces the value of $k$. But in case of Cu, the value of $k$ increases with the increase in the doping concentration; this is due to the abundance in electron exchange between Zn and Cu ions at all frequencies. Furthermore, it is seen that doping with Cu ions increases the grain size of doped material, so a large value of the dielectric constant may be attributed to the larger grain size and oxygen vacancies. Hence, it can be concluded that Cu doping enhances the polarization of ZnO nanoparticles.

*(2) Effect of Atomic Number of Transition Metal on Dielectric Constant.* The third factor is the atomic number of the dopant material. In order to compare the relative behaviour of various transition metal-doped ZnO nanocomposites, we have selected fixed doping concentration of 5% and fixed frequency of 1 MHz for every metal-doped ZnO nanocomposite. Figure 7 shows the variation of $k$ value of various transition metal-doped ZnO nanoparticles at 5% doping and at 1 MHz frequency with respect to their atomic number. It has been observed that the value of $k$ of various transition metal-doped ZnO nanoparticles increases exponentially as the atomic number increases from 24 to 29. The exceptional increase in the dielectric constant of Cu doped ZnO nanoparticles is due to the anomalous electronic configuration of Cu having atomic number 29, $3d^9 4s^2$ (ground state) having unfilled d orbital and fully filled s orbital. But instead of exhibiting this electronic configuration, it exhibits anomalous configuration having fully filled d orbital and partially filled s orbital, as the full 3d orbital is more stable than a full 4s orbital. This configuration causes the alignment of spin of the electrons resulting in long-range ferromagnetic order in Cu doped ZnO. Doping at the 3d transition metal site by transition metal ions in ZnO lattice has observed a variety of magnetic and electronic orders with spatially correlated charge spin and orbital degree of freedom. Hence, enhanced dielectric constant is observed.

*6.1.2. Rare Earth Metal-Doped ZnO.* Rare earth metal-, Ce [139], Gd [130, 140], La [141], Nd [142], Ho [143], Er [144], and Tm [145], doped ZnO nanoparticles have been discussed for their dielectric studies in this article.

Figure 8 shows the variation of the dielectric constant of various rare earth metal-doped ZnO nanoparticles with respect to frequency in the range of 0 Hz to 10 MHz. In the graph, it has been observed that the value of the dielectric constant of every rare metal-doped ZnO is high at low frequencies since all four sources of polarization are active at
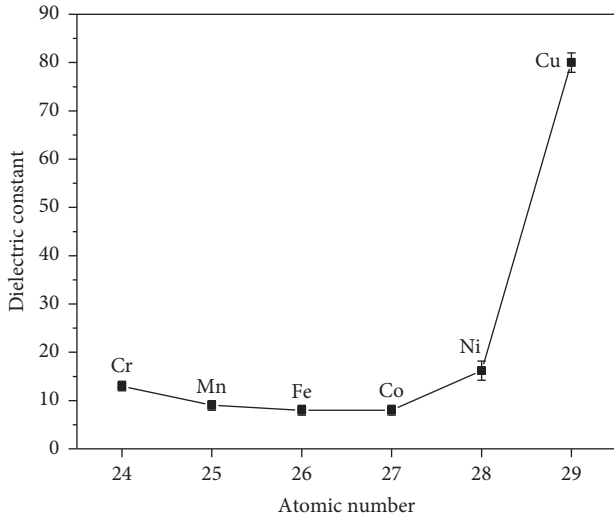
FIGURE 7: Variation of the dielectric constant of various transition metal-doped ZnO nanoparticles at 5% doping and at 1 MHz frequency with respect to their atomic number.
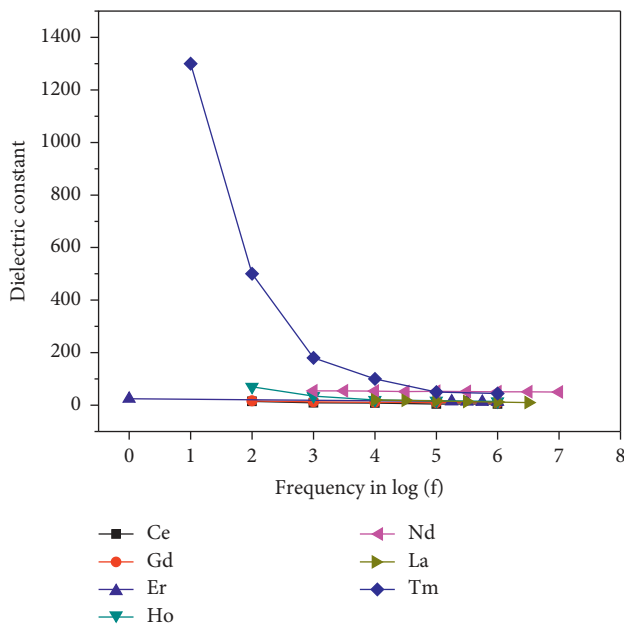


FIGURE 8: Variation of the dielectric constant of various rare earth metal-doped ZnO nanoparticles with respect to frequency.

low frequencies. ZnO is a polar molecule so all the dipoles get aligned in the direction of the applied electric field and give rise to a higher value of $k$. As the frequency increases, the value of $k$ starts decreasing and after a particular frequency it becomes constant, as hopping between different metal ions cannot follow the alternating field. Hence, the value of $k$ decreases at high frequencies [141–145]. The phenomena of dielectric dispersion are attributed to the Maxwell–Wagner model. Among all the rare earth metals discussed in this article, Tm is showing a significant dielectric dispersion. The value of $k$ is very high at low frequencies because Tm is highly polarizable and a higher value

of $k$ at low frequencies can be explained based on interfacial polarization due to inhomogeneous structure and defects like oxygen vacancies and grain boundary defects.

*(1) Effect of Rare Earth Metal Doping Concentration on Dielectric Constant.* Figure 9 shows the variation of $k$ for some rare earth metal-doped ZnO nanoparticles with different doping concentrations at 1 MHz frequency. In Ce-doped ZnO nanoparticles, initially, the value of $k$ decreases due to the high density of defects. But a further increase in doping concentration increases the value of $k$ due to less electronegativity of Ce than Zn that makes the ionic bonds weaker [139]. The dielectric constant of Gd-doped ZnO nanoparticles decreases with the increase in doping concentration due to a decrease in relaxation time where ionic and orientational polarization are closely connected with each other. Gd acts as an acceptor and absorbs free charge carriers, hence decreasing the value of $k$ [130].

A larger ionic radius of $Nd^{3+}$ ions substituted for $Zn^{2+}$ ions in Nd-doped ZnO indicates local distortion in the doped ZnO lattice, due to which, initially, the value of $k$ increases with doping but as the doping concentration is increased further, the sample becomes porous and hence the value of $k$ starts decreasing, as these pores get filled with air which has low dielectric constant, hence decreasing the overall dielectric constant of the doped sample [142].

The value of $k$ in Ho-doped ZnO increases monotonically with the increase in doping concentration. This is because the decrease in columbic interaction causes an increase in exciton binding energy which leads to an increase in the value of $k$ [143]. The value of $k$ in Er-doped ZnO nanoparticles varied according to its size. Initially, up to 3% of doping, the value of $k$ decreased and then it increased again because initially the nanoparticles had a rod-like structure and with the further increase in concentration, it started to change into a plate-like structure. Hence, the value of $k$ started to increase with the increase in the concentration of Er [144].

*(2) Effect of Atomic Number of Rare Earth Metal on Dielectric Constant.* Figure 10 shows the variation of $k$ value of various rare earth metal-, La [141], Ce [139], Nd [142], Gd [140], Er [144], and Tm [145], doped ZnO nanoparticles at 5% doping and at 1 MHz frequency with respect to their atomic number. It has been observed that the value of $k$ of various rare earth metal-doped ZnO nanoparticles increases exponentially as atomic number increases from 57 to 69. Significant enhancement in the dielectric constant of Tm-doped ZnO is due to the anomalous electronic configuration of Tm having atomic number 69, $4f^{13} 6s^2$ (ground state) having unfilled f orbital and fully filled s orbital. But instead of exhibiting this electronic configuration, it exhibits anomalous configuration having fully filled f orbital and partially filled s orbital as the full 4f orbital is more stable than a full 6s orbital. This configuration causes the alignment of the spin of the electrons resulting in long-range ferromagnetic order in Tm doped ZnO. Doping at the posttransition metal site by rare earth ions in ZnO lattice has observed a variety of magnetic and electronic orders with spatially correlated

# 7. Polymer

Polymers are of great attraction for researchers due to various interesting properties like low cost, flexibility, lightweight, easy processing, and wide range of operating frequency [35]. Petrochemical plastics were used widely in consumer-based products, food packaging, lightings, automobiles, and many electronic applications such as printed circuit board (PCB) due to their easy modification of shape, structure, and properties [146].

*7.1. Polymer Nanocomposites.* The small size particles called nanofillers are used to distribute into the polymer matrix which acts as a flexible base matrix which gives rise to a polymer nanocomposite [147, 148]. So, the properties of a composite material are greatly affected by the choice of polymer matrix and the nanofiller. Polymer-based nanocomposites have many applications in the field of medical and engineered technology due to their good electron transport and mechanical and optical properties [149, 150]. They are good aspirants for energy storage applications. Degradation resistance improved thermal and mechanical properties along with improved dielectric breakdown strength being the added advantages of polymer nanocomposites [151, 152].

*7.1.1. Polymer-Based ZnO Nanocomposites.* A lot of work is done to explore the dielectric and electrical properties of polymer nanocomposites. Figure 12 shows the variation of the dielectric constant of various polymer-based ZnO nanocomposites with respect to frequency in the range of 0 Hz to 1 MHz. In the graph, it has been observed that the value of the dielectric constant of every polymer-based ZnO nanocomposite is high at low frequencies because all four sources of polarization are active at low frequencies and ZnO is a polar molecule, so all the dipoles get aligned in the direction of applied electric field and thus give rise to a higher value of $k$. As the frequency increases, the value of $k$ starts decreasing and after a particular frequency, it becomes constant since dipoles cannot follow the alternating field and are unable to align themselves in the direction of the electric field. Hence, the value of $k$ decreases at high frequencies [141–145]. The dielectric dispersion in the case of polymer nanocomposites is attributed to Maxwell–Wagner–Sillars (MWS) effect which represents the collection of charge at the interfaces of materials having different conductivity which leads to increased polarizability in the composite material [153].

In ethylene-vinyl acetate (EVA)/ZnO nanocomposite, the larger value of dielectric constant at low frequencies is due to the space change polarization which arises because of impurities and defects present in the nanocomposite [38]. In case of polyvinyl chloride (PVC)/ZnO nanocomposite, the value of dielectric constant is high at low frequencies and the value starts decreasing as the frequency is increased. This is
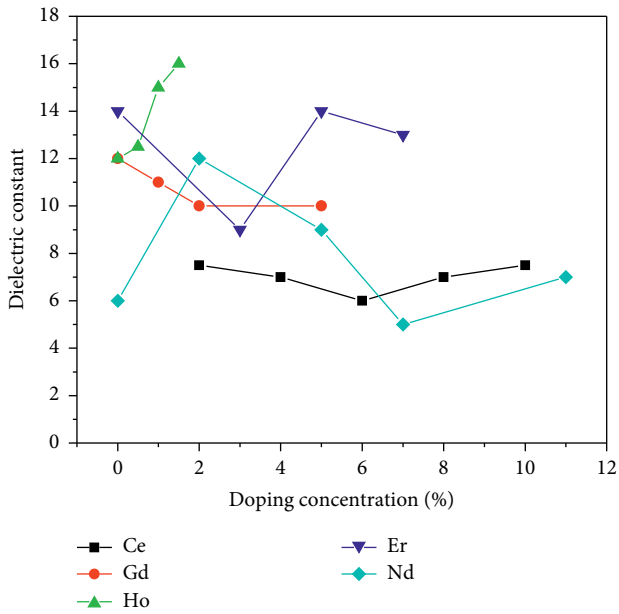


FIGURE 9: Variation of dielectric constant of rare earth metal-doped ZnO nanoparticles with respect to doping concentration at 1 MHz frequency.
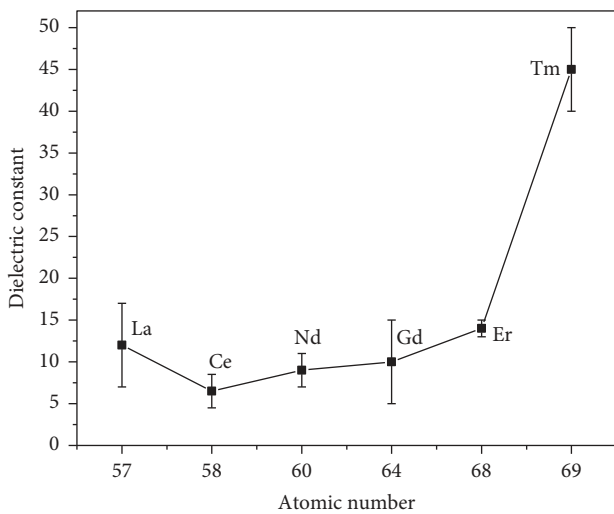


FIGURE 10: Variation of the dielectric constant of various rare earth metal-doped ZnO nanoparticles at 5% doping and at 1 MHz frequency with respect to their atomic number.

charge spin and orbital degrees of freedom. Hence, large ferromagnetism and enhanced dielectric constant are observed.

Table 3 expresses the value of $k$ of various metal-doped ZnO nanoparticles for 5% doping at an operating frequency of 1 MHz and their respective electronic applications. The graph in Figure 11 shows that Cu among transition metals and Tm among all the rare earth metals discussed in this article attain the highest value of $k$, 80 and 45, respectively, at 5% doping and at 1 MHz frequency.

TABLE 3: Various metal-doped ZnO nanoparticles and their potential applications.

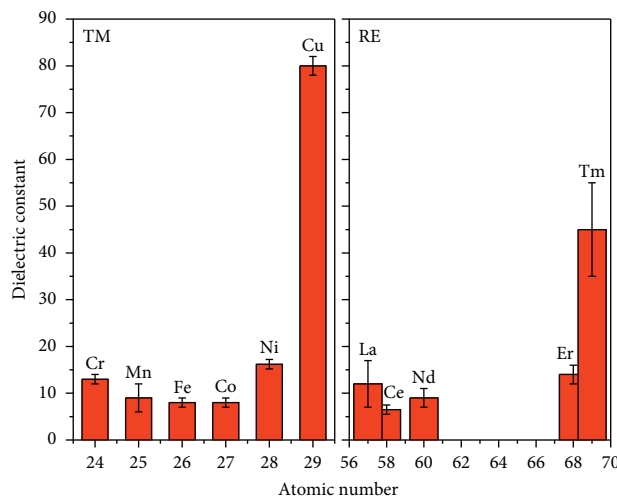| Metal | Doping (%) | Frequency (MHz) | Dielectric constant of metal-doped ZnO nanoparticles | Applications |
|---|---|---|---|---|
| Ce | 5 | 1 | 6.5 | High-frequency device applications |
| Fe | 5 | 1 | 8 | Nano-optoelectronic applications |
| Co | 5 | 1 | 8 | Optical applications |
| Mn | 5 | 1 | 9 | Spintronic applications |
| Nd | 5 | 1 | 9 | Pressure sensors, piezoelectric nanogenerators for energy harvesting |
| La | 5 | 1 | 12 | Storage applications |
| Cr | 5 | 1 | 13 | LEDs, optoelectronic devices |
| Er | 5 | 1 | 14 | High-frequency device applications |
| Ni | 5 | 1 | 16.2 | Optoelectronic applications |
| Tm | 5 | 1 | 45 | Dye-sensitized solar cell applications. |
| Cu | 5 | 1 | 80 | High dielectric constant applications |



FIGURE 11: Dielectric constant of various metal-doped ZnO nanoparticles for 5% doping at 1 MHz frequency.
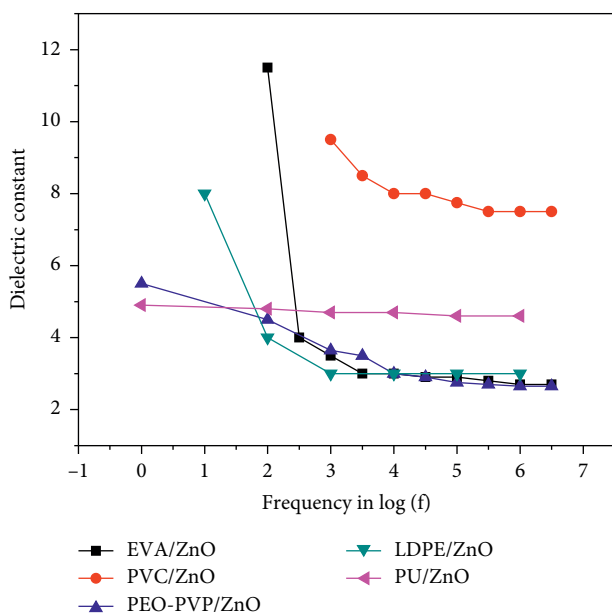


FIGURE 12: Variation of the dielectric constant of various polymer-based ZnO nanocomposites with respect to frequency.

due to the reduction in orientational polarization at high frequencies [154]. In poly(ethylene oxide) (PEO) poly(vinyl-pyrrolidone) (PVP) blend matrix PEO-PVP/ZnO nanocomposite, the larger value of $k$ at low frequencies is attributed to the interfacial polarization in the nanocomposite material [155]. In case of low-density polyethylene (LDPE)/ZnO nanocomposite, the value of dielectric constant is higher at low frequencies due to the presence of interfacial polarization and as the frequency increases, the value of $k$ decreases as interfacial polarization is no longer active at high frequencies [156].

Figure 13 shows the variation of $k$ of the above-discussed polymer ZnO nanocomposites with different doping concentrations at 1 KHz frequency. In every polymer nanocomposite, the value of $k$ increases as the concentration of ZnO nanofiller is increased, this is because these polymers have a low dielectric constant as compared to the ZnO nanoparticles. So, when a composite is prepared with ZnO as a nanofiller, the effective value of $k$ of the polymer nanocomposite is increased. And as the concentration is increased, the effective value of $k$ further increases. Along with this, homogeneous dispersion of nanofillers in the polymer matrix are required to ensure that there is no agglomeration
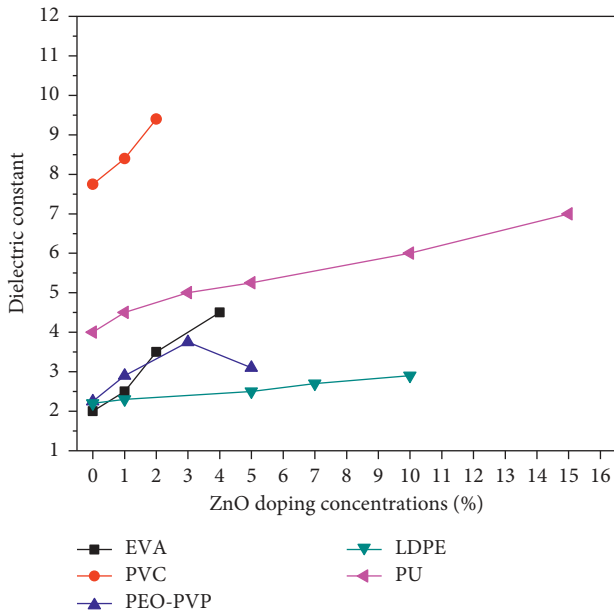
Figure 13: Variation of the dielectric constant of various polymer-based ZnO nanocomposites with respect to doping concentration at 1 KHz frequency.

of nanoparticles that provide tunnelling, hence reducing the dielectric breakdown strength of polymer nanocomposite [157]. In case of PEO-PVP/ZnO nanocomposite, the value of $k$ increases up to 3% filler concentration due to the fact that the dielectric constant of PEO-PVP is less than ZnO nanoparticles hence with the addition of ZnO, there is an increase in the effective dielectric constant of the polymer nanocomposite. But after that at 5%, it started decreasing which may be due to the electrostatic interaction between functional groups of polymer blend and ZnO nanoparticles resulting in a reduction of the orientation of dipoles, hence resulting in reduced dielectric polarization in the nanocomposite. In case of PU/ZnO and LDPE/ZnO nanocomposite, the value of $k$ increases very linearly with the increase in the concentration of ZnO nanoparticles. But in case of EVA/ZnO and PVC/ZnO nanocomposite, an abrupt change in $k$ is observed with the increase in the concentration of ZnO nanoparticles. Due to the lack of availability of data points in the literature, a conclusion cannot be made if the values of the dielectric constant will increase exponentially or like a parabola with the increase in concentration. Hence, in case of polymer-based ZnO nanocomposites, there is still room available to study the effect of concentration on the dielectric properties.

*7.1.2. Applications of Polymer Nanocomposites.* Polymer nanocomposites are used in several electronic applications due to their promising electrical, dielectric, optical, and mechanical properties. Polymer nanocomposites are used in the devices like organic field effect transistors [158–161], gas sensors [162–166], solar cells [167–171], high power and high storage density capacitors and supercapacitors [172–177], electromagnetic interference shielding [178], thermochromic and electrochromic devices [179], electronic antenna [180], optoelectronic devices [181], high capacity memory devices [182], insulating materials [183, 184], nanodielectric applications [185, 186], and printed circuit board [187–189].

*7.2. Biodegradable Polymers.* Although polymer nanocomposites show very good and promising dielectric properties and enhanced dielectric breakdown strength, the biggest disadvantage is the toxic nature of petroleum-based polymers. When the devices or products made up of petroleum-based polymers are disposed of, they contaminate the environment, which is harmful to living organisms and human beings [97]. So, the electronic products are required to be made up of a material which is biocompatible and biodegradable and is safe for human beings along with other living organisms, hence promoting the green technology.

*7.2.1. Biodegradable Polymer Nanocomposites.* Nowadays, biofriendly or echo-friendly materials are high in demand in every field as they are nontoxic [38,190]. Biodegradable polymer nanocomposite is a category of materials that have very interesting properties compared to pure biodegradable polymers and old composites. They have all the properties and advantages of polymers, but an additional advantage is that they are biodegradable. Biodegradable polymers are made up of living organism's protein and DNA. Biodegradable polymers nanocomposites are doing very well in various fields. They are used for food packaging to improve the shelf life of food [191–193], as a photocatalyst to purify water [194, 195], cancer treatments [196–198], drug delivery in the medical field [199–201], antimicrobial activities [202–204], tissue engineering [205], and sensors [206, 207].

Various researchers have made use of biodegradable polymers to develop some new biodegradable nanocomposites. There are various biodegradable polymers like poly(3-hydroxybutyrate) (PHB), polylactic acid (PLA), polycaprolactone (PCL), poly(butylene succinate) (PBS), poly(butylene adipate-co-terephthalate) (PBAT), poly(3-hydroxybutyrate-co-3-hydroxyvalerate) (PHBV), (poly-lactide-co-glycolide) (PLGA), chitosan, cellulose, and gluten. These are 100% biodegradable polymers, and they have several applications in different areas. Polyhydroxyalkanoates (PHA) is a class of polymers which is made from renewable sources synthesized by bacteria from carbon and oxygen [208, 209]. In this class, PHB is the most widely studied and useful biopolymer. But it has some disadvantages like it is very brittle, cost of production is very high, it does not have any resistance to thermal degradation, and it has a high melting point, due to which it is not suitable for industrial applications [209–212].

*7.2.2. Biodegradable Polymer-Based ZnO Nanocomposites and Their Applications.* All the LEDs emit UV radiations in addition to visible light which is very harmful to the living organisms [213]. Poly(3-hydroxybutyrate-co-3-hydroxyvalerate) (PHBV) polymer/ZnO nanocomposite provides

the absorption of entire UV region. This enables the material to be used as an encapsulation material for LEDs [190]. This material absorbs all the UV radiation spectra and emits only visible light. The PHB-ZnO nanocomposite material can be used in textile industry for UV blocking applications [214–216]. The same material can also be used to manufacture printed circuit boards (PCB) which are nontoxic and do not contaminate the environment when disposed off [190]. The material is good for antibacterial applications [217]. PHBV and ZnO nanocomposite is very good for food packaging applications [218–221]. Enhanced value of dielectric constant of polylactide acid (PLA) and poly(butylene adipate-co-terephthalate) (PBAT) with graphene nanocomposites is useful for EMI shielding applications [222, 223]. Poly(butylene succinate) (PBS) and ZnO nanocomposite films are very suitable material for food packaging to improve the shelf life of packed food due to its good antimicrobial and mechanical properties [224].

Enhancement in mechanical, antimicrobial, and dielectric properties of chitosan/ZnO nanocomposite films with the increase of ZnO concentration is very good for food packaging applications [225–227]. Chitosan-ZnO/polyaniline ternary nanocomposite material is recommended for storage of charge in supercapacitors [228]. Chitosan is a nontoxic, biocompatible, and biodegradable polymer which is obtained from shells of lobsters or crabs and also in the cell walls of fungi. It has antimicrobial and UV protection properties as well [229–231]. The enhanced photoluminescence intensity of the Chitosan capped ZnO nanocomposite makes the material suitable for display devices and biomedical applications [125, 232, 233].

*7.3. Future Aspects.* The field of biodegradable nanocomposites is still being explored because it has a lot more to give to the society and electronic industry:

(1) Higher value of dielectric constant for ZnO nanoparticles can be achieved by doping it with some rare earth metals or transition metals. As ZnO is a nontoxic material, it is safe for the human being and the environment to be used in electronic products like thin-film transistors, memory cells, and high-speed integrated circuits.

(2) Keeping in mind environmental safety there is a need to design and synthesize nontoxic and biodegradable dielectric material for energy storage applications. Hence, ZnO-based biodegradable polymer nanocomposites can be used as a dielectric material for developing energy storage devices like capacitors to enhance their energy density.

(3) To enhance the energy storage density, a multilayered structure of ZnO-based biodegradable polymer nanocomposite can be developed to achieve a higher value of energy density.

(4) A sandwiched structure of ZnO-based biodegradable polymer nanocomposites with different concentrations of ZnO nanoparticles as filler in the biodegradable polymer as the base matrix can be used to enhance the energy density of the material further.

## 8. Conclusion

Dielectric constant is a very important factor of a material which determines its applications in the electronic industry as a capacitor, thin-film transistor, memory devices, high-speed IC packaging, and EMI shielding. ZnO is one of the most demanded materials in the electronic industry due to its very attractive properties like wide availability, nontoxicity, and cost-effectiveness. Nanocomposites of ZnO have been explored by various researchers. Two groups of metals, transition metal and rare earth metal, have been chosen to be a dopant in ZnO for this review. The literature revealed that the value of the dielectric constant of a synthesized material depends upon various factors. The first factor is the size of nanoparticles of synthesized material, the larger the size of nanoparticles, the higher will be the value of dielectric constant due to the fact that larger particle contains a greater number of dipoles which get aligned under the influence of the electric field. Second, the thickness of films also affects the value of the dielectric constant. The value increases with the increase in the film thickness of ZnO thin films due to the quantum confinement effect. Third, the density of the material: if the material is porous, the value of the dielectric constant will be less and if the material is of high density, the value of the dielectric constant will be high. Fourth, operating frequency: at low frequency the value of the dielectric constant is high and at high frequencies, the value becomes low for every material since all four sources of polarization are active at low frequencies, and at high frequencies, the dipoles are unable to align themselves with the changing direction of the applied field. Fifth, the defects in the material, such as oxygen vacancies, zinc interstitials, and inhomogeneities like grain boundaries, are inherent to the synthesis process of the material. The higher the defects, the higher will be the value of dielectric constant of the material at low frequencies.

Among all the metals discussed in this article, transition metal Mn and rare earth metal Tm are showing a significant dielectric dispersion. The higher value of $k$ at low frequencies can be explained based on interfacial polarization due to inhomogeneous structure and defects such as oxygen vacancies and grain boundary defects. The exponential increase in the dielectric constant of Cu among transition metals and Tm among rare earth metal-doped ZnO at the same doping concentration is due to the anomalous electronic configuration.

Petroleum-based polymer ZnO nanocomposites are also observed to show promising dielectric properties. But they lag due to their toxic nature which contaminates the environment when they are disposed of. Biodegradable ZnO nanocomposites are explored by various researchers which shows good mechanical, antibacterial, antimicrobial, and dielectric properties which enabled these materials to be used as food packaging materials and in various medical

applications. Still, there is much room available to study the effect of filler concentration on the dielectric properties of polymer ZnO nanocomposites. To experience all the advantages of a polymer material along with the nontoxicity, the dielectric properties of biodegradable ZnO nanocomposites need to be explored in depth so that these materials can be used in electronic applications as storage materials and many more electronic applications. Hence, electronic products need to be made up of the material, which is biocompatible, biodegradable, and safe for living organisms as well as human beings, hence promoting green technology.

## Data Availability

The data used to support the findings of this study in the manuscript are available from the corresponding author upon request.

## Conflicts of Interest

The authors declare that they have no conflicts of interest.

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