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Effect of NiO Filler on the morphology, Dielectric Loss Factor and Loss Tangent of ZnO using Solid State Method at Microwave Frequency

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***Abstract*-** **Among transition metal oxides, nickel oxide (NiO) both bulk and nano size have received considerable attention due to its wide range of applications in different fields. The aim of this research is to characterize the dielectric loss factor and loss tangent of doped zinc-oxide composites using solid state method at microwave frequency. The methods used in this research are solid state method for sample preparation and open ended coaxial probe (OECP) for determining the dielectric loss factor and loss tangent. The OECP results shows a sequential increase in dielectric loss factor and loss tangent as pure ZnO is doped incrementally with the filler (NiO). It also shows a sequential decrease in dielectric loss factor and loss tangent as the frequency increases. The SEM result revealed agglomeration of particles as doping increased for all compositions. Therefore NiO can be used as a filler for improving dielectric loss factor and loss tangent of ZnO as a matrix. The composite can also serve as a good agent for constructing dielectric based materials which can be used in electronic and telecommunication gadgets. It is also proven that solid state method is suitable for synthesis of powdered sample like ZnO and NiO for determining their dielectric loss factor and loss tangent.**

Keywords—Nickel Oxide (NiO); Zinc Oxide (ZnO); Dielectric loss factor; loss tangent; Solid State Method.

I. INTRODUCTION

A nanocomposite is formed when two or more materials are mixed on a nanometer scale to obtain a new material with properties that depend on the contribution of each component in the mixture. The physical properties of nanomaterials are mainly affected by the morphology, structural properties, chemical composition and the size of the nanoparticles. For a nanocomposite, these properties can be controlled by determining the volume fractions of the constituent phases. ZnO is a wide band gap (3.2 eV) n-type semiconductor that has found a wide range of applications as a transparent conducting oxide (TCO) electrode in photovoltaics, photocatalysis, sensing, fuel cells and other optoelectronic devices (Albert & Abiola, 2017). Umit, (2010), reported that, ZnO is an attractive material for applications in electronics, photonics, acoustics, and sensing. In optical emitters, its high exciton binding energy (60 meV) gives ZnO an edge over other semiconductors such as GaN if reproducible and reliable p-type doping in ZnO were to be achieved, which currently remains to be the main obstacle for realization of bipolar devices. On the electronic side, ZnO holds some potential in transparent thin film transistors (TFTs) owing to its high optical transmittivity and high conductivity.

NiO is a p-type wide band gap (4.2 eV) semiconductor that has been used in similar applications. ZnO and NiO readily form a p-n junction that has shown good electrical properties for gas sensing, fuel cell electrodes and photocatalysis (Albert & Abiola, 2017). The applications of Nickel oxide (NiO) today is found in semiconductors, capacitor–inductor devices, tuned circuits, transparent heat mirrors, thermistors and varistors, batteries, micro-supercapacitors, electrochromic and chemical or temperature sensing devices. It is used in preparation of nickel cermet, plastics and textiles, in nanowires, nanofibers and specific alloy and catalyst applications. It is also used as an antiferromagnetic layers, accelerators and radar absorbing materials, aerospace and active optical filters (Sani *et al*., 2019; AzoNano, 2013).

Raju and Murphy (2012), synthesized a series of nanocomposites of nickel–zinc ferrite + paraformaldehyde successfully using the mechanical milling process and studied the dielectric loss factor and loss tangent of the composites. He reported that, with the increase in the volume of polymer, the dielectric loss factor and loss tangent of all the composites decreases. Sourav *et al.* (2014), used thermal decomposition route to synthesize Nickel nanocomposites. The dielectric loss factor and loss tangent was observed to decrease rapidly with increase in frequency in the low frequency range and it reach a constant value at the high frequency range which is independent of frequency. Ahmad *et al*. (2015), fabricated the composite of oil palm empty fruit bunch fiber (OPEFB) which is the waste product of oil palm industry, environmentally friendly polycaprolactone (PCL) and nickel oxide (NiO) by compounding all materials in the

Thermo Haake blending machine. The dielectric loss factor and loss tangent of the substrates were obtained with the open ended coaxial method for microwave frequency range between 0.2 MHz and 20 GHz. The results revealed that the permittivity values of the composite can be tuned by changing the ratio of OPEFB/PCL/NiO prior to compounding and blending.

Gaurav *et al.* (2015), studied the improvement in dielectric loss factor and loss tangent of nematic liquid crystal (NLC) by doping of nickel oxide (NiO) nanoparticles. They observed that the dielectric loss factor and loss tangent decrease as the frequency increases. Ajai *et al. (*2017), synthesized polyaniline-nickel ferrite (PANI/$NiFe_2O_4$) composites using interfacial polymerization method. They reported that, dielectric loss factor and loss tangent of PANI/NiFe2O_4 composites are large at lower frequencies and decrease steeply with increasing frequency. In the other hand, Ramesh *et al.* (2003), measured ultralow dielectric loss factor and loss tangent values on Ni–Zn ferrites prepared using Fe_2O_3 as a starting material and sintered in a microwave field. Dielectric loss factor and loss tangent were observed between microwave-sintered Ni–Zn ferrites prepared using Fe_3O_4 (T34) and those starting with Fe_2O_3 (T23) ingredients. The ultralow dielectric loss factor and loss tangent values observed on T23 ferrites show that this procedure is highly suitable for preparing Ni–Zn ferrites for high-frequency switching applications. Bahari *et al. (*2012), used sol-gel method in fabricating polyvinylpyrrolidone / Nickel oxide (PVP/NiO) using 0.2 g of PVP at different working temperatures of 80, 150 and 200 ºC. The obtained results demonstrate the feasibility of using high dielectric loss factor and loss tangent nanocomposite PVP/NiO as gate dielectric insulator in the organic thin film transistors (OTFTs).

Aldar *et al.* (2014), reported that the dielectric loss factor and loss decreases with increase in frequency. The dielectric loss factor and loss decreases rapidly at low frequencies and becomes quite slow at high frequencies. The higher value of dielectric loss factor and loss tangent at lower frequencies is explained on the basis of space charge polarization.Ghassan and Naeem (2016), investigated the effect of volume filler content ϕ on dielectric loss factor and loss tangent of polyethylene PE filled with nickel (Ni) powders. They observed a sudden increase in the dielectric loss factor and loss tangent of such composites at a critical volume concentration indicating a semi-conducting behaviour. Ranga *et al.* (1999), investigated the dielectric loss factor and loss tangent of polycrystalline mixed nickel–zinc ferrites in the frequency range 100 kHz–1 MHz. It was shown that, the dielectric loss factor and loss tangent for these ferrites is approximately inversely proportional to the square root of the resistivity.

John *et al*. (2008), prepared nanostructured nickel ferrite samples through a chemical precipitation method followed by thermal processes. The dielectric loss factor and loss tangent was studied and the nanoparticles of nickel ferrite showed substantial variation in the values of dielectric loss factor and loss tangent. Banerjee *et al. (*2012), synthesized Barium strontium titanate (BST) ceramics $(Ba_{0.6}Sr_{0.4})TiO_3$ by solid state sintering using barium carbonate, strontium carbonate and rutile as the precursor materials. The samples were doped with nickel oxide in different proportions. It was observed that the dielectric loss factor and loss tangent of BST were modified significantly with nickel oxide doping. Muhammad *et* al. (2012), focus on the preparation of nickel oxide nano-crystallites by a novel low cost sol-gel auto-combustion technique and the dielectric loss factor and loss tangent was studied. The value of dielectric loss factor and loss tangent decreased with the increase of frequency up to 1 MHz, which they attributed to the space charge polarization. Rajashekhar *et al*. (2015), synthesized NiO nanoparticles by self-propagating high temperature (SHS) method. These synthesized NNP are used to blend with PANI to get PANI -NiO (PN) composites. The result of the dielectric behaviour of the above synthesized nanocomposites suites for battery applications.

Osama *et al*. (2014), reported that, addition of SiO_2 in the presence of ZnO and NiO leads to better densification by minimizing the present of closed pores. Results of firing shrinkage as a function of temperature show increase of shrinkage with temperature. The dielectric loss factor and loss tangent shows a decreasing trend for all the samples, the decrease is rapid at lower frequency and slower and stable at higher frequency. Nisha *et al*. (2008), prepared nanoparticles of nickel-cobalt oxide by chemical co-precipitation method. The effect of frequency on the dielectric behaviour have been studied for nanosized samples of nickel-cobalt spinel oxide. It is seen that, with the decreasing frequency, the dielectric loss factor and loss tangent increases much more obviously than that of the conventional materials.

Suresh *et al*. (2017), used the sol–gel technique in the chemical synthesis and characterizations based on electrical studies of pure and Ag-doped zinc oxide (ZnO) nanoparticles. The dielectric studies proved that both the dielectric loss factor and loss tangent reduces as the frequency increases. Latif *et al.* (2012), investigated the dielectric behavior of the polar (CPVA) /ZnO nanocomposite films. The results show that the dopant composition has great influence on the magnitude of dielectric loss factor and loss tangent. The results also show that the composite polymer films have both electric and electronic properties. Wang *et al*. 2017), prepared the samples of 1%, 2%, 3% and 4% Zinc Oxide (ZnO) nano-composite silicone rubber by mechanical method. The dielectric properties of each sample were measured by dielectric spectroscopy. The experimental results showed that the dielectric loss factor and loss tangent of the silicone rubber composite increases with the increase of the content of nano-ZnO.

From the review above, so many methods of preparation were chosen but only Banerjee *et al. (*2012), used solid state which is very cheap and environmentally friendly. The final product remained in solid form and structurally pure with the desired properties. Instead of Barium strontium titanate as in the case of Banerjee *et al. (*2012), ZnO is doped with NiO in this research and among other advantages of ZnO are significant physical and chemical stabilities, high catalytic activity, effective antibacterial and bactericide function, and intensive ultraviolet and infrared absorption.

II. EXPERIMENT

A. Sample Preparation

In our work, the solid state method is used in the preparation of a doped zinc oxide. The materials used for the synthesis are 100 g of Zinc oxide (99.7% purity) and 50 g of Nickel oxide (99.7% purity) all in powdered form. The Zinc oxide was obtained at Halishuaib Chemicals while the Nickel oxide was obtained at CEMAN Chemicals Ventures. During the preparation of the composites, 5 g of Nickel oxide was mixed with 25 g of Zinc oxide using pestle and mortar, the combination was grinded continuously using mortar and pestle for about 60 minutes for perfect homogeneity. The mixed sample was taken to furnace and was heated to about 1000^0C for appreciable reaction to take place. The same procedure was applied for 7.5 g of Nickel oxide and 22.5 g of Zinc oxide, 10 g of Nickel oxide and 20 g of Zinc oxide, 12.5 g of Nickel oxide and 17.5 g of Zinc oxide, and 15 g of Nickel oxide and 15 g of Zinc oxide. Five different mixtures with different proportions of Zinc oxide and Nickel oxide were prepared. The summary of materials composition is shown in Table 1. The prepared composites are then ready for characterization.

TABLE 1: COMPOSITION OF SAMPLES

Sample	NiO		ZnO		Total	
	(g)	*%*	*(g)*	*%*	*(g)*	*%*
A	5.0	16.7	25.0	83.3	30.0	100
B	7.5	25.0	22.5	75.0	30.0	100
C	10.0	33.3	20.0	66.7	30.0	100
D	12.5	41.7	17.5	58.3	30.0	100
E	15.0	50	15.0	50	30.0	100

B. Characterization

Characterization of the samples for dielectric loss factor and loss tangent were carried out using open ended coaxial probe HP85071C within a microwave frequency range of 8.2 to 12.2 GHz. The SEM machine Inspect S50 microscope was used to study the surface morphology.

II. RESULTS AND DISCUSSION

A. Loss Factor

The result shown in Figure 3.1 is the comparison of the loss factor obtained for all the samples under study.

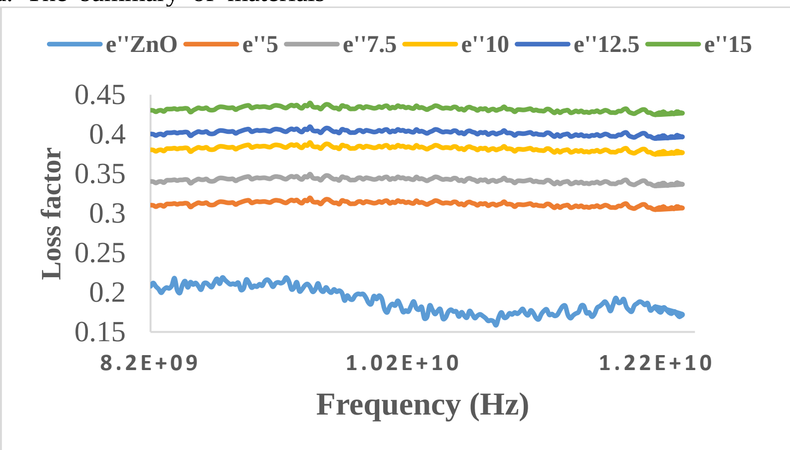

Fig. 3.1: Comparison of loss factor for pure ZnO and doped ZnO composites

Analysis shows that a total increase in loss factor of 0.23 representing 47.4 %. Further observation shows that loss factor decreases as frequency increases in all doped samples. At 8.2 GHz, it was observed that the pure ZnO have a loss factor of 0.20, 5 g doped ZnO have 0.31, 7.5 g doped ZnO have 0.34, 10 g doped ZnO have 0.38, 12.5 g doped ZnO have 0.40 and 15 g doped ZnO have 0.43. Also at 10.2 GHz, it was observed that the pure ZnO have a loss factor of 0.18, 5 g doped ZnO have 0.30, 7.5 g doped ZnO have 0.33, 10 g doped ZnO have 0.38, 12.5 g doped ZnO have 0.39 and 15 g doped ZnO have 0.43. Similarly at 12.2 GHz, it was observed that the pure ZnO have a loss factor of 0.17, 5 g doped ZnO have 0.30, 7.5 g doped ZnO have 0.33, 10 g doped ZnO have 0.37, 12.5 g doped ZnO have 0.39 and 15 g doped ZnO have 0.42. The dielectric loss factor decreases rapidly at low frequencies and becomes quite slow at high frequencies. The higher value of dielectric loss factor at lower frequencies is explained on the basis of space charge polarization (Aldar *et al.*, 2014; Muhammad *et* al., 2012). However, Sourav *et al.* (2014) reported that high value of dielectric loss factor at low frequency and almost constant value at higher frequency is due hopping contribution. Yakubu *et al.* (2015), reported that high value of dielectric loss factor at lower frequencies is attributed to the interfacial ionic polarizations due to localized ion motion within the sample. It is suggested that NiO consists of well conducting grains separated by thin insulating grain boundaries. This causes the localized accumulation of charges under the applied external field and thereby enhancing the space charge polarization.

B. *Loss Tangent of Samples*

Shown in Fig. 3.2 is the comparison of the loss tangent obtained for all the samples investigated. The loss tangent was calculated from the relation,

$$\tan\partial = \varepsilon'' / \varepsilon' \quad (4.1)$$

Where ε'' is the loss factor and ε' is the dielectric constant

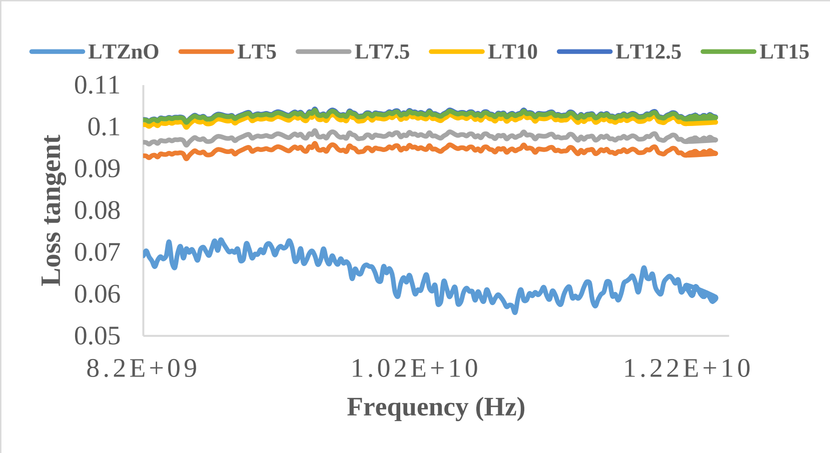

Fig. 3.2: Comparison of loss tangent for pure ZnO and doped ZnO composites

As shown Figure 3.2, the results are in good agreement with results obtained for the dielectric loss factor. This behaviour is attributed to good sample preparation technique leading to uncontaminated samples. The loss tangent was observed to have a total increase of 0.0324 representing 66.7 % increase after doping.

This shows an increase in loss tangent as NiO is added to the ZnO. It also revealed a decrease in loss tangent as the frequency increases. The decrease in loss tangent with frequency indicates dielectric dispersion is at low frequency region which is due to Maxwell-Wagner type of interfacial polarization in agreement with Koop's phenomenological theory (Yakubu *et al.*, 2015). The dielectric properties of all composites decreases as frequency increases. This may be attributed to space charge polarization (Ahmad, 2017). It is suggested that NiO consists of well conducting grains separated by thin insulating grain boundaries. At high frequencies, space charge carriers cannot line up their axes parallel tothe field and thereby reducing the contribution of space charge polarization (Ahmad, 2015).

C. *SEM*

Fig. 6(A), shows defined coarse morphology with nouniform particles that are agglomerated and forms irregular shaped particles. The darks spots indicates NiO with less density in distribution while the white spots indicates ZnO (Ahmad *et al*., 2015). Further observation shows a denser coarse morphology attributed to the increase in the filler content. From Fig. 6(B), the micrograph shows some whitish color image indicating the presence of zinc oxide, which are agglomerated by some doted NiO. Fig. 6(C) shows a micrographs with whitish colour image agglomerated with dark dotes of NiO (Karthikeyan *et al*., 2017). Due to the density of the sample in Fig. 6(D), more light were able to pass through it which is evident in the FTIR analysis. Observation on Fig. 6(E) shows that the zinc oxide and filler are sparsely dispersed through the matrix of the composite materials, and visible traces of white and black coloring are seen indicating the equal percentage of mixture in the sample (Yakubu *et al*., 2015).

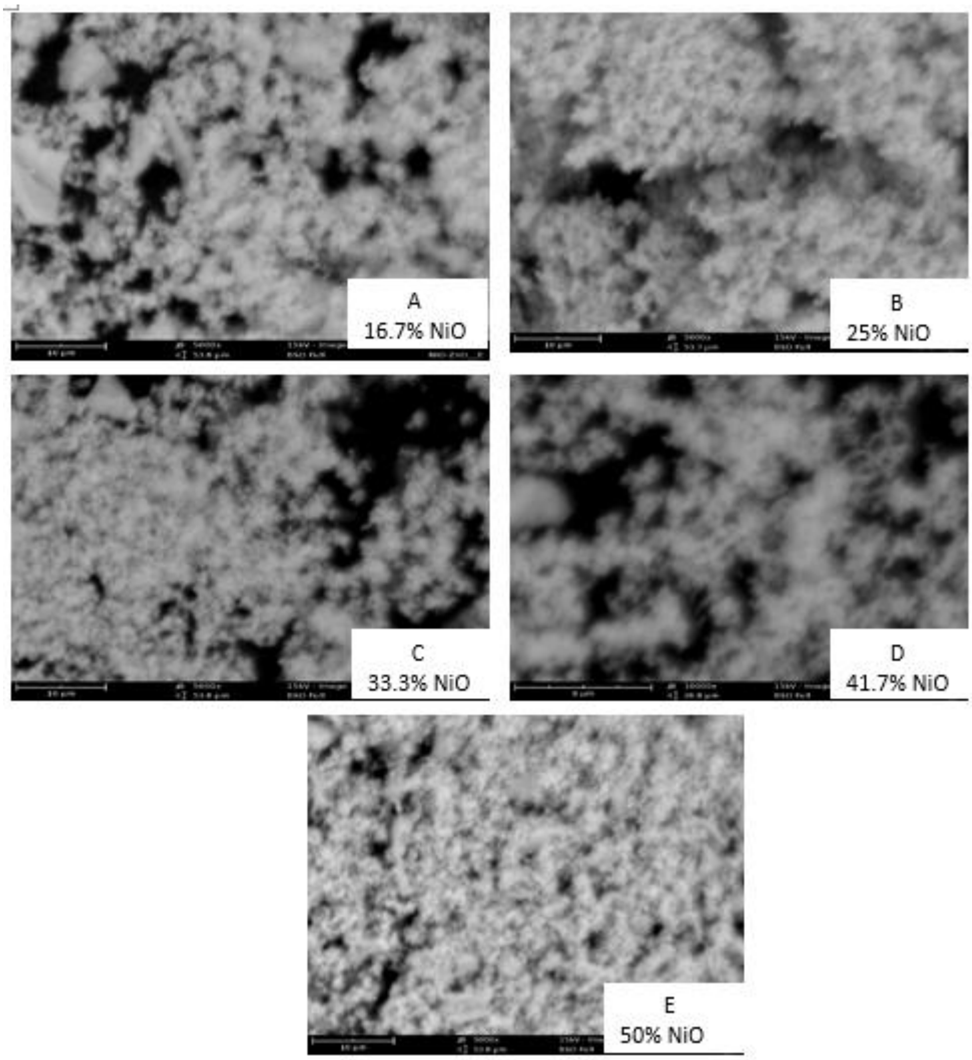

Fig. 3.3: SEM micrographs for doped composites

III. CONCLUSION

NiO/ZnO nanocomposite samples were prepared by a simple solid state method in this research. Investigations were carried out according to the objectives to determine the effect of NiO on the dielectric loss factor and loss tangent of ZnO using open ended coaxial probe (OECP). It was found that NiO can be used as a filler for improving dielectric loss factor and loss tangent with ZnO as a matrix. The SEM result revealed agglomeration of particles as doping increased for all compositions. The composite (NiO/ZnO) can also serve as a good agent for constructing capacitors and other dielectric based materials that can be used in constructing electronic and telecommunication gadgets.

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