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Research Article

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Tuning in Dielectric and Electrical Transport Properties of CuFe_2O_4 with Incorporation of Metal oxides (ZnO, WO_3 & MgO) Nanostructures

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Abstract

Copper ferrite (COF) nanocomposites with different weight fractions (10 to 50 wt. %) of metal oxides (MOs; M= Zn, Mg & W) were synthesized by powder mixing method. The phase analysis and microstructure were examined by XRD, SEM and TEM techniques. The XRD analysis showed the formation of required nanocomposites without any other impurities. The examination of microstructure revealed the different morphology like; spherical, nano-discs, nano-plates and nano-spindle etc. The behavior of dielectric constant (ϵ'), and dissipation factor ($\tan\delta$), of $(\text{MOs})_x/(\text{COF})_{1-x}$ are consistent with Maxwell-Wagner polarization and Koop's phenomenological theory. Moreover, the ϵ' & $\tan\delta$ show decreasing behavior with incorporation of MOs nano-structures. The a.c. conductivity with frequency follow the dynamical law $\sigma(\omega) = A \omega^s$. Finally, the incorporation of MOs wt. fractions reduce the conductivity due to substitution of Fe^{3+} and Fe^{2+} ions by metal cations (Zn^{2+} , Mg^{2+} and W^{3+}) from lattice site.

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1. Introduction

Spinel ferrites, having compositions $\text{Fe}_2\text{O}_3\text{-MO}$, where M is generally a transition metal ion, encompasses a gigantic potential in different areas of science and technology [1-15]. The chemical formula of spinel ferrite is $\text{M}^{2+}\text{Fe}_2^{3+}\text{O}_4$, where M and Fe are divalent and trivalent cations respectively. The properties of ferrites depend on many factors like; composition, crystal structure, synthesis route and morphology. Moreover the chemical and physical properties of ferrites depend on distribution of M^{+2} and Fe^{+3} cations reside on the tetrahedral “A” and octahedral “B” sites. Ferrites revealed significant magnetic losses due to their ferromagnetic natural resonance (FMR). Owing to these FMR properties, they are one of the best materials for microwave absorption in required frequency [20, 21]. Among the spinel ferrites, COF is the most efficient ferrimagnetic ceramic that used in magnetic data storage and electromagnetic devices due to their magnetic, dielectric and semiconducting properties [17-19]. The COF exhibit prominent properties like moderate value of saturation magnetization (M_s) and coercivity (H_c), high value of magneto crystalline anisotropy (H_k) and chemical stability at nanoscale as compared to other spinel ferrites [22].

However, pristine ferrite restricts their applications in different areas of science and engineering due to their limited electrical and magnetic properties. However the material for required applications e.g. electronics, medicines, defence; food industry, textiles and sensors devices can be synthesized by tuning their electrical, chemical and magnetic properties. This tuning is only possible by doping of different elements in ferrites structure at desired concentration. The doping only tunes the properties at limited level of interest. To tailor the properties on large scale, the nanocomposites of ferrites with different materials like; metal oxides, conducting and insulating polymers, hard and soft ferrites, allotropes of carbon like graphite, graphene and multi walled carbon nano-tubes etc. can be prepared. These

nanocomposites tailor the inherent properties of ferrites and induce required electrical and magnetic properties [23-25].

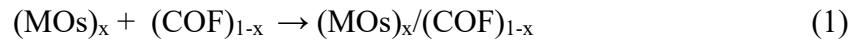
In this study, we chemically synthesized $(\text{MOs})_x/(\text{COF})_{1-x}$ nanocomposites and study their structure, dielectric, and electrical transport properties in detailed. The MOs nanostructures contents in COF produce heterogeneous system of two different permittivity zones ϵ_1 and ϵ_2 , having conductivities σ_1 and σ_2 . This difference in conductivity produces accumulation of charges in the presence of applied A.C. field. These charges enhance the interfacial polarization and hence interact with field and dissipate more energy in the form of heat. Hence, this multiphase system is beneficial for absorption of electromagnetic (EM) energy due to enhancement of interfacial polarization. The main focus of present research is to tune the dielectric properties of COF with incorporation of different MOs nanostructures.

2. Experimental Section

2.1 Synthesis Method

The nanocomposites of $(\text{MOs})_x/(\text{COF})_{1-x}$ were prepared by using chemical impregnation method. The analytical grade of chemicals used are; $\text{CuCl}_2 \cdot 2\text{H}_2\text{O}$, $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$, ZnCl_2 , $\text{MgSO}_4 \cdot 7\text{H}_2\text{O}$, Na_2WO_3 , HCl and NaOH . Firstly, for the synthesis of COF nanoparticles 0.2M ($\text{CuCl}_2 \cdot 2\text{H}_2\text{O}$) and 0.4M ($\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$) solution was formed separately in distilled water under vigorous magnetic stirring. Moreover, the 3M solution precipitating agent (NaOH) added drop wise in former metal chlorides solution till PH-10~11 was obtained. The black brown precipitate was formed and brought to reaction at 80 °C for one hour under strong magnetic stirring. The obtained colloidal suspension was cooled down to room temperature and centrifuge to separate the precipitate from the solution. The obtained precipitate still contains water contents, which was removed by keeping the sample in an oven at 100 °C for 10 hours. The black brown sample was obtained which was grinded to fine powder by using mortal pestle.

Secondly, (MOs) nanostructures were prepared by using 0.2M solution of metal chlorides. The, 2M solution of NaOH and HCl (for synthesis of WO₃) was separately prepared and added dropwise in metal chlorides solution to form a colloidal suspension till PH-10~11 was obtained. This colloidal suspension was brought to reaction at 60 °C for one hour and separated by solution by using the centrifuge machine. This precipitate was dried by heating the sample in an oven at 100 °C for 10 hours. The dried sample was grinded in to fine powder by mortal pestle. The nanocomposites of (MOs)_x/(COF)_{1-x} were obtained by following the empirical formula;



2.2 Characterization Techniques

The (MOs)_x/(COF)_{1-x} nanocomposites were characterized by using different experimental techniques. The phase analysis was elucidated by x-ray Diffraction (Model JDX-11, of Joel Company Ltd., Japan). The SEM (Model; Tescan-Vega-03) was used to study the surface morphology and particle size. To calculate the grain size and effect of MOs percentage (10-50 wt. %) on morphology of nanostructures, the TEM (Model: Philips EM-400 with electron acceleration voltage 60 kV) was also used. Finally, the dielectric, and electrical transport properties were measured by LCR Meter Bridge (Model; LCR Meter IET 7600 Plus Precision).

3. Results and Discussions

3.1 Structure and Morphology Analysis

The phase analysis was confirmed by using the x-ray diffraction (XRD) technique. Fig.1 shows the XRD pattern of (ZnO)_x/(COF)_{1-x} nanocomposites. It is evident from Fig.1, that the intensity of peaks with (hkl) planes (101), (100) and (110) increases with impregnation of ZnO contents in nanocomposites. This intensification of intensity is attributed to increase in charge density of Zn²⁺ in these planes that enhance the constructive

interference after scattering the x-rays and hence results in the amplification of intensity. Moreover, the intensity peaks revealed a shift towards lower 2θ angle which is ascribed to slightly increases in ionic radius of Zn^{2+} ($0.74 \text{ \AA}$) as compared to Cu^{2+} ($0.72 \text{ \AA}$). According to Bragg's Law; $d \sim \frac{1}{\theta}$, the larger ionic radius of ions enhance the interplanar spacing between the diffraction planes and hence reduce the diffraction angles. Therefore with impregnation of ZnO contents shift the diffraction peaks toward lower 2θ values. Moreover, the intensity of COF phase mentioned with (*) in Fig.1 diminishes; this lessening in intensity is due to decrease in Cu^{2+} ions in this grains that eventually reduce the intensity. The Debye-Sherrer's formula as shown in Eq.3 was used to measure the crystallite size from different diffraction peaks and lies in the range of 27 to 79 nm due to increase in crystal growth [15].

$$D = \frac{0.9\lambda}{\beta \cos \theta} \quad (3)$$

Where λ is the wavelength of x-ray, β is full width at half maximum of the peak at diffracting angle θ .

Fig.1 (b) shows the diffraction pattern of $(\text{WO}_3)_x/(\text{COF})_{1-x}$ nanocomposites; the observed at 2θ values with (hkl) planes are; 23° (001), 24.15° (200), 26.7° (120), 34° (220), 52.41° (221), 48° (240), 55.7° (132) and 62.25° (422) according to JCPDS card no. 32-1395 [32]. There is decrease in intensity of COF phase decreases marked with symbol (*) due to suppression of COF grains by WO_3 . Moreover, at higher concentration of WO_3 , addition of new peaks appeared; and their intensity also increased due to presence of W^{3+} ions that raise the scattering of x-raye in WO_3 grains. The higher concentration also broadens the peaks which are ascribed to decrease in nonmagnetic contents that quenche the magnetic interaction between different magnetic ions of COF. The particle size was calculated from diffraction peaks using Eq.3 and lies in the range of 23-89 nm [27].

Finally, the XRD pattern of $(\text{MgO})_x/(\text{COF})_{1-x}$ nanocomposites shown in Fig.1 (c) shows the amplification in MgO diffraction peak (marked with arrow). This strengthening in peaks intensity is attributed to increase in charge density of Mg^{2+} ions. There is a little shift in peaks position towards higher 2θ values was observed and was made prominent by drawing straight lines. This shift is attributed to increase in ionic radius of Mg^{2+} ion ($0.79 \text{ \AA}$) as compared to Cu^{2+} ion ($0.73 \text{ \AA}$) and consistent with Eq.2. The Eq.3 was used to measure the crystallite size l and found in the range of 21 to 39 nm. This increase in particle size is due to agglomeration of MgO grains

The microstructure analysis of prepared nanocomposites were analyzed by using SEM and TEM techniques. Firstly, the SEM images of $(\text{ZnO})_x/(\text{COF})_{1-x}$; $x = 10$ to 50 wt.% nanocomposites, shown in Fig.2 revealed the nano-plates like shape with thickness 30-80 nm, which are separated from each other. At higher concentration of ZnO (50%) nano-plates agglomerated and exhibit a nanoflowers like morphology as can be observed in red circle shown in Fig.2 (a). Secondly, the $(\text{MgO})_x/(\text{COF})_{1-x}$ micrographs shown in Fig.2 (b) revealed the spherical morphology having size 19-43 nm. The higher concentration of MgO enhance the agglomeration between the particles because the Cu^{2+} is a weak magnetic ion having magnetic moment $1\mu_B$. Furthermore, 50wt.% concentration the morphology change from spherical to spindle like shape due to rapid increase in growth of particles along x-direction as compared to y and z directions. Thirdly, Fig. 2(c) shows the SEM images of $(\text{WO}_3)_x/(\text{COF})_{1-x}$ nanocomposites having spherical morphology with size 40-118 nm. At lower concentrations of WO_3 , the morphology remains spherical, but as the contents increase the agglomeration becomes dominated and produce increase in grain $> 100 \text{ nm}$.

Finally, to estimate the grain size and morphology of prepared nanocomposites, the TEM images were taken at same resolution only for 50 wt.% of MOs contents in nanocomposites. The TEM images revealed the spherical morphology of COF with size 30 nm. Moreover, he

higher contents (50.wt.%) of MOs , deteriorate the spherical morphology as shown in 2 (a-c). This deterioration can be attributed due to induction of defects like; bond angle , bond length and the force exerted by high density material at the edges and interface of low density material.

3.2 Dielectric Properties

The dielectric constant (ϵ) is an important parameter that measures the extent of different types of polarization like; space charge, dipolar, ionic and electronic, etc. contributing in the material. The dielectric properties of material can be elucidated by measuring the dielectric parameters. The real part (ϵ') of dielectric constant measure the energy stored in the material while the imaginary parts (ϵ'') determine the energy dissipation of electromagnetic energy in the form of heat [4, 14, 19]. Figure-4 (a-c) shows the ϵ' of COF and (MOs)_x/(COF)_{1-x}; x; varies from 10 to 50 wt.% nanocomposites in the frequency range of 1kHz-2MHz (in logarithmic scale). It is observed from fig.4 that ϵ' first decreases significantly with frequency and becomes constant at $\geq \log 4$ Hz.

The higher value of ϵ' at low frequency is achieved due to activation of space charge polarization. The diminution in ϵ' with increasing frequency is ascribed to the actuality that at higher frequencies the contribution of dominated polarization becomes low because the permanent and induce dipole does not follow the fast variation of electromagnetic field [23]. The Koop's model and Maxwell's Wagner theory were used to elucidate the dielectric response of (MOs)_x/(COF)_{1-x} nanocomposites which state that these nano-composites consist of small conducting grains, separated by insulating grain boundaries. The grains become more active at lower frequencies whereas; the resistive grain-boundaries restrain no such response and causing electrons to pile up at the interface and produced the interfacial polarization. Thus, at lower frequencies the movement of electron occur from grain to grain-boundary, consequently, results in larger values of ϵ' at lower frequencies.

Moreover, this behavior is quite different at higher frequencies because it is difficult for the electrons to move between Fe^{+2} and Fe^{+3} from conducting grains towards high resistive grain-boundaries. Thus, at high frequency, the electron reaches at their destination by hopping mechanism, and shows minimum values ϵ' .

The compositional analysis were also studied by varying the MOs contents 10~50 wt.% . Fig.4 (a) shows that the value of ϵ' decrease with ZnO contents in entire frequency range and calculated values at frequency 1k Hz is shown in table-1. This decrease in the value of ϵ' with ZnO contents is ascribed to the increase in particle size as observed by SEM images in Fig.2(a). There are two mechanism can be involved for this decrease in ϵ' . Firstly, the density of a material play an essential role, i.e., the high porosity results in low ϵ' and high dielectric losses [28]. Secondly, the presence of ZnO phase; enhance the particle size and hence the large number of grain boundaries are formed in the material. The insulating nature of grain boundaries reduces the hopping of charge carriers between Fe^{2+} and Fe^{3+} and consequently a decline in ϵ' was observed [29]. Similar trend with ϵ' was also observed up to 40 wt.% of MgO. But a higher value in ϵ' (44.78) was observed at 50 wt.% of MgO. As the ionic radius of Mg^{2+} is greater than the ionic radius of Fe^{2+} and Fe^{3+} . At 50 wt. percent of MgO contents substitute the Fe^{2+} and Cu^{2+} ion from lattice sites; as a result of which, the lattice is expanded. This increase the Fe-O bond lengths at B site enhance the atomic polarizability and consequently the ϵ' [9, 17]. Furthermore, a decreasing behavior in ϵ' for $(\text{WO}_3)_x/(\text{COF})_{1-x}$ was also observed up to 30 wt.% of WO_3 . The secondary phase is conscientious for this decrease in ϵ' which restrict the motion of charges at the grain boundaries and reduce the polarization and hence ϵ' . But at 40 wt.% concentration the value of ϵ' increases up to 81.19 which is higher than pristine COF, this higher value is due to increase in bond length of Fe-O at B site which enhance the polarizability and hence ϵ' . But at 50 wt. % the ϵ' once again decreases because at this concentration the quantity of W^{3+} ions increases which substitute the Fe^{2+} and

Fe³⁺ ions from the octahedral sites. This substitution of Fe³⁺ and Fe²⁺ ions from lattice sites reduce the hopping of charges between Fe³⁺ and Fe²⁺ and consequently the ϵ' decreases accordingly as shown in Fig.4 (b).

The variation of dielectric loss tangent ($\tan\delta$) with frequency is shown in Fig.5 and it can be observed that $\tan\delta$ decreases with applied frequency [31-32]. This decrease in $\tan\delta$ with frequency is ascribed to the fast variation electric field as compared to dipoles and electromagnetic (EM) radiation pass through the material without interaction. Moreover,, with further increase in frequency a constant behavior of $\tan \delta$ with frequency was observed. Moreover, there is a resonance peak was observed at 50 wt. % of MgO, which is due to the matching of hopping frequency of electrons with the applied frequency and this frequency is called a resonance frequency “ f_r ” . [33, 34].

3.3 Electrical Transport Properties

The A.C. conductivity (σ_{ac}) of prepared nanocomposites in the logarithmic scale is shown in Fig.7. The conductivity of material depends on many factors like; grain size, distribution of Fe³⁺ and Fe²⁺ ions, applied frequency and composition of materials etc. The a.c. conductivity of all samples show an increasing trend with frequency and follow the dynamical law $\sigma(\omega) = A \omega^s$, where A is a parameter with the units of conductivity and s is the slope of the linear plot [34-36].

The σ_{ac} was calculated by using the electrical parameters given as below [15];

$$\sigma_{ac} = 2\pi\epsilon_0\epsilon'f \tan \delta \quad (4)$$

Where f is frequency of applied field, ϵ_0 is the vacuum, ϵ' is dielectric constant, $\tan\delta$ is the dissipation factor, and σ_{ac} is the a.c. conductivity [15, 24]. Figure-7 shows that, σ_{ac} shows a linear trend with frequency, representing the small polaron conduction. This linear increase in σ_{ac} with frequency is ascribed to conduction by hopping of charge carriers between Fe²⁺ and Fe³⁺ [19].

The effect on σ_{ac} with MOs contents show a non-systematic behavior. For example with enhancement of ZnO contents (10 to 50 wt. %) decrease the conductivity values. This decrease in conductivity values is attributed to substitution of Fe^{2+} ions by Zn^{2+} ions from the lattice sites. This substitution reduces the chance of electron jumping between Fe^{2+} and Fe^{3+} , and consequently decreases the σ_{ac} [38]. Furthermore, the Zn^{2+} ion contains strong coulomb field as compared to Cu^{2+} ion and confined the charge carriers to prevent the jumping of electrons in its neighbourhood and hence reduces the conductivity accordingly. Similarly, the enhancement of MgO & WO_3 contents in COF also reduces the concentration of Fe^{2+} & Fe^{3+} at octahedral sites to balance the charge, strains, and crystallographic energy in the material as shown in Fig.7. It can be observed from Fig.8, that the ZnO show decreasing behavior up to 50 wt. percent concentration in COF. On the other hand, MgO show decrease in conductivity up to 40 wt.% and an sudden increase in conductivity value (-4.91) at 50 wt.% of MgO was observed. This increasing behavior in σ_{ac} is attributed to the incorporation of impurity states between band gap that results in decreases in band gap and consequently enhance the conductivity. Lastly, the ZnO show increase in conductivity at 30 and 40 wt.% in COF which is due to increase in grain size that lessen the oxygen rich insulating layer and allow the conduction of charges within material.

Conclusion

The $(MOs)_x/(COF)_{1-x}$ nanocomposites were prepared by powder mixing method in which required phases are coexistent. The structure and morphology was examined by XRD, SEM and TEM respectively. The dielectric and electrical transport properties were also studied systematically. The ϵ' and $\tan \delta$ show the normal dispersion of ceramic material in the given frequency range. The electrical transport properties show decreasing trend with increase in MOs concentration because the secondary phase hinders the movement of charge carriers and substitution of M^{2+} (Zn^{2+} , Mg^{2+} and W^{3+}) ions in place of Fe^{3+} ion reduce the

jumping frequency of electron between Fe^{2+} and Fe^{3+} . This decrease in ϵ' with incorporation of MOs nanostructures is suitable for high frequency application. This low value of ϵ' increases the penetration depth of electromagnetic waves and decreases the skin effects. Hence these nanocomposites can be a potential candidate for absorption of microwave in required frequency range.

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- **Conflict of Interest**

The authors have claimed that the work reported in this article will not generate any conflict of interest or economic disputes among them.

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- **Competing Interest**

There is no competing interest in this research work .All research work is original and carried out in our Nano science and Technology Lab.

- **Research Data Policy and Data Availability Statements**

The data sets generated during and/or analyzed during the current study are available from the corresponding author on reasonable request.

Figures

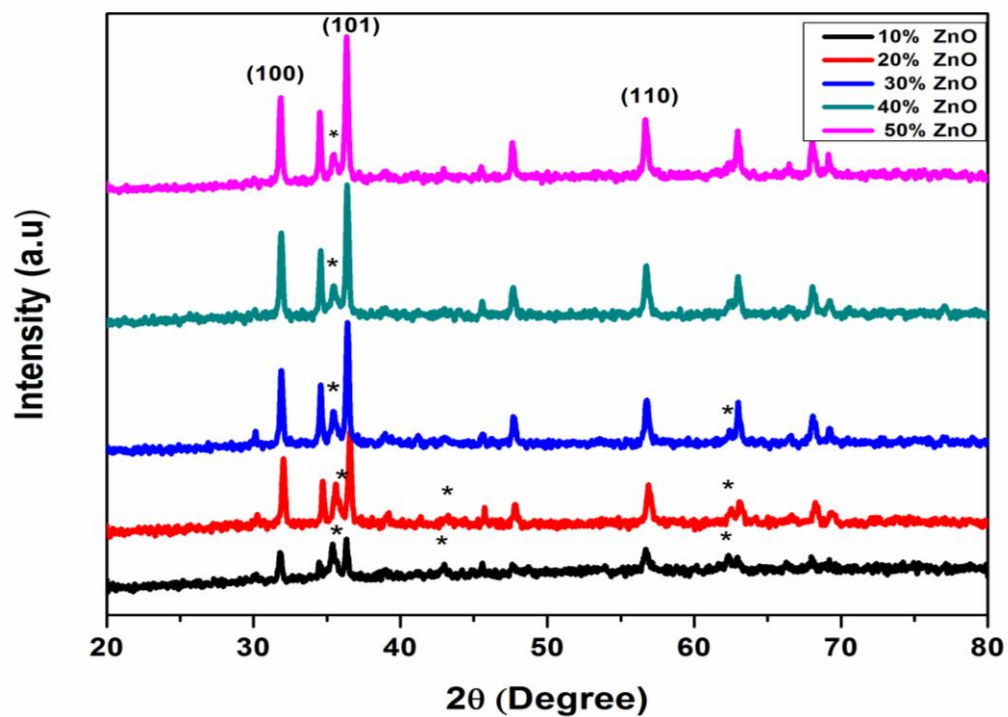


Figure-1(a): XRD pattern of $(\text{ZnO})_x/(\text{COF})_{1-x}$; $x=10$ to 50 wt.% nanocomposites

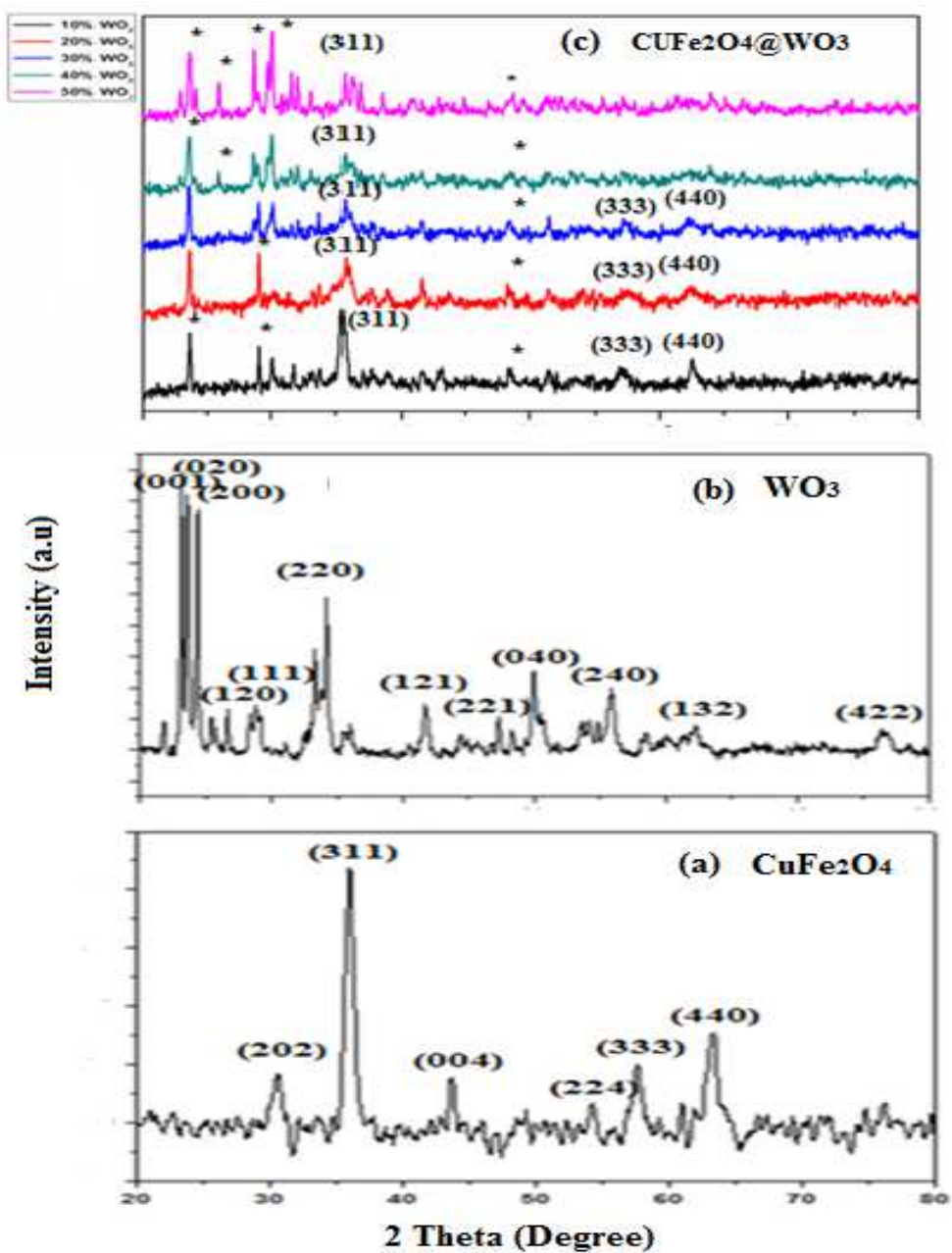


Figure-1(b): XRD pattern of $(\text{ZnO})_x/(\text{COF})_{1-x}$; $x=10$ to 50 wt.% nanocomposites

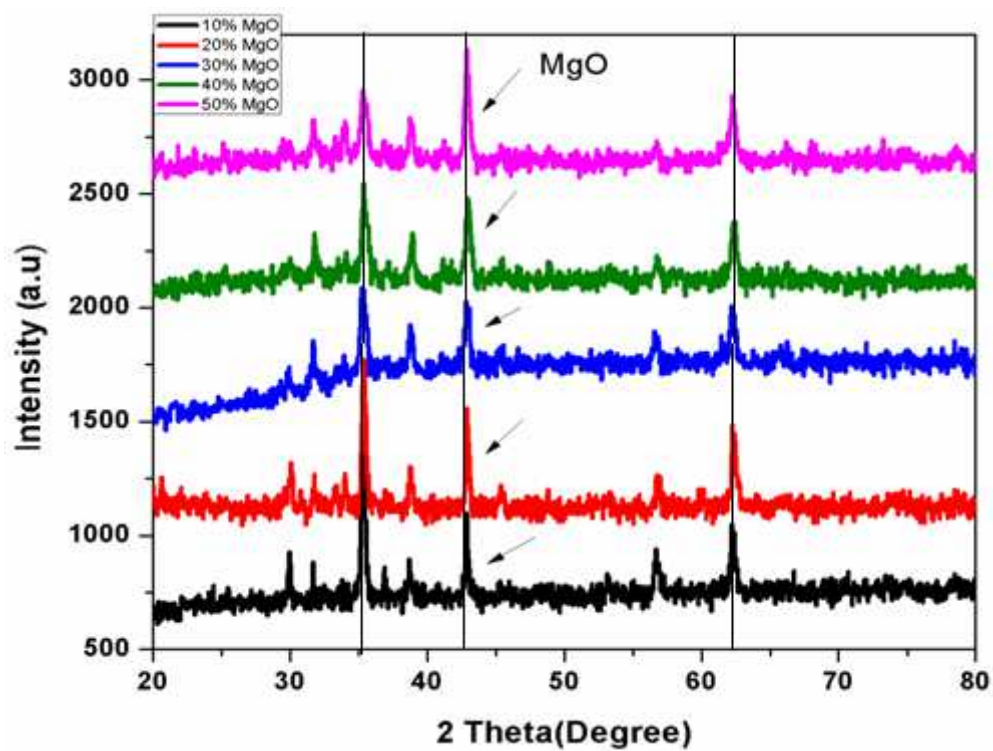


Figure-1(c): XRD pattern of $(\text{WO}_3)_x/(\text{COF})_{1-x}$; $x=10$ to 50 wt.% nanocomposites

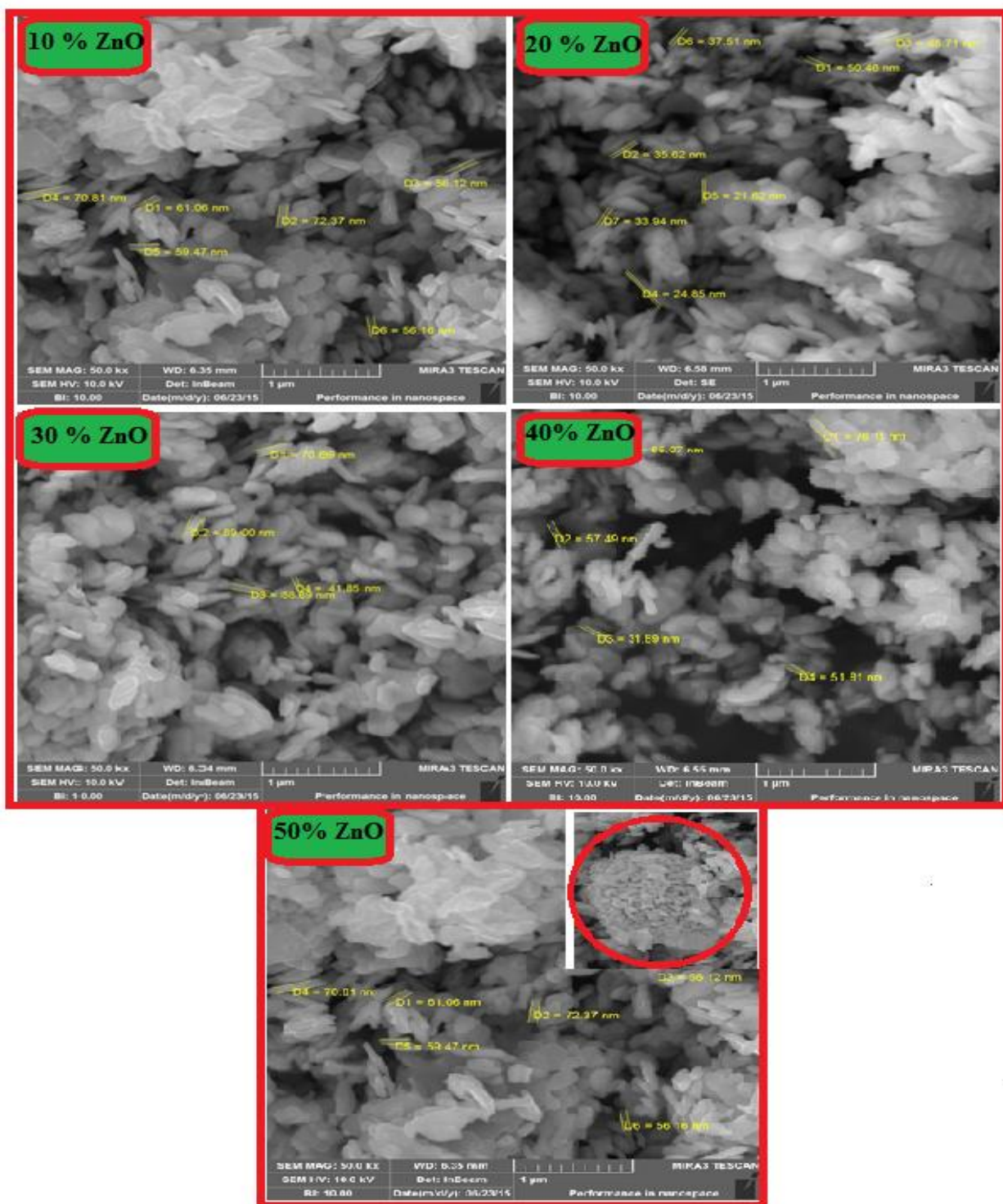


Figure- 2(a): SEM images of $(\text{ZnO})_x/(\text{COF})_{1-x}$; $x=10\%$ to 50% wt.% nanocomposites

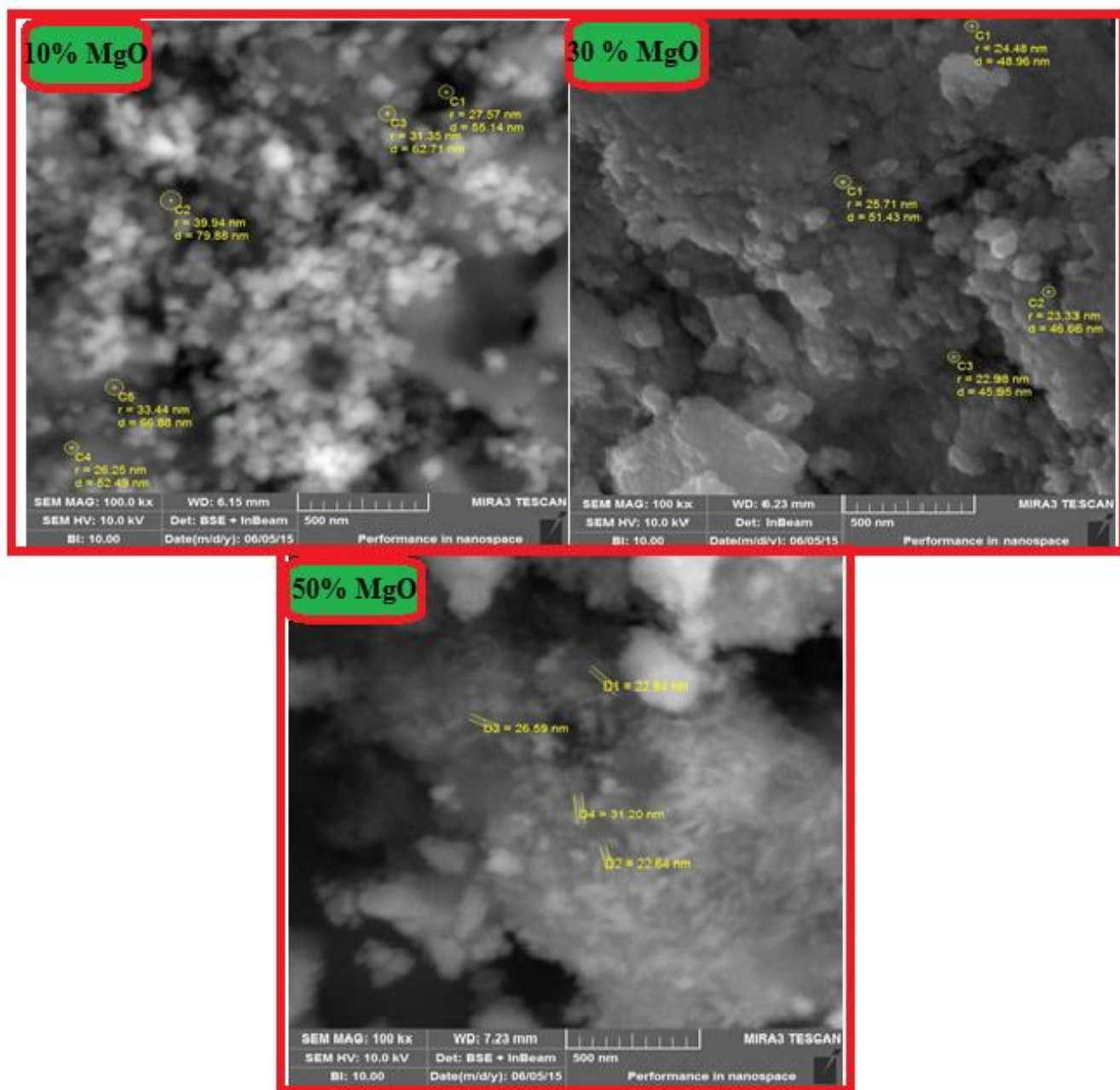


Figure- 2(b): SEM images of $(\text{MgO})_x/(\text{COF})_{1-x}$; $x=10\%$ to $50 \text{ wt.}\%$ nanocomposites.

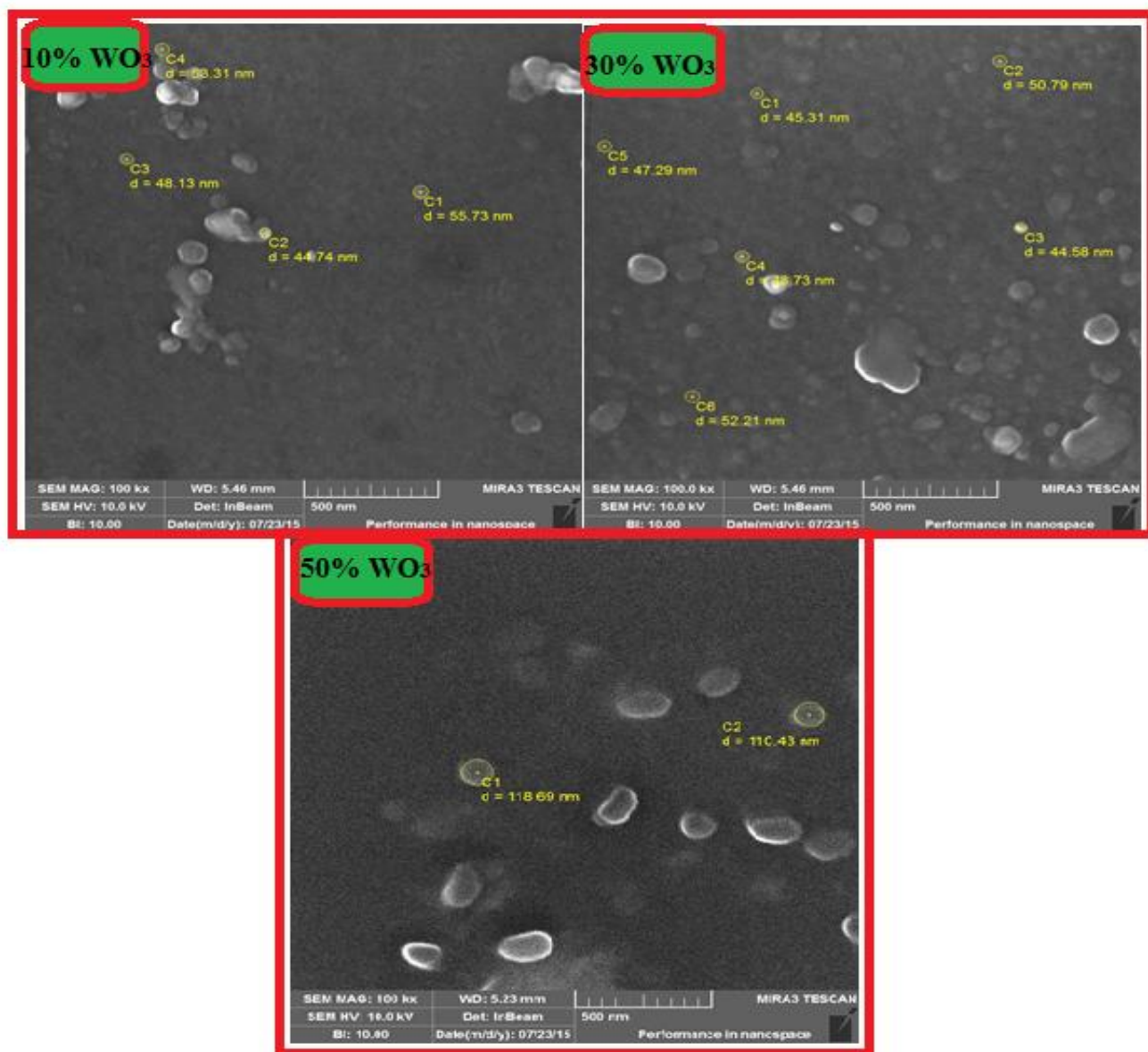


Figure 2 (c): SEM images of $(\text{WO}_3)_x/(\text{COF})_{1-x}$; $x=10\%$ to $50\text{ wt.}\%$ nanocomposites.

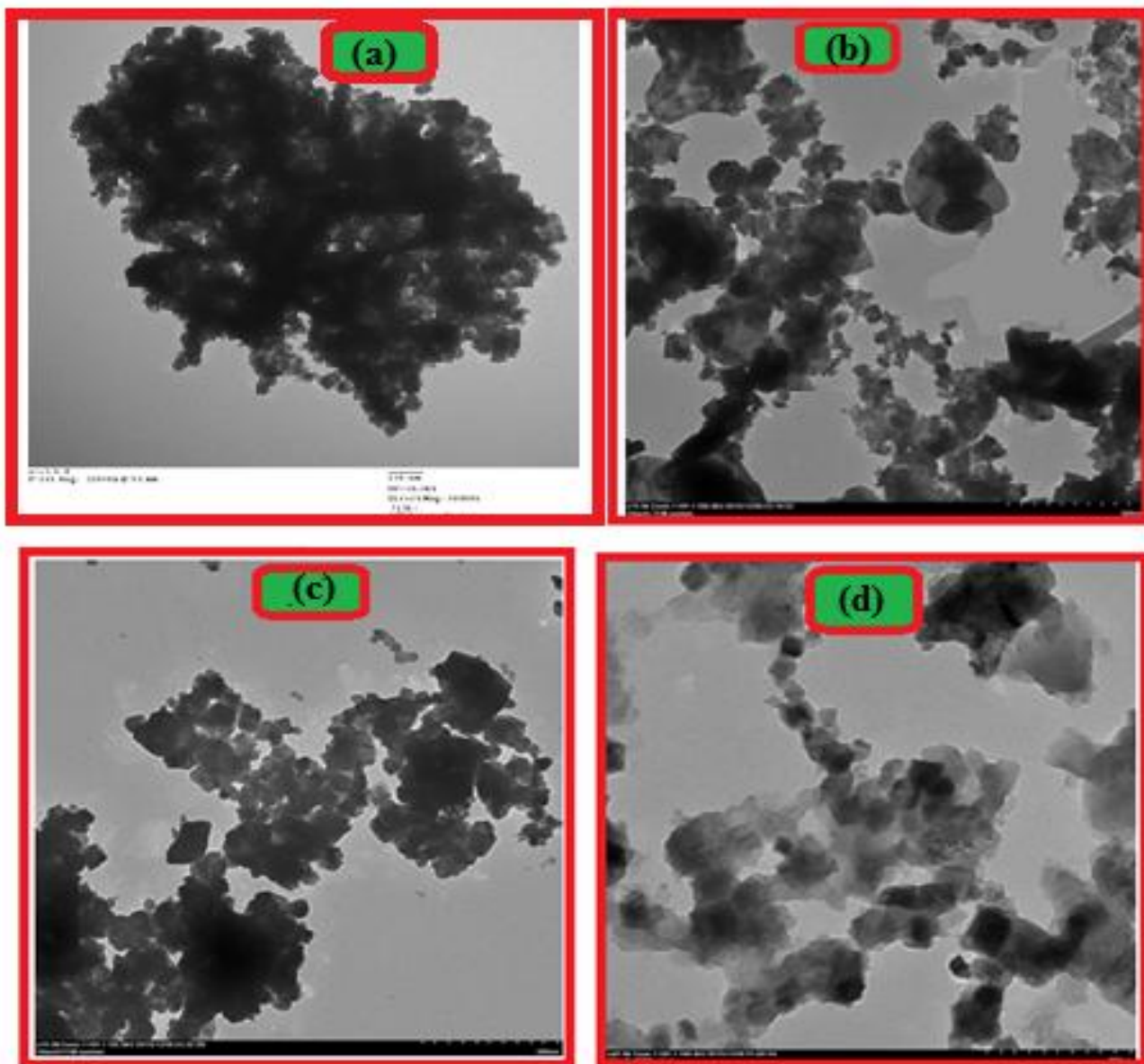


Figure 3: TEM images of (a) Pristine COF (b) 50 % ZnO (c) 50% WO₃ (d) 50% MgO

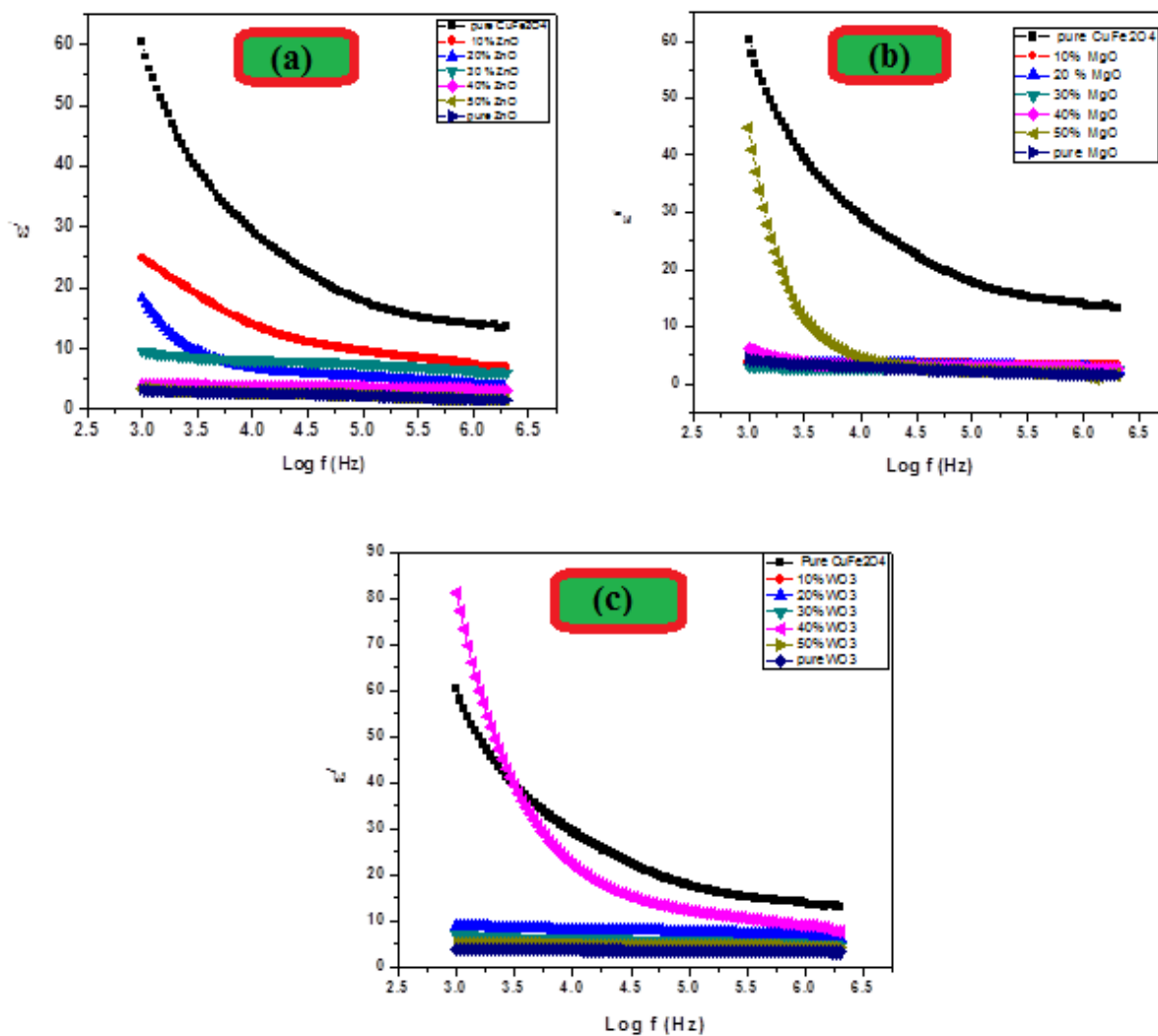


Figure- 4: Variation of dielectric constant (ϵ') with $\log f$: (a) $(\text{ZnO})_x/(\text{COF})_{1-x}$ (b) $(\text{MgO})_x/(\text{COF})_{1-x}$ (c) $(\text{WO}_3)_x/(\text{COF})_{1-x}$; $x=10$ to 50 wt.% nanocomposites

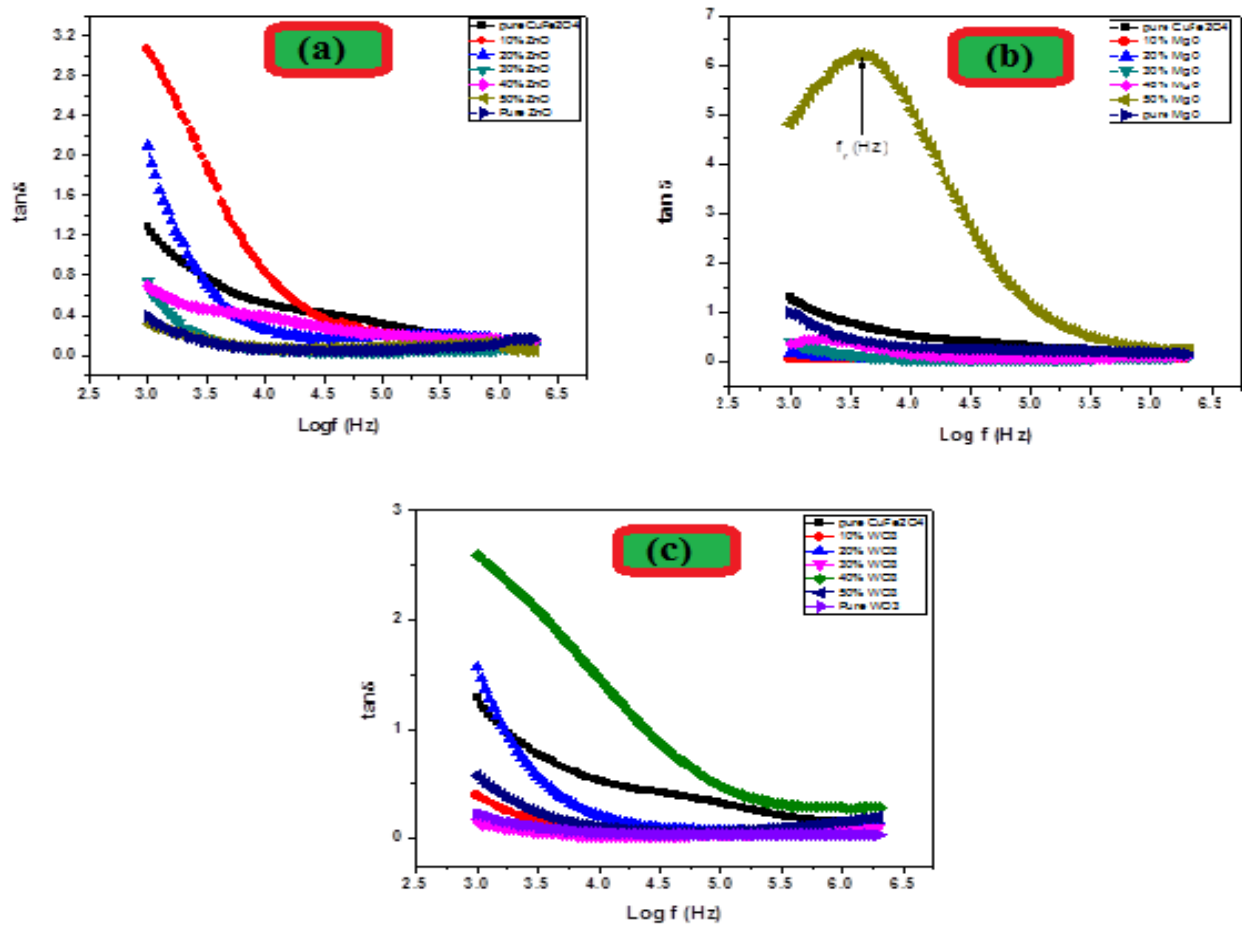


Figure 5: Variation of dielectric loss tangent ($\tan \delta$) with $\log f$: (a) $(\text{ZnO})_x/(\text{COF})_{1-x}$ (b) $(\text{MgO})_x/(\text{COF})_{1-x}$ (c) $(\text{WO}_3)_x/(\text{COF})_{1-x}$; $x = 10$ to 50 wt.% nanocomposites.

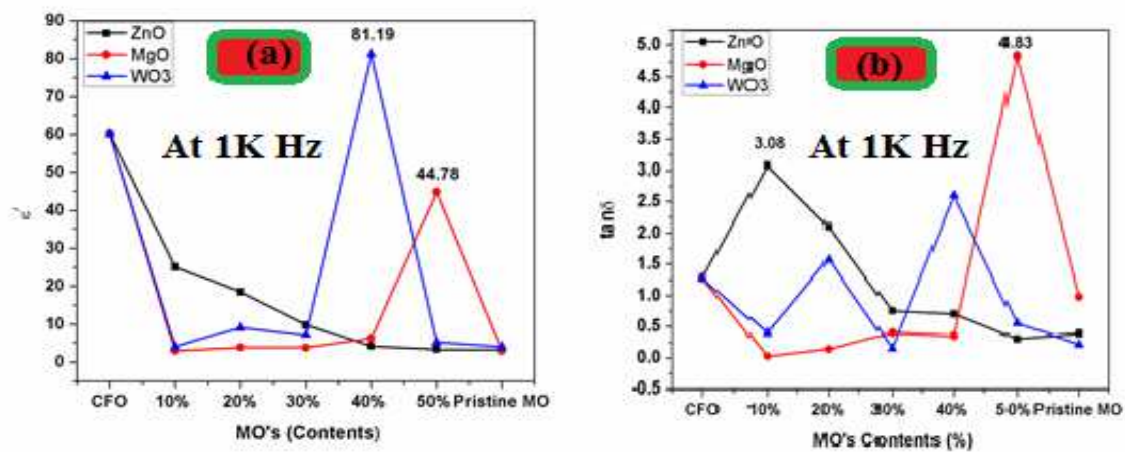


Figure 6: Variation of dielectric constant (ϵ') & tangent loss ($\tan \delta$) with MOs contents at 1kHz.

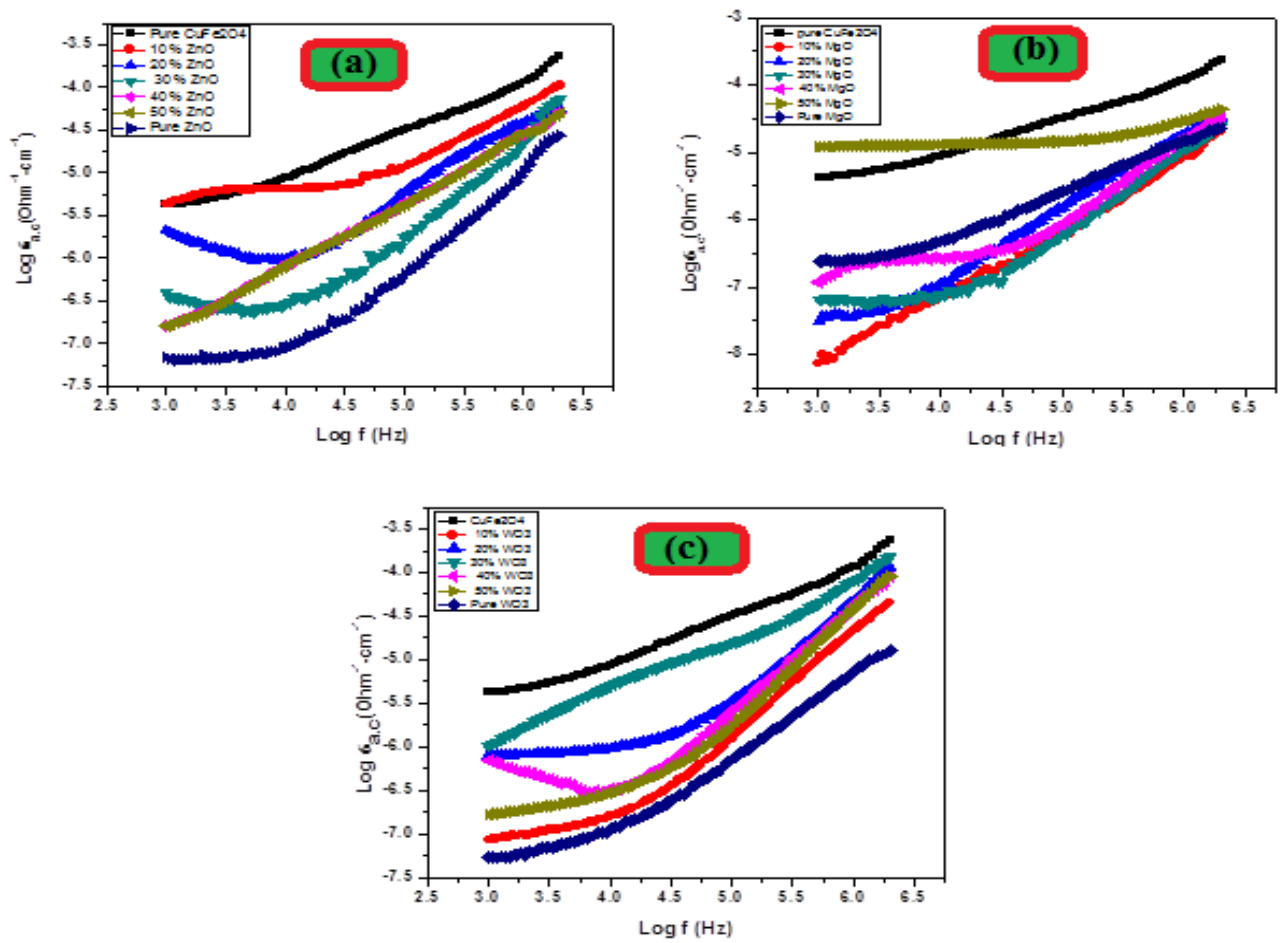


Figure7: Variation of $\log \sigma_{a,c}$ with $(\log f)$: (a) $(\text{ZnO})_x/(\text{COF})_{1-x}$ (b) $(\text{MgO})_x/(\text{COF})_{1-x}$ (c) $(\text{WO}_3)_x/(\text{COF})_{1-x}$; $x=10$ to 50 wt.% nanocomposites.

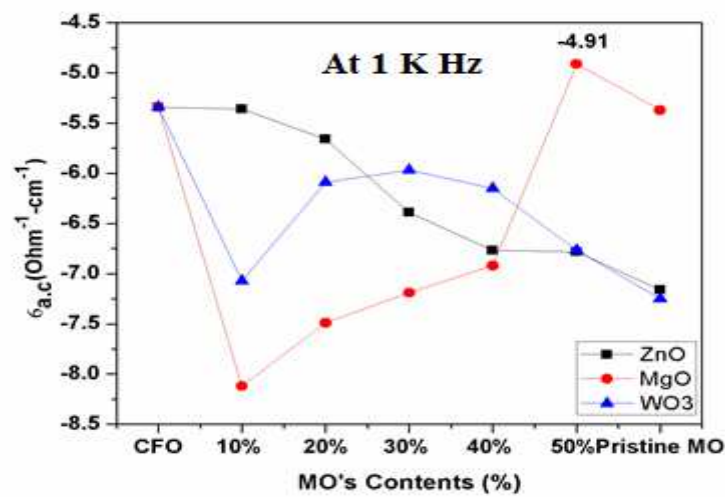


Figure 8: Variation of $\log \sigma_{a,c}$ versus MOs contents at 1 kHz.

Table-1: Dielectric Parameters of (MO)_x/(COF)_{1-x} (x=10 to 50 wt.%) nanocomposites at 1 k Hz

Composition	ϵ'	$\tan \delta$	$\sigma_{a.c}$
Pristine COF	60.15	1.28	-5.34
(ZnO) ₁₀ /(COF) ₉₀	25.15	3.08	-5.36
(ZnO) ₂₀ /(COF) ₈₀	18.47	2.10	-5.66
(ZnO) ₃₀ /(COF) ₇₀	9.83	0.75	-6.39
(ZnO) ₄₀ /(COF) ₆₀	4.18	0.70	-6.77
(ZnO) ₅₀ /(COF) ₅₀	3.35	0.31	-6.78
Pristine ZnO	3.15	0.39	-7.16
(MgO) ₁₀ /(COF) ₉₀	2.99	0.02	-8.12
(MgO) ₂₀ /(COF) ₈₀	3.83	0.14	-7.49
(MgO) ₃₀ /(COF) ₇₀	3.73	0.40	-7.19
(MgO) ₄₀ /(COF) ₆₀	6.20	0.35	-6.92
(MgO) ₅₀ /(COF) ₅₀	44.78	4.83	-4.91
Pristine MgO	2.94	0.98	-5.37
(WO ₃) ₁₀ /(COF) ₉₀	3.96	0.39	-7.07
(WO ₃) ₂₀ /(COF) ₈₀	9.15	1.57	-6.09
(WO ₃) ₃₀ /(COF) ₇₀	7.16	0.15	-5.97
(WO ₃) ₃₀ /(COF) ₇₀	81.19	2.6	-6.15
(WO ₃) ₃₀ /(COF) ₇₀	5.12	0.58	-6.77
Pristine WO ₃	4.0	0.21	-7.25