Physical and Optical Spectroscopic Properties of Glass Doped with Multi-Transition Metal Oxides

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Abstract

Glass doped with transition metal oxides plays a vital role in various technological and industrial applications. The incorporation of metal oxides is important to improve the optical glass properties. The chemical composition of the network is 55 B2O3,35 ZnO, (10-x) Na2O, x Co_2O_3, y CuO (x=0, 0.05, 0.1, 0.15, 0.2, 0.25; y=0.1, 0.15, 0.20, 0.25, 0.30, 0.35). The preparations of glass samples were using the conventional melting-quenching technique. The physical and optical characterizations of the prepared glasses were studied using the UV-VIS spectroscopic technique to examine the structure of the preparing glass networks. The XRD technique assured the amorphous nature of the network. The Archimedes method measures the density and molar volume of a network. The influence of increasing concentration of cobalt and copper oxides on the optical properties of the prepared glass specimens, such as refractive index, permittivity, extinction coefficient, electric susceptibility, dielectric constants, and optical conductivity were examined and studied. The glass network was analyzed using optical spectroscopy over a range of 190-2500 nm to investigate the effect of blue-green band-pass glass filters, band-pass absorption glass filters, and UV cut-off. The observation illustrated that the density increased and molar volume decreased with increasing the concentration of doping materials of cobalt and copper oxides. The decreasing optical energy gap with increasing concentrations of Co_2O_3 and CuO from 2.96 to 2.50 eV illustrated that the glass samples can be considered semiconductors in nature. Moreover, UV cut-off regions increased by increasing the concentration of doping materials.

Keywords: Blue-green bandpass filter, Cobalt oxide, Borate glass, Copper oxide, optical conductivity, Semiconductor.

1. Introduction

Optical Glass filters have recently been the most important technical in optical applications. An optical bandpass filter is an optical device that allows a specific range of wavelengths or colors of light to pass while attenuating the other wavelengths outside this range [1–5]. These filters were used in various optical applications. Optical bandpass filters played an important role in controlling the spectral properties of light for a wide range of scientific, [2,6-8] industrial and technological applications [8,9]. Glass of borate contained a significant amount of boron oxide (B_2O_3) as one of its primary glass a components which gave borate glass with unique optical properties compared to other types of glass [3–8]. The optical properties of borate glass included: Transparency and tending to have lower ultraviolet (UV) absorption compared to some other types of glasses [3,6,9,11-13]. These make it suitable for the UV-transmitting applications [2,8] including UV optics, UV-curable materials, nonlinear Optical Properties, and high Refractive Index [14–16]. Different borate glass formulations can result in variations in transparency, refractive index, dispersion, and other optical characteristics [2,6,10,17]. These properties made borate glass a versatile material with applications in a wide range of optical devices and systems [4,18–20]. Copper oxide was introduced as doping in borate glass, it had several impacts on the properties of glass, and it interacted with light and influenced the glass's optical properties, such as absorption and emission spectra [11-26]. This property was utilized in applications like optical filters [27-30]. Sodium ions impact the glass structure, influencing its physical and chemical properties. In glass technology, cobalt ions act as nucleating agents that promote the crystallization process [14,31]. Their influence on the structural characteristics of glasses influenced sample composition, synthesis method, and parameters. Actually, in glass network materials cobalt ions occur in two stable valence states, divalent as Co^{2+} and trivalent as Co^{3+}[4,5,15,21]. Cobalt ions were known for their ability to create deep blue colors in glass [1,2,4,7]. Cobalt-doped glass can absorb ultraviolet (UV) light, making it convenient for applications of UV-blocking [32,33]. Zinc oxide can affect the refractive index of the glass, making it useful for optical applications [23]. ZnO has well-known antibacterial properties. Copper, cobalt, zinc, and sodium oxide doped borate glass indicated a type of glass that contained multiple dopants. The quite effective application of CuO and Co_2O_3 (0.05 – 0.2 mg) to the bioactive glass has been assured to be effective in the inhabitation of bacteria growth [34]. Incorporating these dopants can produce glass with a wide range of properties and potential applications. The prepared glass sample was used as a blue-green bandpass filter for UV-preventing applications such as protection of UV laser [29]. The work aims to prepare a new composition of Optical Glass filters based on add ion of multi-transition metal oxides of cobalt and copper with different molar ratios to zinc-sodium oxides of borate glass to enhance their physical and optical properties.

2. Experimental and methods

2.1. Materials:

Analytical grade raw materials to prepare the ZnO, Na_2CO_3, Co_2O_3, CuO and H_3BO_3 glass specimens in powder form. Pure oxides of zinc oxide as (ZnO) (sigma-Aldrich = 99.9%), Sodium oxide (Na_2O) as sodium carbonate powder (Na_2CO_3) (sigma-Aldrich = 99%), Cobalt oxide as (Co_2O_3) (sigma-Aldrich = 99.99%), Copper oxide as (CuO) (sigma-Aldrich = 99.88%) and B_2O_3 was introduced as boric acid (H_3BO_3) (fluka, purity 99.99%).

2.2. Preparation of glass:

In this research, using the glass quenching melting technique to prepare network of glass with formula 55 B_2O_3, 35 ZnO, (10-x) Na_2O, x Co_2O_3, , y CuO (x=0, 0.05, 0.1, 0.15, 0.2, 0.25; y=0.1, 0.15, 0.20, 0.25, 0.30, 0.35), Pure chemical compounds were incorporated and ground by utilization mortar for 30 minutes of each sample and then carried to porcelain crucibles. The first step is the utilization of a muffle furnace at a temperature of 250 ˚C to liberate gases such as NH_3, H_2O, NO_2, and CO_2. All samples were melted in disposable porcelain crucibles by using an electrical muffle furnace for 1 h at 1000 ˚C, the molten material output was shaken many times to ensure the homogeneity of samples and got ride gas bubbles [3, 14, 29, 31]. The molten components were on a preheated stainless plate which was subsequently annealed at 250 ˚C for 30 minutes with a pressing plate to get thin disks with small thickness in millimeter units then samples were left to be cooled at room to avoid internal stress of glass samples then production samples got ride to cool slowly to room temperature.

2.3. The characterization methods:

The crystallite phase of transparent samples was analyzed using X-ray diffraction (XRD) with a Brucker axis D8 diffract meter and crystallographic data software Topas 2. A CuKα (λ = 1.5406 Å) radiation was used at 40 kV and 30 mA at a rate of 2°/min. The diffraction data were collected for 2θ values ranging from 4° to 70°. Density measurements were taken at room temperature using the Archimedes technique with ethanol as the liquid of immersion (density ρ_o= 0.888g/cm³). The specimens' density (ρ) and molar volume (Vm) were determined using Eqs. (1, 2), respectively [1, 2, 7].

$$\rho = \frac{W1}{W1-W2}\,\rho \qquad (1)$$

Where w1 and w2 are the weights of samples in air and ethanol, respectively.

$$\mathrm{Vm} = {M\omega}/{\rho} \qquad (2)$$

Where Mw is a molar mass

OPD (oxygen packing density) represents the density of packed oxygen atoms in amorphous or disordered structures [1, 2, 7, 21, and 29].

$$\mathrm{OPD} = \frac{n_o * \rho}{M\omega} \qquad (3)$$

Where n_o is the number of oxygen atoms per formula unit.

(N) The ionic concentration of glass indicated to relative abundance of different types of ions within the glass composition and it affects the structure and properties of glass [49]. It is determined by the following equation.

$$N = \frac{NA * P * \rho}{Mw} \qquad (4)$$

NA: It is the Avogadro's number, **ρ**: density, p: mol fraction of glass composition.

(r) Inter ionic distance of the glass samples is given by the following equation. It is between adjacent ions in glass structure [49]. It can affected packing arrangement of ions and lead to arrangement of ions and changes the interionic distance

$$r = (1/N)^{1/3} \qquad (5)$$

P = n*x, x: mole fraction of the glass composition, (n) is the number of atoms of element ions in the given oxide.

2.4. Measurements:

In this work the optical absorption and transmission spectra of the studied glass samples were recorded at room temperature using UV/VIS absorption (JASCO V570) spectrophotometer in the range of wavelength (190–2500) nm. Optical and physical properties of specimens were measured and calculated. Thicknesses of the samples of matrix were measured by Vernier caliper.

3. Results and discussion

3.1. XRD

Fig.1. shows the XRD spectra of the prepared glass network. The XRD pattern of the studied specimens was composed of vacillations and no sharp peaks were necessitated indicating that these samples had an unorganized temple of all specimens. Moreover, there is no alteration in its amorphous nature before and after successive incorporation of metal oxides (Co_2O_3 and CuO). On the other hand, XRD patterns for doped glass with metal oxides don't show any characteristic peaks for metal oxide which can be attributed to the low concentrations in the prepared glasses.

3.2 Density, Molar volume, ionic concentration, inter ionic distance and Oxygen packing density:

The density and molar volume of glass species were numbered and the results were summarized in Table 1. The results showed that the density of the S1 sample of undoped cobalt oxide was the lowest density which is equal to 2.753 g cm^{-3} while the glass network of doped Co_2O_3 slightly increments from 2.753 to 2.902 g cm^{-3} with increasing cobalt oxide concentration, the ionic concentration and the inter ionic-distance were calculated and tabulated in Table 2. A direct relation between the increase in ion concentration with increasing Co_2O_3. An inverse relation was observed between (N) and (r). Fig.2. Due to variation of atomic weights of Na, Co and Cu. Otherwise, the molar volume illustrated to be decreased from 26.08 to 24.60 cm^{-3} mol^{-1} with increment concentricity of cobalt oxide as shown in Fig.2. The addition of Co_2O_3 and CuO can induce structural changes in the borate glass network. These changes may include modifications in bond lengths, angles, or coordination environments of the atoms. Such alterations can lead to a more compact structure, resulting in a decrease in molar volume. The increase in density and decrease in molar volume of borate glass when adding increasing concentrations of cobalt oxide and copper oxide are primarily due to the higher atomic mass and solid solution formation of these oxides, which result in a more densely packed glass structure [35,36].

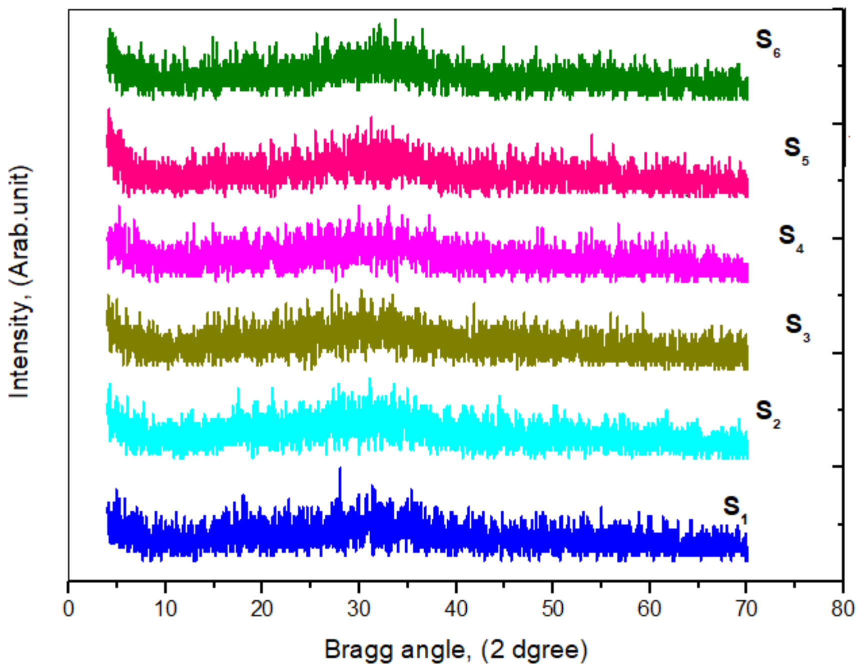

Fig. 1. XRD pattern of the prepared glass specimens

Additionally, any reduction in porosity and the volume occupied by the additives themselves can further contribute to these changes. The specific effects will depend on the composition and processing conditions of the glass. Table 3.1 exhibits that oxygen packing density (OPD) values of doped cobalt oxide samples S2-S6 are less than that of S1 specimen (89.38) and the difference is inversely proportional to the amount of doping cobalt oxide.

As a result, the structure of doped glass matrix is less tightly packed and the degree of disorder increases by the existence of Co_2O_3, i.e. formation of open structure which explains the observation of increasing density and decreasing molar volume.

Table. 1. Components, density (ρ), molar volume (Vm), and oxygen packing density (OPD) of glass network.

Samples	Glass components (mol %)					(Mw)*	ρ (g cm^{-3})	Vm ($cm^{-3}mol^{-1}$)	OPD**
	B_2O_3	ZnO	Na_2O	Co_2O_3	CuO				
S1	55	35	9.9	0	0.1	71.41	2.75	26.08	89.38
S2	55	35	9.8	0.05	0.15	71.48	2.79	25.75	88.43
S3	55	35	9.7	0.1	0.2	71.56	2.79	25.65	86.27
S4	55	35	9.6	0.15	0.25	71.64	2.81	25.49	85.72
S5	55	35	9.5	0.2	0.3	71.72	2.87	24.87	85.41
S6	55	35	9.4	0.25	0.35	71.79	2.90	24.60	84.33

*Mw: Molar Mass, **OPD: Oxygen Packing Denisity.

Table 2. of ionic concentration and inter- ionic distances of 55 B_2O_3, 35 ZnO, (10-x) Na_2O, x Co_2O_3, , y CuO (x=0, 0.05, 0.1, 0.15, 0.2, 0.25; y=0.1, 0.15, 0.20, 0.25, 0.30, 0.35).

No. of samples	Ionic concentrations (N) (ions/cm^3)	Inter- Ionic distance (cm)
S2	2.00776E+21	7.92677E-08
S3	3.91729E+21	6.34363E-08
S4	5.83845E+21	5.55351E-08
S5	7.75606E+21	5.05188E-08
S6	9.57276E+21	4.70964E-08

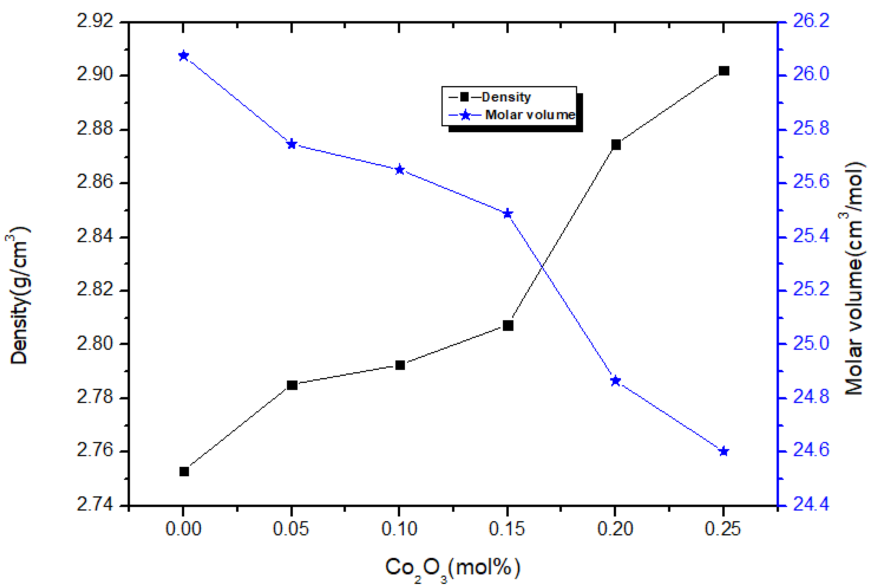

Fig.2. Influence of Co_2O_3 on density and molar volume of the equipped network of glass.

3.3 UV-VIS spectroscopy

This section studies the influence of Co_2O_3 and CuO concentrations on the optical parameters of the glass species and illustrates the absorption and transmission of the glass samples in the range of (190–2500 nm). Fig.3. clarified the optical absorption spectra of the glass network, showed fluctuations increased in these spectra due to variations of ions of cobalt ions and copper ions [7,8,14,20,37,39]. The interpretation of bands broadening due to multi absorption which showed as transitions [1,29]. As doping of cobalt, and copper oxides increase as absorption spectra increase. The first specimen without cobalt oxide (Co_2O_3) has a very light blue color but the doping network of cobalt oxide has an intensity of blue color increases as the concentration of Co_2O_3 and CuO increase.

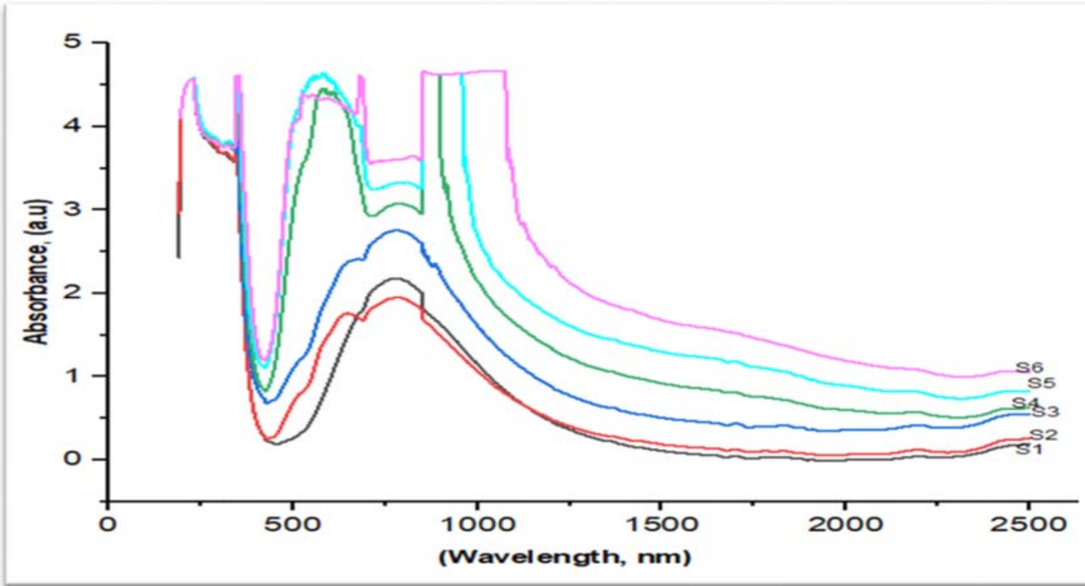

Fig.3. Optical absorption of considered glass matrix

Fig.4. Illustrated transmission spectra decrease from 64% to 5.9% due to doping material increment from cobalt oxide and copper oxide, the obtained glass matrix is used as an optical glass filter of UV for preventing UV and Nonlinear optical application [1,4,8,20,30,33]. The UV cutoff region of the glass matrix is illustrated as shown in Table 3. and Fig.5. showed an increase from 370 to 390 nm as increasing the concentration of cobalt oxide and copper oxide. This increase can be interpreted as the conversion of bridging oxygen atoms to non-bridging oxygen atoms. Likewise, the negative charges that existed on the non-bridging atoms of oxygen produced electron excitations with higher wavelengths [1].

Table 3. The cutoff region of UV-VIS of the glass matrix.

Co_2O_3 concentration	S1	S2	S3	S4	S5	S6
UV-Vis cut off λ (nm)	370	374	378	382	386	390

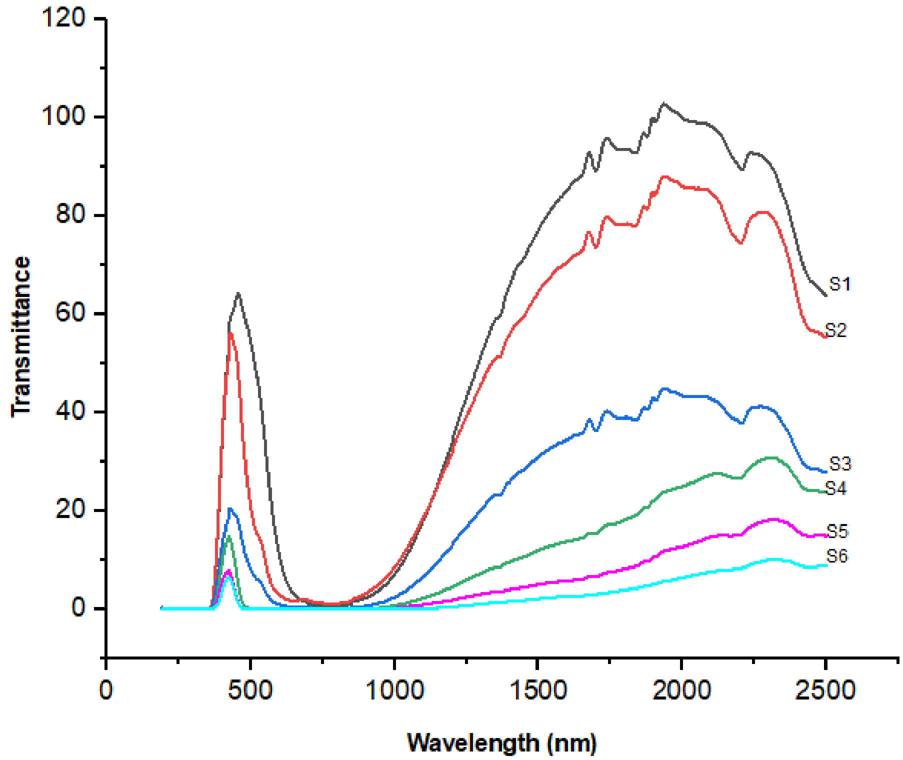

Fig.4. The optical transmission of the glass samples.

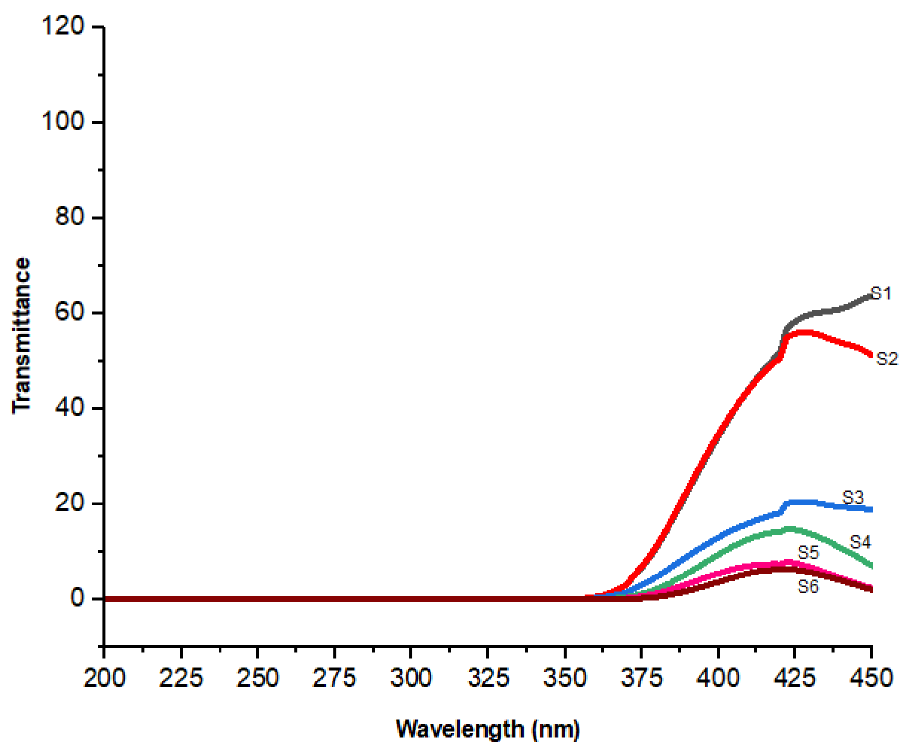

Fig.5. The cutoff region of UV-VIS versus wavelength glass samples.

The refractive index of glass depended on various factors such as the nature of components in the network, the parameters of glass oxides, two types of transmission bridging, non-bridging in the UV region, and the lattice vibration of glass specimens in the infrared region [1,2,38]. The refractive index increased due to two main reasons. Firstly, there was a difference in ion refraction between Co, Cu, and Na. The ion refraction of doping material of Co and Cu was higher than Na. Secondly, adding oxides of cobalt and copper to the glass matrix shrinked the interfacial distance of the glass network and reduced mean boron separation. This led to an increment of non-bridging oxygen, which increased the refractive index of glass samples [1,2,4,20]. The illustration in Fig.6 shows this effect.

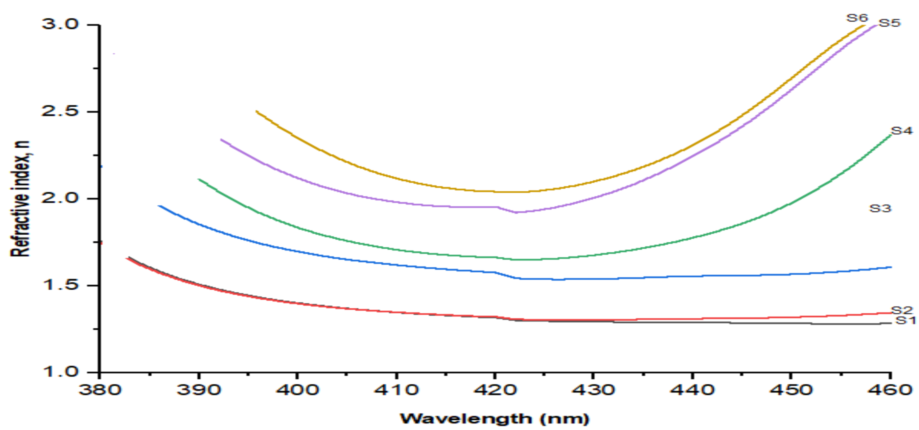

Fig. 6. The refractive index of the glass prepared at wavelengths between380 and 460 nm

The permittivity was a measure of how much electric field was reduced in a medium compared to a vacuum which depended on polarization, electronic transitions, and vibrations of molecules and atoms in material. According to some studies the permittivity had two peaks in UV and one peak in the infrared region. These peaks were caused by absorption resonances of borate glass and doping ions which are related to electronic transition and energy gap between levels that can be affected by the composition of the glass matrix. Both cobalt and copper ions introduced new absorption bands in visible and IR regions due to their d-d transitions. Therefore, as the concentration of cobalt and copper oxides increased, the permittivity curve tended to shift lower wavelength, had more peaks, and had higher value due to the existence of more BO_4 and non-bridging oxygen, more d-d transitions, and more polarization was induced in a network of glass [39] that illustrate in Fig.7.

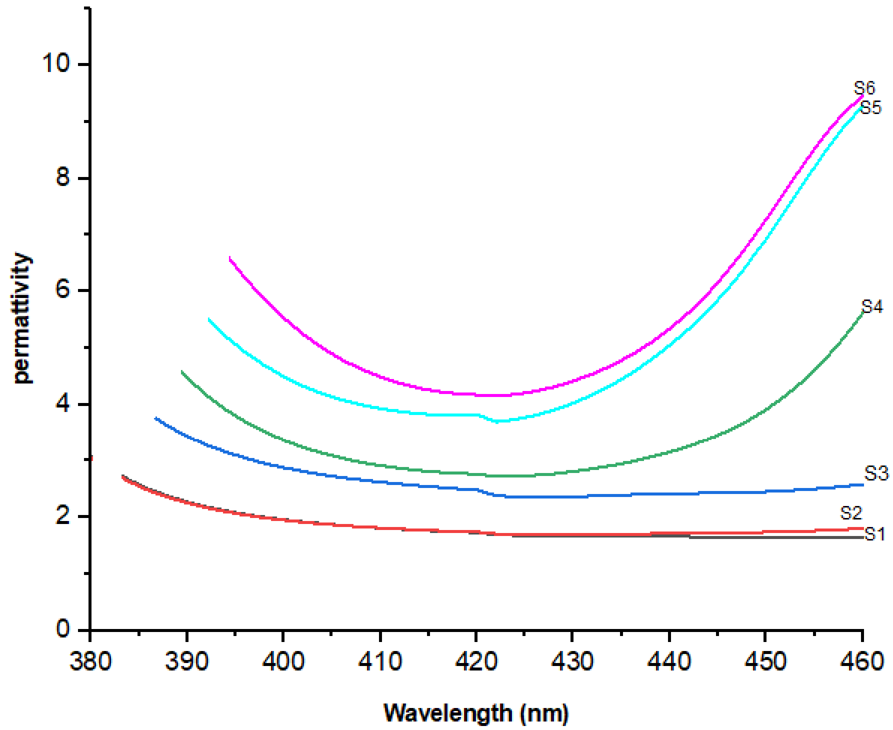

Fig.7. The permittivity of a glass matrix varied with different wavelengths.

Electric susceptibility was a measure of how easily it can be polarized by an electric field. It was influenced by various factors, including the nature of the polarizable entities, their concentration, and the wavelength of the incident electric field. It may decrease with increasing wavelength in certain regions of the electromagnetic spectrum due to the dopant ions becoming less effective at polarizing the material at longer wavelengths. As the dopant concentration increases, interactions between the dopant ions can occur, leading to complex effects on the electric susceptibility [40] as shown in Fig. 8. The dielectric and dielectric double constants distinguished the dielectric properties of the material. The dielectric constant measured the material stores the electric charge, while the dielectric double measured how much electric energy dissipated as heat. At low wavelength, the electric field was absorbed by charge transfer transitions which were very intense, broad, and overlapped with each other so they decreased firstly, then constants with increasing wavelength due to crystal field splitting of d- orbitals and finally, they were increased due to increase in vibrational energy [41] as shown in Figs.9 and 10. Fig.11. Showed that the curve decreased at low wavelengths, then became constant at intermediate wavelengths, and then incremented at high wavelengths. The curve also shifted to higher values with increasing concentrations of cobalt and copper oxides. Moreover, the electronic transition of metal ions in the network, and the Copper and cobalt ions existed in different oxidation states. At low wavelength, the light was absorbed by charge transfer that as wavelength increased the absorption by this transition decreased, at intermediate wavelength, the light was absorbed by d-d transition within the metal ions. These transitions were narrower and less intense than charge transfer transition, the glass network was heterogeneous and amorphous so d-d bands absorption was broadening and smoothing so the extinction was constant at this region. At higher wavelengths the glass matrix became more depolymerized and distorted, resulting in higher vibrational energies and higher extinction coefficients [42]. The extinction coefficient increase as a doping material increase due to the absorption of glass sample increase [36,38] as shown in Fig. 11.

In the field of materials science, optical conductivity was an important measure of a material's ability to conduct electric current when exposed to a specific frequency of electric field. This property was determined by the chemical composition and structure of the glass matrix. When the transitions that absorb light decreased, the optical conductivity was also reduced. At intermediate wavelengths, the light was absorbed by the metal ion's d-d transition, which depends on the crystal field splitting of the d-orbital. This, in turn, was influenced by the ligand field around the metal ions. As a result, the optical conductivity remains constant in this region. The transition related to the stretching and bending mode of borate glass depended on glass composition and modifier of content so as the concentration of copper and cobalt oxides increased as the distorted glass matrix increased, vibrational energy increased and optical conductivity increased with increasing wavelength [43-44], the optical conductivity of the material was closely related to the extinction coefficient indicating an increase in it [5] as illustrated in Fig.12.

The reflectance spectrum depended on chemical composition, it conducted in UV, visible or near in red. At UV wavelength. Reflectance in the visible region for wavelength between 400 and 700 nm. It might increase due to the nature of the glass system and the existence of certain elements or impurities [2] as shown in Fig. 13.

The refractive index at a certain wavelength can be calculated using reflectance (R) by the following relation [1,2,50-51].

$$R=\frac{n-1}{n+1} \qquad (6)$$

On the other hand, the reflectance (R) was enumerated by using the following equation, [1,2,25,45,46]

$$A + R + T = 1;\ \text{So}\quad R = 1 - A - T \qquad (7)$$

Where, A: absorbance R: reflectance T: transmittance

The permittivity (ε), the electric susceptibility (χ), and the polarizability (γ) of the glass samples were calculated using the following equations [44- 46, 50-51]

$$\varepsilon = n^2 \qquad (8)$$

$$\grave{\varepsilon} = n^2 - k^2 \qquad (9)$$

$$\ddot{\varepsilon} = 2nk \qquad (10)$$

$$\chi = \frac{\varepsilon-1}{4\pi} \qquad (11)$$

The extinction coefficient (k) and optical conductivity were calculated by using the following relation [29,30, 45- 46]

$$k = \frac{\alpha\lambda}{4\pi} \quad (12)$$

Optical conductivity = $\alpha n c / 4\pi$ (13)

Where α is the absorption coefficient, λ is the wavelength.

$(\alpha \upsilon) = (1/d) \ln (I_o/I) = 2.303\ (A/d)$ (14)

Fig.13 shows that as doping chemical composition of copper oxide and cobalt oxide increases as reflectance decreases. Electric constant, Dielectric constant, electric susceptibility and permittivity of glass composition described its ability to store the electric energy and doping material influenced on electronic properties so increment with introducing cobalt and copper oxides[1,29,30,38,46] as shown in figs.8, 9, 10. Finally, the optical band gap, e.g., of the borate glass was calculated using the relation [1,47].

$ah\upsilon = B\ (h\upsilon - Eg)^{2}$ (15)

Where, B is constant and α is the absorption coefficient that was calculated by using the relation [1,29,47].

Where I_0 and I are intensities of the incident and transmittance beam respectively, A was absorbance and d was the thickness of glass samples.

The optical band gap energy, often simply referred to as the "band gap," is a fundamental property of materials, especially semiconductors and insulators.

The band gap energy of borate glasses generally falls within a range of approximately 2.5 to 4.0 electron volts (eV) [11,47], depending on factors such as the concentration of boron oxide and the presence of other additives or dopants**.** For amorphous material, the plot of $(h\upsilon\alpha)^{1/2}$ as a function of photon energy (hυ) is used to determine the optical band gap [1,5,7,36] as shown in fig.14. Increment Co_2O_3 and CuO concentricity in glass decrement in the optical band gap from 2.95 to 2.58 eV that illustrated that the glass samples considered semiconductor in nature [1-2] with as shown in figure .14, which was attributed to increasing in number of non-bridging oxygen (NBO) atoms**.** The studied glasses possessed many properties that qualify them for optoelectronics and nonlinear optics applications [1,10,21,40,48]. The results showed that the glass matrix was used as an optical blue-green band pass filter which is located at 350 – 590 nm [1,2,4,7, 45,46].

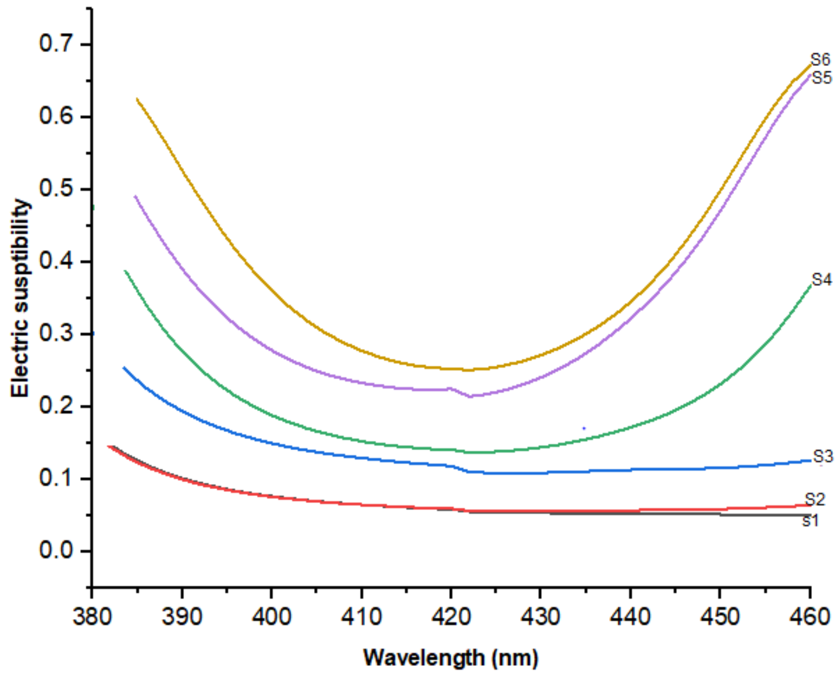

Fig.8. The electric susceptibility versus wavelength.

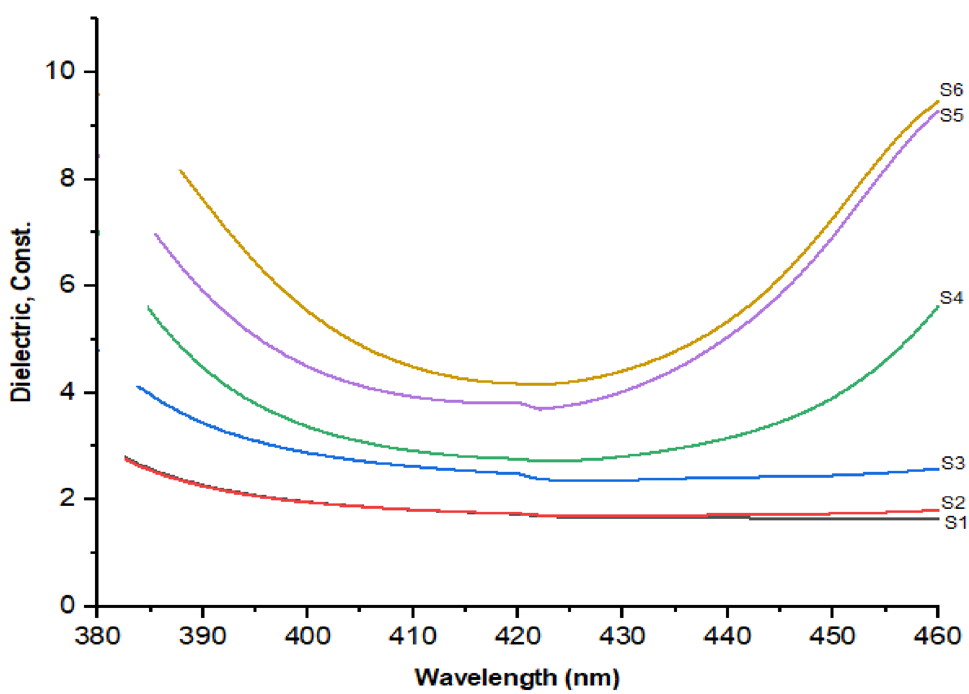

Fig.9. The dielectric constant versus wavelength

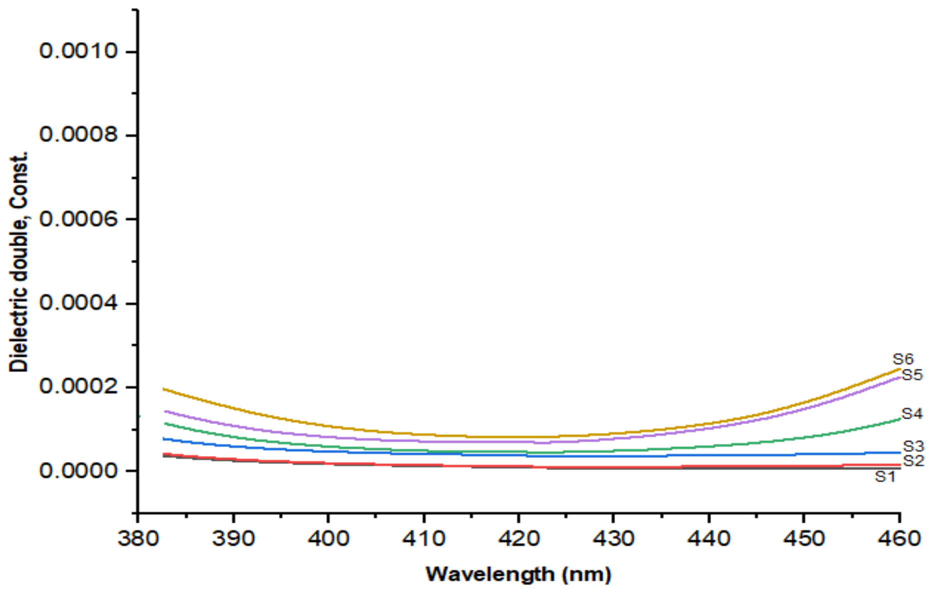

Fig.10. The dielectric constant versus wavelength for glass samples.

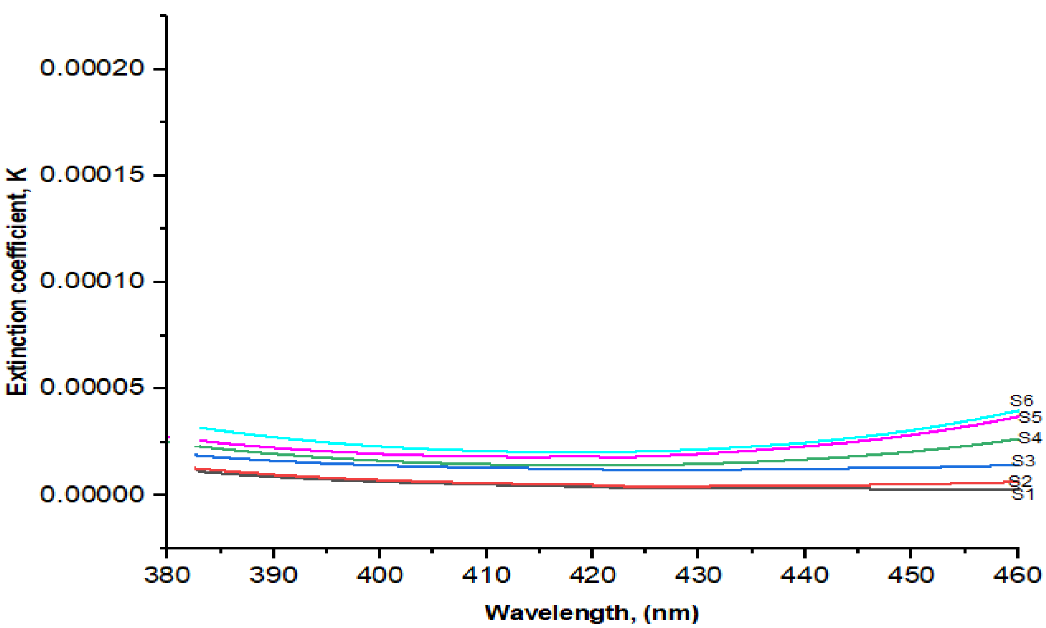

Fig.11. the extinction coefficient, k versus wavelength for glass specimens

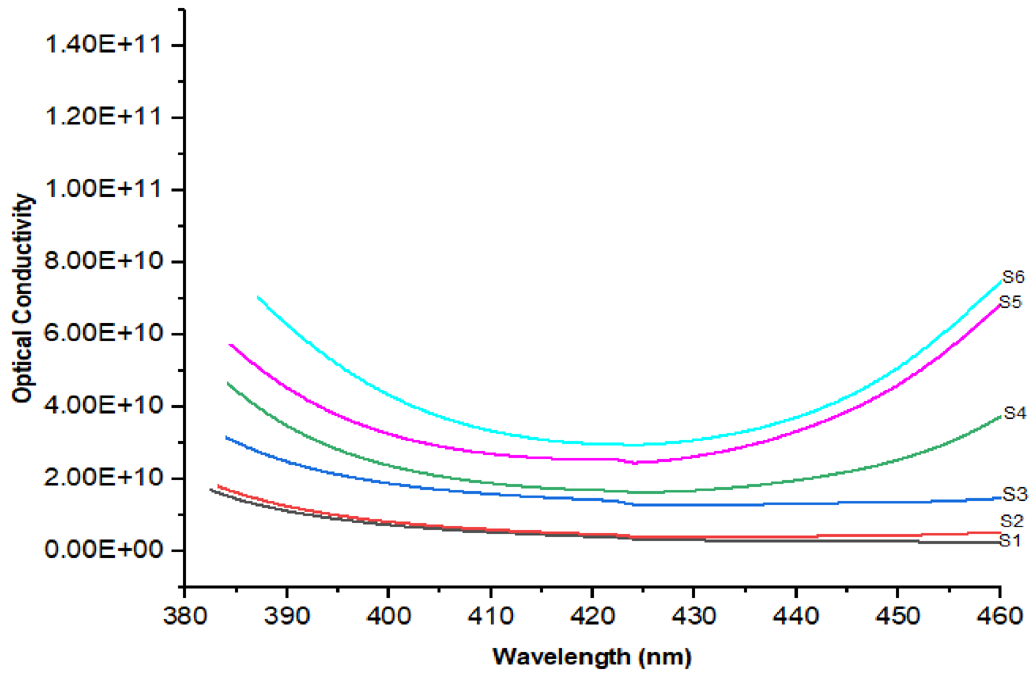

Fig.12. the optical conductivity versus wavelength for glass matrix

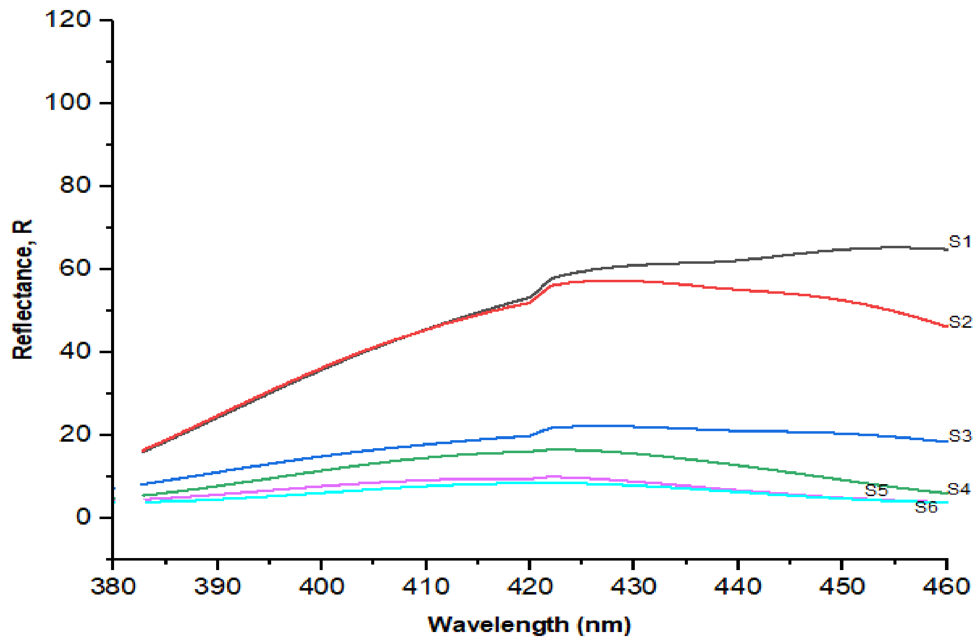

Fig.13. The reflectance versus wavelength for glass network.

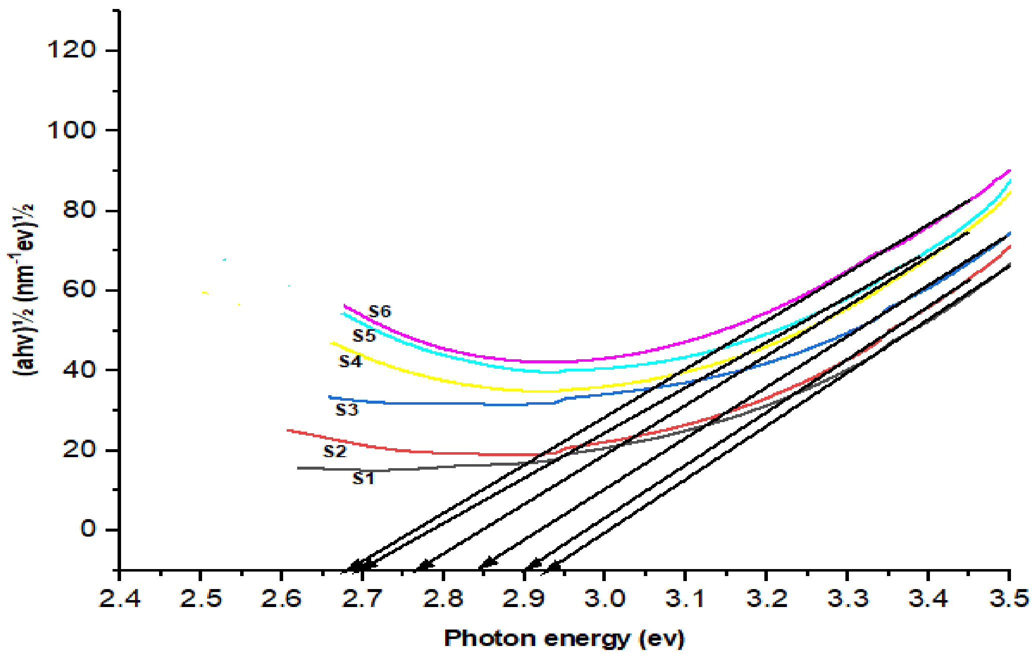

Fig.14.The optical energy gap for present glass samples

Conclusions

This paper showed the influence incorporation of Co_2O_3 and CuO on the zinc and sodium oxide glass network. All prepared glass systems are made amorphous by using XRD. The glass network illustrated that the density and ionic concentration increased, while molar volume and inter-ionic distance decreased by increment concentration of doping material of cobalt and copper oxides. The studied glass samples showed increasing the cutoff region in UV-VIS with increasing doping material, and the optical band gap energy decreased from 2.95 to 2.58 eV so this illustrated that the glass matrix is considered semiconductor material in nature. Experimental results clarified that the refractive index, extinction coefficient, permittivity, optical conductivity, electric constant, Dielectric constant, and electric susceptibility increased as increasing doped material of CuO and Co_2O_3. The experimental data showed that the obtained glass network was used as an optical blue-green band pass filter which is located at 350 – 590 nm.

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