Comparison of Physical and Optical Properties of Glass Doped with Cobalt Oxide from Chemical and Sugarcane Leaf Ash

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Abstract

This research investigates and compares the physical and optical properties of cobalt oxide-doped glass prepared using conventional chemical methods and glass doped with cobalt oxide using sugarcane leaf ash as a source of SiO_2 and CaO. The composition of sugarcane leaf ash was analyzed at various sintering temperatures using X-ray fluorescence (XRF) spectroscopy, revealing a high SiO_2 content, with the maximum value reaching 69 wt%. The glass composition was formulated based on the ratio (50–x)SiO_2 (with sugarcane leaf ash used as a partial substitute for SiO_2 and CaO): $25B_2O_3$: $10Na_2O$: 8CaO: 7SrO: xCoO, where x represents the cobalt oxide concentration (0.00, 0.01, 0.02, 0.03, 0.04, and 0.05 mol%). The results showed that both the density and refractive index increased with higher CoO concentrations, while the molar volume decreased. The optical absorption spectra in the wavelength range of 350 – 2000 nm exhibited an increasing trend with rising CoO content. The cobalt oxide-doped glass displayed a blue color, whereas the glass doped with cobalt from sugarcane leaf ash exhibited a reddish-blue hue, as confirmed by CIE Lab* color measurements.

Keywords: Glass; CoO; Sugarcane leaf ash; Physical properties; Optical properties

1. Introduction

Sugarcane is an important economic crop in Thailand. Sugarcane is grown on an area of 19.58×10^9 m² (1 rai = 1600 m²), covering the northern, eastern, northeastern, and central regions of the country. Each year, sugarcane farmers harvest a large amount of waste material, which is sugarcane leaves. Normally, sugarcane leaves are produced at about 0.9375 kg m⁻² (1 rai = 1600 m²), leaving about 18 million tons of sugarcane leaves per year, which is a large amount. However, a large amount of sugarcane leaves is not used. Although sugarcane leaves are currently processed into processed products, household items, and decorative items, there are still a large amount of sugarcane leaves left [1]. Some farmers leave the sugarcane leaves in the fields without doing anything, while some farmers burn the sugarcane leaves to prepare for another round of planting. When the burned sugarcane leaves were examined for chemical composition, it was found that the most common chemical component in burnt sugarcane leaves was silica (SiO_2), which is the main component used in the glass industry [2, 3].

Glass has an amorphous structure, is a solid material that is not crystalline (no fixed shape), has a short-range atomic arrangement, is transparent and fragile because the atoms inside the glass are arranged in a specific disorder. Most glass melting uses SiO_2 because SiO_2 has the properties of glass to withstand high heat and is inert to chemicals. In addition, previous research has used agricultural waste ash, which is mainly composed of silica, to melt glass [4, 5]. Therefore, researchers are interested in using sugarcane leaf ash, which has the highest SiO_2 content, to replace SiO_2 in glass melting and doped with cobalt oxide.

Cobalt is one of the transition [6] elements that has a high concentration of blue ions, and only small amounts of cobalt can produce a clear color. Cobalt can be used as a color indicator, with the color changing in proportion to the amount of tetrahedral and octahedral coordination in the glass [7].

Due to the significance mentioned above, the researchers were interested in utilizing sugarcane leaf ash from Nakhon Pathom provinces as a component in glass production. Therefore, this study investigated the chemical composition of sugarcane leaf ash under different temperature conditions using X-ray Fluorescence Spectrometry (XRF) for its suitability in glass manufacturing. Subsequently, the sugarcane leaf ash was incorporated into glass formulations to examine its physical and optical properties, including density, molar volume, refractive index, optical absorption, and color. Cobalt oxide (CoO) was added at varying concentrations (x = 0.00, 0.01, 0.02, 0.03, 0.04, and 0.05 mol%). The objective of this study is to provide a foundational approach for the development of glass-based products for various applications.

2.   Materials and Methods

In this research simulated the burning of sugarcane leaf at different temperatures using. Analysis of the chemical composition of sugarcane leaf ash using X-ray Fluorescence (XRF) (Minipal-4, Panalytical) revealed that the main chemical component of the leaves is silica (SiO_2), constituting 69.70 wt% by weight use temperature at 600°C. Additionally, contaminants that affect color formation in glass, such as MnO, Fe_2O_3, and CuO, were also detected but it does not significantly affect the color of the glass [8]. It was noted that increasing the sintering temperature does not affect the amount of silica (SiO_2) as shown in Table 1.

Table 1 The composition of sugarcane leaf ash was analyzed at different sintering temperatures.

Chemical composition (wt%)	Temperature (°C)				
	Raw	400	600	800	1,000
SiO_2	68.87	69.15	69.70	68.65	69.00
P_2O_5	2.15	2.08	1.91	2.20	2.69
K_2O	4.65	4.63	4.40	3.20	3.72
MgO	2.00	2.60	2.40	2.70	2.50
CaO	18.98	17.95	17.88	18.78	20.06
MnO	0.08	0.08	0.08	0.09	0.10
Fe_2O_3	0.54	0.48	0.44	0.70	0.73
SO_3	2.64	2.95	3.13	3.54	1.08
CuO	0.02	0.02	0.02	0.03	0.04
Total	100	100	100	100	100

After analyzing the chemical composition, the results presented in Table 1 showed that SiO_2 had the highest concentration at all temperatures tested, with the maximum value observed at 600°C. Therefore, sugarcane leaf ash obtained at this temperature was selected for glass fabrication. As shown in the table, SiO_2 and CaO were present in relatively high amounts. Consequently, the glass composition was formulated to reflect a ratio of SiO_2 and CaO consistent with the X-ray fluorescence (XRF) analysis. The glass was synthesized using the following molar formula: $(50–x)SiO_2$ (where SiO_2 and CaO were partially substituted with sugarcane leaf ash): $25B_2O_3$: $10Na_2O$: 8CaO: 7SrO: xCoO, where x represents the molar concentration of CoO (0.00, 0.01, 0.02, 0.03, 0.04, and 0.05 mol%). Then, it was melted in an electric furnace at a temperature of 1200°C for 3 h. The molten glass was poured into a graphite mold and baked at a temperature of 500°C for 3 h, followed by cooling at room temperature. After the glass is annealed at 500°C, the cooling process is important. The cooling process should be gradual, cooling at about 33°C per 1 h, to reduce the heat stress and improve the quality and strength of the glass. The cooled glass samples were cut into the size of $1.00 \times 1.50 \times 0.30$ cm³ [9], as shown in Fig. 1. These samples were then analyzed for their physical and optical properties, including density (Densitometer, A&D HR-200), molar volume, refractive index (Abbe refractometer, Atago), X-ray diffraction (XRD) (Shimazdu XRD-6100), absorption spectra (UV-VIS-NIR Spectrophotometer, Shimazdu UV-3600), and color coordinates (UV-VIS Spectrophotometer, Cary-50).
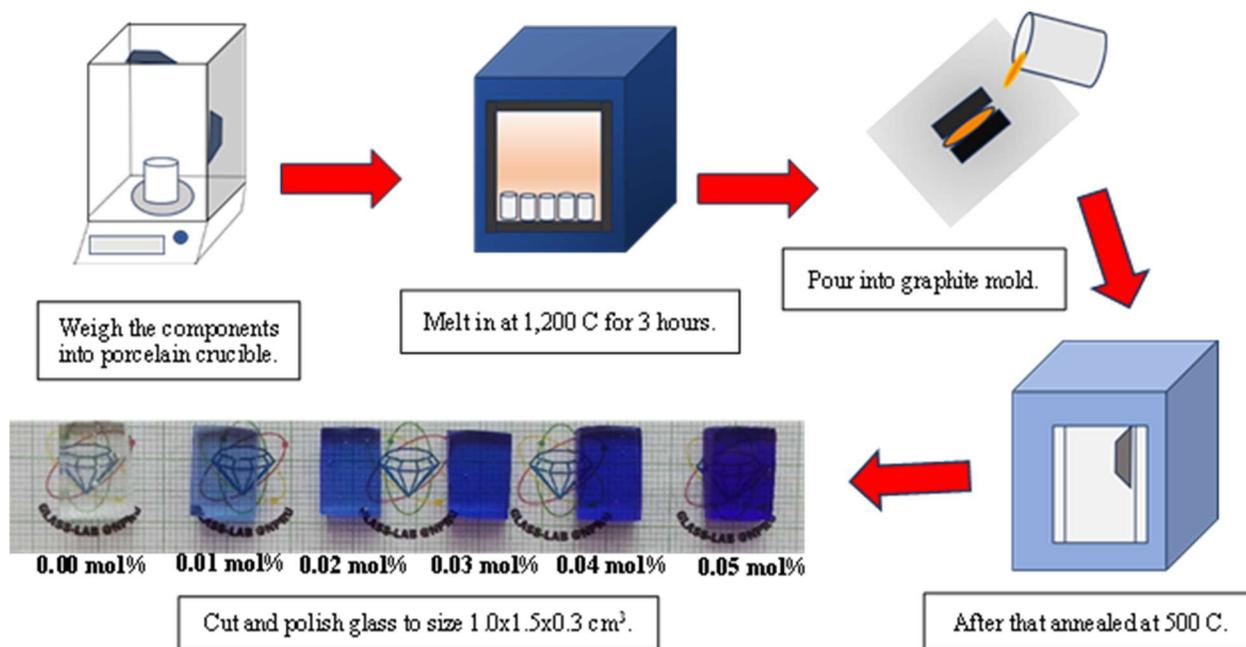
Fig. 1 Preparation of glass with sugarcane leaf ash substituting for SiO_2 and CaO doped CoO.

3. Results and Discussions

The glass samples were prepared using the molar formula $(50 - x)SiO_2$ (with sugarcane leaf ash used as a partial substitute for SiO_2 and CaO): $25B_2O_3$: $10Na_2O$: 8CaO: 7SrO: xCoO, where x represents the CoO concentration (0.00, 0.01, 0.02, 0.03, 0.04, and 0.05 mol%). The glass synthesized without CoO is clear and colorless. Upon the addition of CoO, the glass turns blue, and the color intensity increases with the CoO concentration. This is due to Co^{2+} ions absorbing light within the visible wavelength range, as illustrated in Fig. 2 (a). For the glass produced using sugarcane leaf ash, the sample without CoO appears clear with a slight yellow tint, attributed to the presence of iron ions in the ash. After CoO is added, the glass exhibits a blue-red hue [7], and this coloration becomes more intense with increasing CoO content, as shown in Fig. 2 (b).
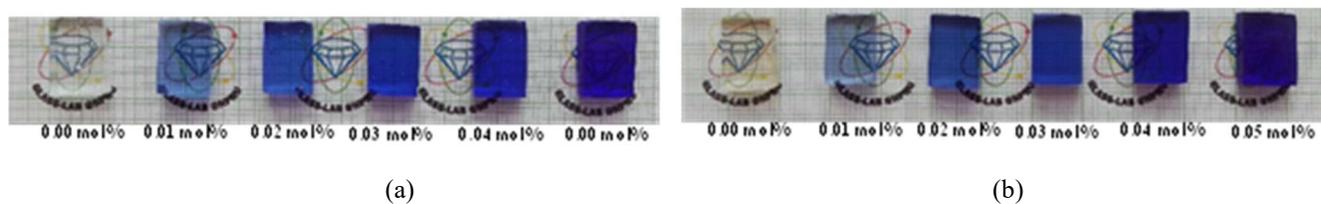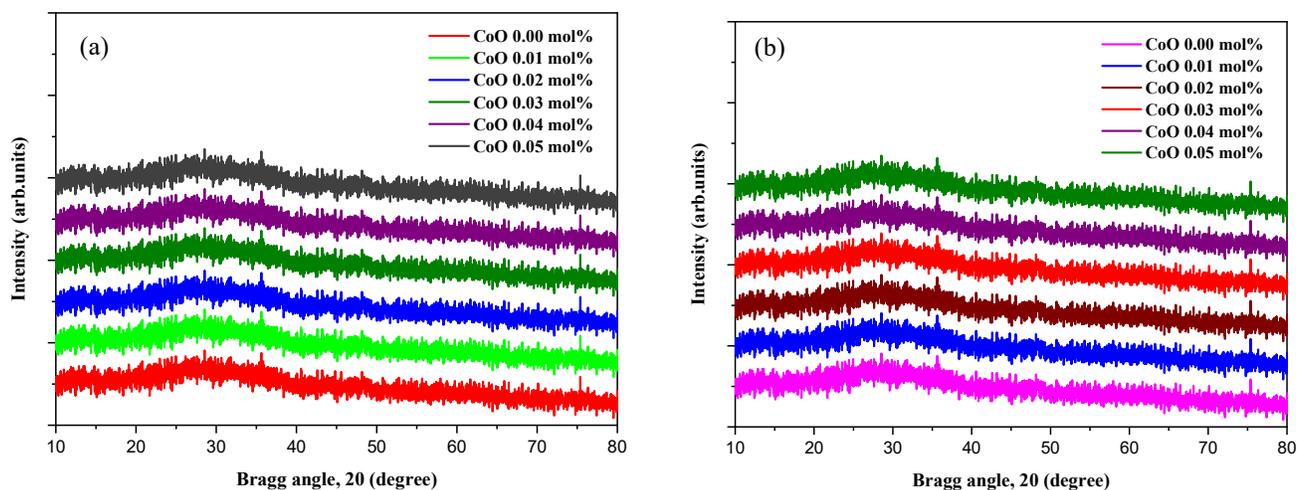

(a)   (b)

Fig. 2 (a) Glass doped with CoO at various concentrations using SiO_2 chemical (GCC) and (b) Glass doped with CoO at various concentrations using sugarcane leaf ash (GSC).

The XRD pattern analysis of the glass samples revealed no obvious crystalline phases, as the as-prepared glasses were completely amorphous. The results showed that the synthesis process successfully produced a homogeneous glass structure without any crystals, confirming the purity of the amorphous phase in the glass samples, indicating that the preparation method could effectively eliminate the formation of crystalline structures within the glass matrix. The study consistently reported similar broad diffraction patterns for amorphous glasses as shown in Fig. 3 [10,11].

Fig. 3 (a) X-ray diffraction (XRD) of glass using SiO$_2$ chemical (GCC) and (b) X-ray diffraction (XRD) of glass using sugarcane leaf ash (GSC) with various concentrations of CoO.
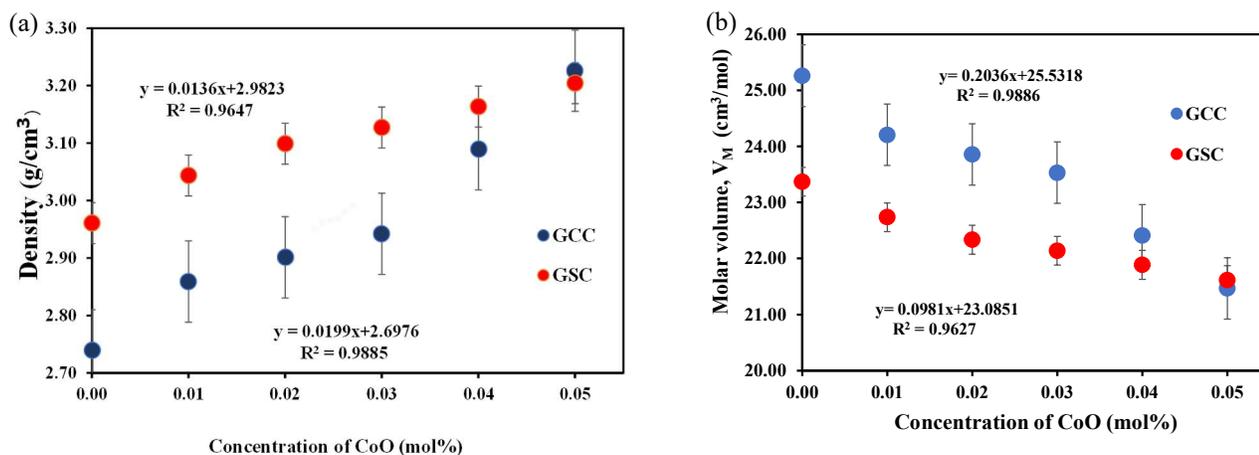

Fig. 4 (a) Density of glass doped CoO. and (b) molar volume of glass doped with various concentrations of CoO.

Density measurements of CoO-doped glasses synthesized from both chemical precursors and sugarcane leaf ash at concentrations of 0.00, 0.01, 0.02, 0.03, 0.04, and 0.05 mol% were conducted using Archimedes' principle. Both glass systems demonstrate a concurrent increase in average molecular weight with increasing CoO content, indicating stoichiometrically consistent incorporation of the dopant into the glass network. For glasses fabricated from chemical precursors, the measured density values ranged from (2.7394 ± 0.0004) to (3.2259 ± 0.0001) g·cm^{-3}, while those produced using sugarcane leaf ash exhibited densities ranging from (2.9607 ± 0.0003) to (3.2041 ± 0.0003) g cm^{-3}. This increase in density is attributed primarily to the higher molecular weight of CoO relative to SiO$_2$; as CoO substitutes for SiO$_2$ in the glass matrix, the overall molecular mass increases, leading to enhanced structural compactness. The introduction of heavier cobalt ions into the network contributes to denser atomic packing, as these ions occupy positions previously held by lighter silicon atoms, thereby improving the packing efficiency of the glass structure. Notably, a consistent disparity in density is observed between the two glass types: ash-derived glasses display systematically higher densities than their chemically synthesized counterparts. This difference is ascribed to the presence of additional heavy metal oxides such as CaO, MgO, and Fe$_2$O$_3$ in the sugarcane ash, which serve as effective network modifiers. These oxides introduce supplementary ionic crosslinks and enhance network rigidity, resulting in a more tightly packed and structurally compact glass matrix compared to glasses prepared from compositionally simpler chemical precursors [12, 13]. The relationship between density and CoO concentration for both glass systems reveal a strong positive linear trend. For glasses synthesized from pure chemicals, the linear regression yields a correlation coefficient (R²) of 0.9885. Similarly, glasses derived from sugarcane ash yield an R² of 0.9647, as depicted in Fig. 4 (a). These high coefficients

of determination confirm a robust linear dependency of density on CoO content, emphasizing the reliability and reproducibility of CoO doping in modulating the structural compactness of the glass matrix.

The molar volume, calculated based on the ratio of molecular weight to density, indicates a decreasing trend with increasing CoO concentration. This calculation is critical understanding how the incorporation of different components affects the glass structure. Glass using SiO_2 chemical the molar volume values ranged from $25.2577 - 21.4658$ cm³ mol⁻¹ and glass using sugarcane leaf ash the molar volume values ranged from $23.3696 - 21.6129$ cm³ mol⁻¹. The decrease in molar volume indicates that as the concentration of CoO increases, the glass network becomes more compact, possibly due to the interaction between CoO and the oxygen atoms bridging the glass matrix. Since CoO ions replace some silicon ions in the silicate network, this substitution changes the network connectivity, leading to stronger bonding interactions with the bridging oxygens. This results in a reduction in the free space within the glass structure, leading to a more stable glass network. As a result, the overall density of the glass increases while the molar volume decreases, indicating a complex balance between the composition and the physical properties. The glass using SiO_2 chemical resulting graph shows a strong linear relationship with a correlation coefficient (R^2) of 0.9886. This trend is visualized, and glass using sugarcane leaf ash resulting graph shows a strong linear relationship with a correlation coefficient (R^2) of 0.9627. This trend is visualized in Fig. 4 **(b)** [14].

The refractive index (n_d) of glass doped with cobalt oxide from chemical and sugarcane leaf ash, conducted using an Abbe refractometer, found that the refractive index tends to increase with higher CoO concentrations. The refractive index of glass using SiO_2 chemical values ranged from (1.5605 ± 0.0003) to (1.5922 ± 0.0002). The refractive index of glass using sugarcane leaf ash values ranged from (1.6488 ± 0.0002) to (1.6686 ± 0.0001), indicating a consistent increase as the CoO content was elevated. This observed trend mirrors that of the density, reinforcing the understanding that the refractive index is intrinsically governed by both the material's density and the electronic polarizability of its constituent ions. As articulated by the Lorentz-Lorenz relation, the refractive index increases with enhancements in these parameters. Notably, ash-derived glass samples consistently exhibit higher values compared to their chemically synthesized counterparts. This can be attributed to the incorporation of CoO, which introduces heavier cobalt ions into the glass network, thereby augmenting both the polarizability and the density of the system, and consequently leading to an elevated refractive index. In contrast, chemically synthesized glasses, lacking such ions, exhibit a comparatively lower optical density. While CoO incorporation, typically forming octahedral complexes, increases polarizability in both systems, its impact is more significant in glasses using sugarcane leaf ash due to denser ionic arrangements [15]. The relationship between the refractive index and CoO concentration, the resulting graph demonstrates a strong linear correlation. The refractive index of glass using SiO_2 chemical with a coefficient (R^2) of 0.9902, the refractive index of glass using sugarcane leaf ash with a coefficient (R^2) of 0.9789, as shown in Fig. 5.
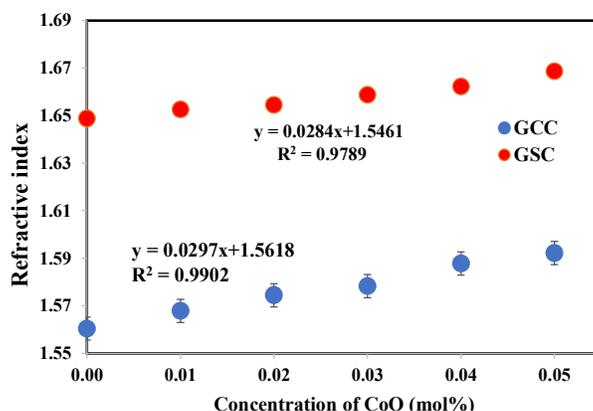
Fig. 5 The refractive index of glass doped with various concentrations of CoO.

The dielectric constant (ε) and the optical dielectric constant both increase with CoO doping and are generally higher in glasses using sugarcane leaf ash. As shown in Fig. 6 (a), the dielectric constant (ε) increases from approximately $2.7185 - 2.7842$ in glasses using sugarcane leaf ash, compared to an increase from approximately $2.4352 - 2.5351$ in glass using SiO_2 chemical. This enhancement is attributed to the reduction of non-bridging oxygen in the glass structure, which lowers polarization losses. Consequently, glasses using sugarcane leaf ash exhibit higher dielectric constants, resulting in improved electrical insulation. Similarly, as shown in Fig. 6 (b), the optical dielectric constant increases from approximately

$1.7185 - 1.7842$ in glasses using sugarcane leaf ash, compared to an increase from approximately $1.4352 - 1.5351$ in glass using SiO_2 chemical. This trend aligns with that observed in the dielectric constant. These values reflect the ease with which the electron cloud of the material can be distorted by an external electric field. The higher dielectric response observed in glasses using sugarcane leaf ash a more polarizable structure, likely due to a greater presence of non-bridging oxygen atoms and complex bonding arising from diverse network modifiers. This observation is consistent with recent findings that report enhanced dielectric properties in structurally heterogeneous glasses [16].
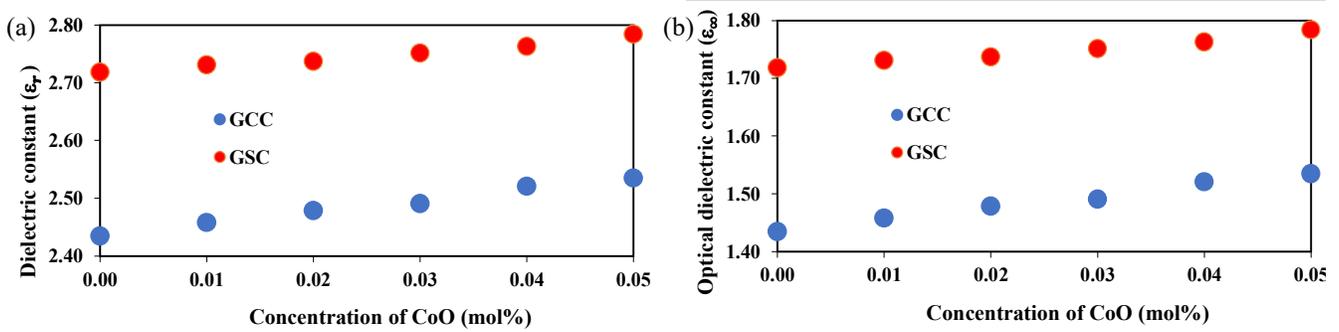

Fig. 6 (a) Dielectric constant of glass doped CoO and (b) Optical dielectric constant of glass doped CoO.

The molar refractivity (R_m), a measure of the total polarizability of one mole of a substance, depends on both the molecular polarizability and the density of the material. It serves as an important distinguishing factor. At higher densities, glass using sugarcane leaf ash exhibit a higher molar refractivity than glass using SiO_2 chemical. This is because CoO molecules in glasses using sugarcane leaf ash bind within the glass network, reducing the amount of non-bridging oxygen. As a result, the refractive index of glass using sugarcane leaf ash is higher than that of glass using SiO_2 chemical. The molar refractivity of glass using sugarcane leaf ash ranges from approximately $8.5114 - 8.0603$ cm³ mol⁻¹, whereas that of glass using SiO_2 chemical ranges from $8.1731 - 7.2660$ cm³ mol⁻¹, as shown in Fig. 7. This suggests that glass using sugarcane leaf ash may have higher densities, which reduce the flexibility of electron groups possibly due to a more extensive ionic network and lower oxygen mobility. In contrast, the less rigid structure of chemically synthesized glasses may allow for greater local polarizability.
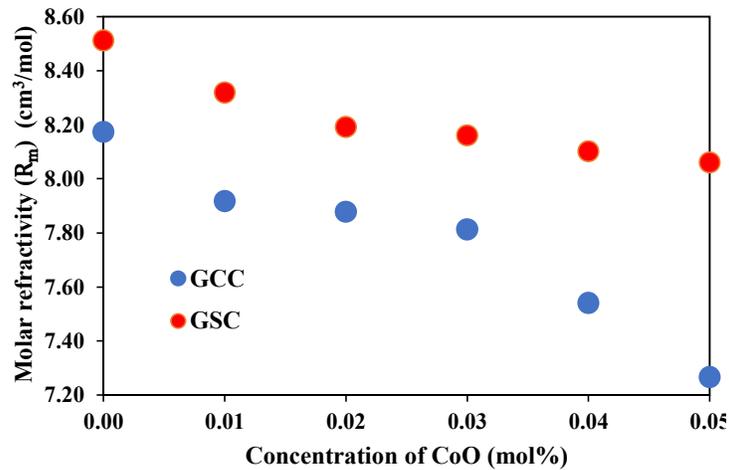

**Fig. 7** The molar refractivity of glass doped with various concentrations of CoO.

The reflection loss (R), which is associated with surface light reflection, increases with higher CoO content and is more pronounced in glass using sugarcane leaf ash. The reflection loss ranges from approximately $5.9996 - 6.2772$ cm³ mol⁻¹ for glass using sugarcane leaf ash, and from approximately $4.7918 - 5.2192$ cm³ mol⁻¹ for glass using SiO_2 chemical, as shown in Fig. 8. This increase is attributed to the incorporation of CoO, which raises the glass density, thereby reducing light transmission and resulting in greater reflection. This trend aligns with the higher refractive index, as predicted by Fresnel's

equation. Although higher reflection losses may reduce optical transparency, the increased glass density may be advantageous for applications such as nonlinear optics or radiation shielding.
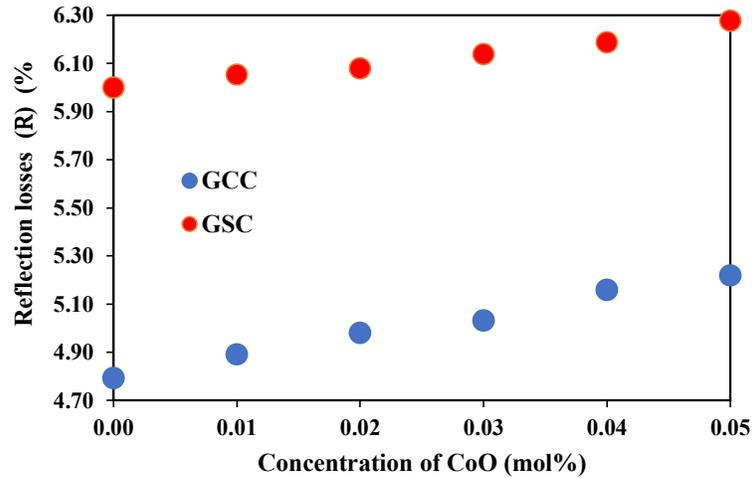
Fig. 8 The reflection loss of glass doped with various concentrations of CoO.

Molecular electronic polarizability (α_m), a molecule's ability to be distorted by an external electric field, resulting in an induced dipole moment essential parameter influencing optical behavior at the molecular level, decreases significantly with increasing CoO concentration in both systems. However, this reduction is more pronounced in glass using sugarcane leaf ash, where α_m drops from approximately 3.3729 to 3.1914 × 10^{-22} m³, compared to a decrease from about 3.2388 to 2.8793 × 10^{-22} m³ in glass using SiO$_2$ chemical, as shown in Fig. 9. This indicates that glass using sugarcane leaf ash exhibit a more constrained electronic structure, likely due to stronger internal electric fields and reduced bond flexibility caused by multivalent cations. This observation aligns with the trend observed in molar refractivity [17].
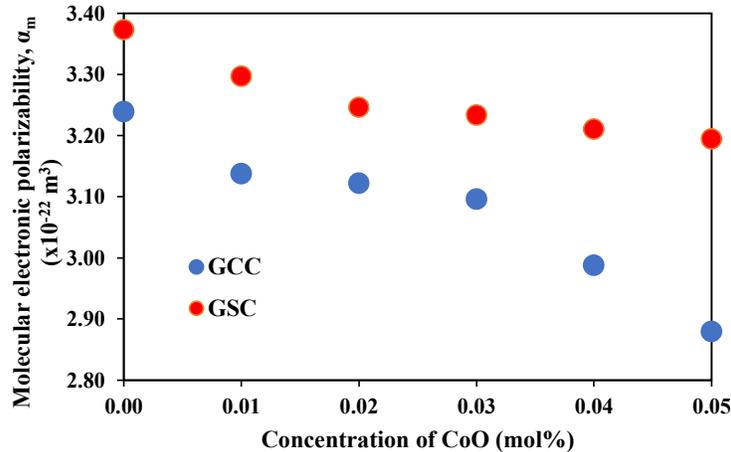
Fig. 9 The reflection loss of glass doped with various concentrations of CoO.

Further insights into the microstructure can be obtained by analyzing the Co-ion concentration (N), polaron radius (r_p), and interionic distance (r_i). Although both glass systems exhibit an increase in ion concentration with higher CoO content, glass using sugarcane leaf ash accommodate slightly more Co ions, possibly due to the presence of a greater number of non-bridging oxygen vacancies. The polaron radius decreases from approximately 29.1306 − 16.7504 Å in glasses using sugarcane leaf ash, compared to a decrease from approximately 29.7452 − 16.7126 Å in glass using SiO$_2$ chemical. As the cobalt ion concentration increases, the number of electrons in the atomic structure also rises, leading to stronger interactions between the central and outer electrons, which reduces the polaron radius. Similarly, the interionic distance decreases from approximately 72.2750 − 41.5589 Å in glass using sugarcane leaf ash, and from approximately 73.7999 − 41.4651 Å in glass using SiO$_2$ chemical. This trend reflects the reduction in distance between cobalt ions as their concentration increases. The smaller polaron

radii and shorter ion spacing indicate a higher density of local charge carriers and a more compact ion arrangement. As a result, the internal field intensity (F) slightly increases, ranging from approximately $0.0471 - 0.1426$ in glass using sugarcane leaf ash, compared to an increase from approximately $0.0452 - 0.1432$ in glass using SiO_2 chemical. These changes may significantly affect charge transport, the electronic band structure, and the overall electrical properties critical factors in developing glass materials for optoelectronic and photonic applications [18], as shown in Fig. 11.
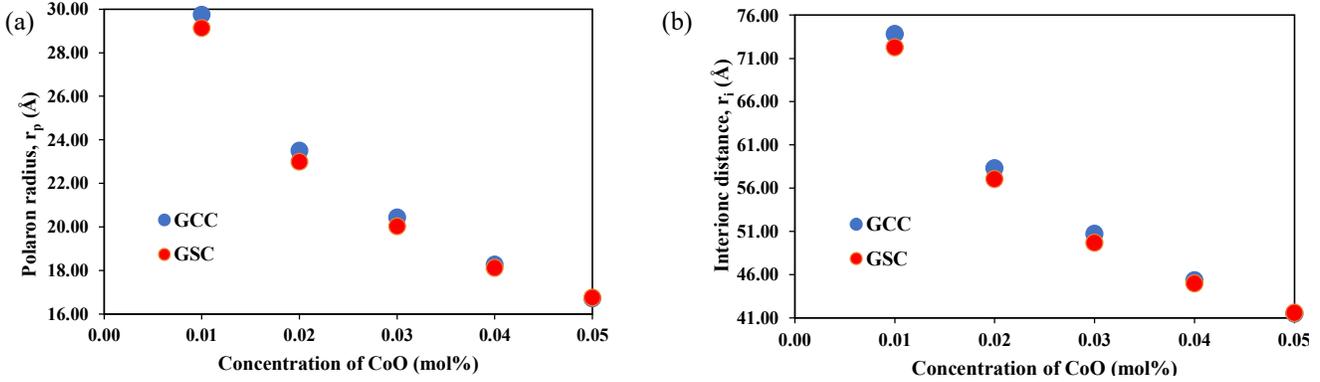

Fig. 10 (a) Polaron radius of glass doped CoO and (b) interionic distance of glass doped with various concentrations of CoO.
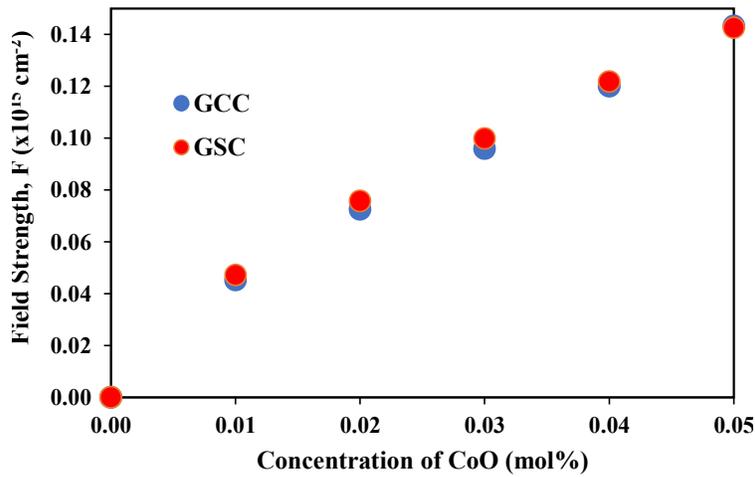

Fig. 11 field strength of glass doped with various concentrations of CoO.

The optical absorption spectra of CoO-doped glass, measured in the wavelength range of $350 - 2000$ nm for concentrations of 0.00, 0.01, 0.02, 0.03, 0.04 and 0.05 mol%, revealed absorption peaks at approximately 521, 576, 630 and 1238 nm. These peaks are attributed to energy level transitions of CoO: $^4T_{2g}(F) \rightarrow {}^2T_{1g}(H)$, $^4A_2(^4F) \rightarrow {}^4T_1(^4F)$, $^4T_2 \rightarrow {}^4E_1$ and $^4A_{2g}(^4F) \rightarrow {}^4T_{1g}(^4F)$ [19, 20]. The intensity of these peaks tends to increase with higher CoO concentrations, as shown in Fig 6.
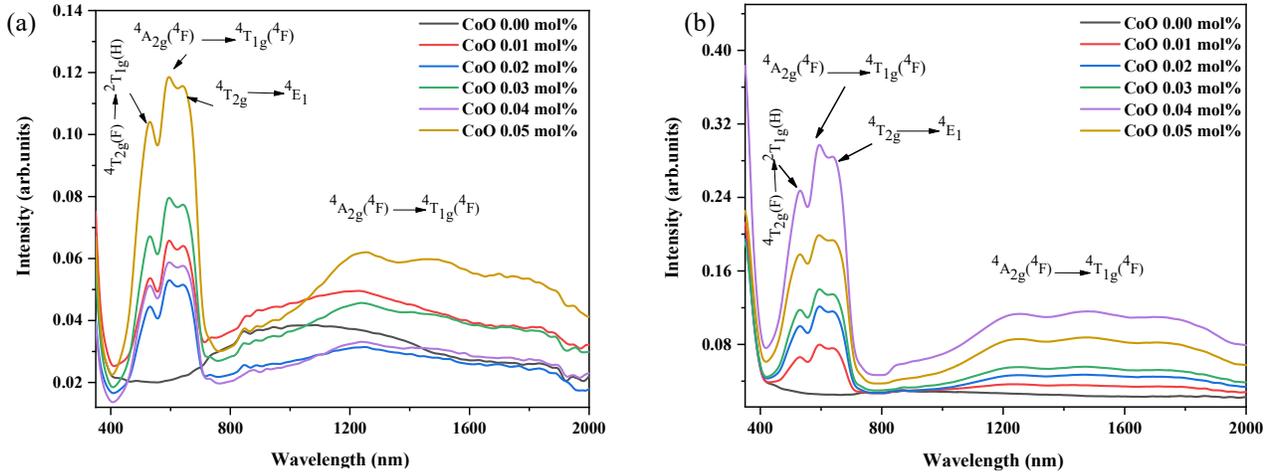
Fig. 6 (a) Absorption spectra of glass using SiO_2 chemical and (b) Absorption spectra of glass using sugarcane leaf ash with various concentrations of CoO.

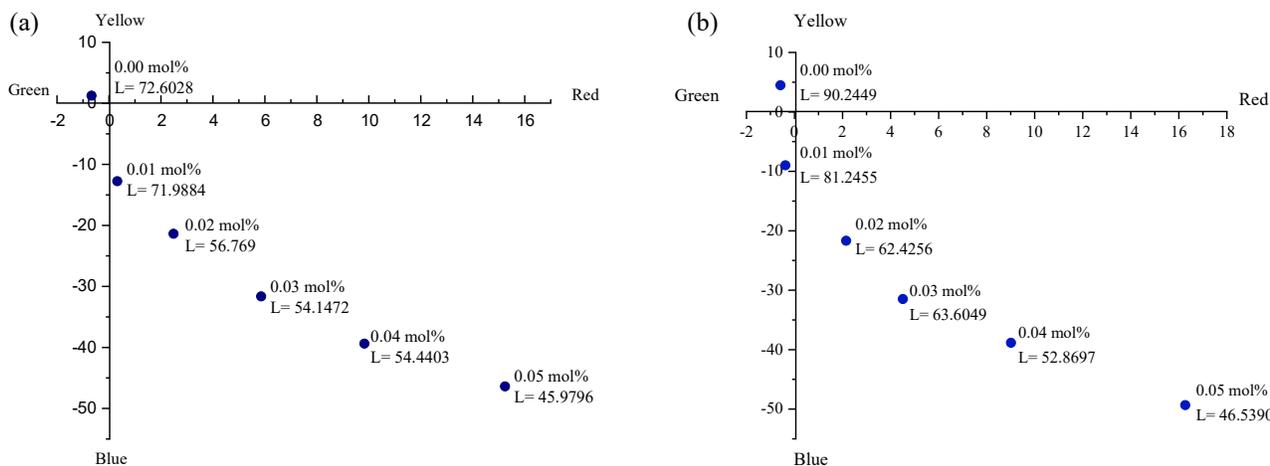*Results of color analysis in the system CIE L*a*b**

Analysis of the CIE lab colour space* analyses of glass filled with CoO at concentrations of 0.00, 0.01, 0.02, 0.03, 0.04 and 0.05 mol %. Glass using SiO_2 chemical, revealing that the brightness (L* values) decreases with increasing CoO concentration. The L* values range from 72.6028 − 45.9796, indicating a shift towards darker shades as the CoO content in the glass matrix increases. In addition to the brightness, the a* and b* chromaticity coordinates are also significantly affected by the CoO concentration. The a* values, representing the green–red axis, range from -0.6692 − 15.2403, while the b* values, representing the blue–yellow axis, range from -46.3978 − 1.2652. The glass using sugarcane leaf ash revealed that the brightness (L* value) decreased with increasing CoO concentration. The L* values ranged from 90.2449 − 46.5390, indicating a shift towards darker hues as the amount of CoO in the glass matrix increased. In addition to the brightness, the a* and b* chromaticity coordinates are also significantly affected by the CoO concentration. The a* values, representing the green–red axis, range from -0.5786 − 16.2743, while the b* values, representing the blue–yellow axis, range from -49.3306 − 4.4782, as shown in Table 4 and Table 5 Positive a* values indicate a reddish tint, while negative b* values indicate a tendency towards blue. The joint analysis of a* and b* coordinates shows that the color values of the glass samples lie on the positive and negative axis, corresponding to the red and blue regions of the color spectrum. In particular, a more negative b* value than a* indicates more blue than red, resulting in a predominantly blue-red color of the glass. Therefore, the glass appears blue color. This observation is confirmed by the absorption peak found in the range of 490 – 650 nm, which includes the blue region of the visible spectrum. This trend is visualized in Fig. 7, showing the color change with different CoO concentrations, to emphasize the significant effect of CoO on the aesthetic properties of the glass.

Table 4 (a) Color analysis in the CIE L*a*b* system of glass using SiO_2 chemical.

Concentration (mol%)	Color measurement (a)		
	L*	a*	b*
0.00	72.6028	-0.6692	1.2652
0.01	71.9884	0.3216	-12.7601
0.02	56.7690	2.4841	-21.3704
0.03	54.1472	5.8559	-31.6388
0.04	54.4403	9.8302	-39.3872
0.05	45.9796	15.2403	-46.3978

Table 5 Color analysis in the CIE L*a*b* system of glass using sugarcane leaf ash doped with various concentrations of CoO.

Concentration (mol%)	Color measurement (b)		
	L*	a*	b*
0.00	90.2449	-0.5786	4.4782
0.01	81.2455	-0.3719	-8.9864
0.02	62.4256	2.1551	-21.6751
0.03	63.6049	4.5148	-31.4869
0.04	52.8697	9.0162	-38.8517
0.05	46.5390	16.2743	-49.3306

Fig. 7 (a) Color analysis in the CIE L*a*b* system of glass using SiO_2 chemical and (b) Color analysis in the CIE L*a*b* system of glass using ugarcane leaf ash doped with various concentrations of CoO.

4. Conclusion

In this study, the physical and optical properties of cobalt oxide-doped glasses prepared by conventional chemical method and cobalt oxide-doped glasses using sugarcane leaf ash as SiO_2 and CaO source in the formula $(50-x)SiO_2$: $25B_2O_3$: $10Na_2O$: $8CaO$: $7SrO$: $xCoO$ with different CoO concentrations (0.00, 0.01, 0.02, 0.03, 0.04 and 0.05 mol%) were investigated and compared. The experimental results showed that the addition of CoO affected the physical and optical properties of the glasses. The density and refractive index of the chemically treated and sugarcane leaf ash-doped glasses increased with increasing CoO concentration, while the molar volume decreased. indicating a more compact structure. In addition, the obtained glasses were found to have an amorphous structure before and after the addition of CoO. The absorption spectra of the chemically treated glasses showed increased absorption intensity, especially at 521, 576, 630 and 1238 nm, which were consistent with the increasing concentration of CoO, resulting in the characteristic blue color. CIE L*a*b* color analysis confirmed that the glass samples displayed blue color. And the absorption spectra of the glasses using sugarcane leaf ash showed increased absorption intensity, especially at 521, 576, 630 and 1238 nm, which was consistent with the increasing concentration of CoO, resulting in the characteristic blue-red color. CIE L*a*b* color analysis confirmed that the glass samples displayed blue-red color, with the color intensity increasing with the amount of CoO added. These results suggest that sugarcane leaf ash is a suitable alternative traditional silica source for glass production. Overall, this research provides valuable insights into the potential utilization of agricultural waste for glass production, including decorative materials.

Acknowledgements

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