

STUDY OF PHYSICAL AND OPTICAL PROPERTIES OF SUGARCANE LEAF ASH GLASS DOPED WITH COPPER OXIDE

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Abstract

In this research, the physical and optical properties of sugarcane leaf ash doped with copper ions in glass were investigated. The composition of sugarcane leaf ash was analyzed at different sintering temperatures using X-ray fluorescence (XRF) spectroscopy, revealing the highest SiO₂ content (69 wt%). The glass formula was developed based on the ratio (50-x)SiO₂ (with sugarcane leaf ash substituting for SiO₂ and CaO): 25B₂O₃: 10Na₂O: 8CaO: 7SrO: xCuO, where x represents the concentration of copper oxide (0.00, 0.10, 0.20, 0.30, 0.40, and 0.50 mol%). The results showed that the density and refractive index increased with increasing CuO concentration, while the molar volume decreased. The absorption spectra of the glass in the wavelength range of 350-1,100 nm was measured and showed a peak around 775 nm, corresponding to the ²B_{1g} → ²B_{2g} transition of Cu²⁺. The studied glass exhibited greenish-blue color coordinates, confirmed by CIE Lab* color measurements.

Keywords: Sugarcane Leaf ash; CuO; Glass; Physical Properties; Optical Properties

Introduction

Currently, the central region of Thailand has extensive sugarcane cultivation across several provinces, including Kanchanaburi, Suphanburi, Ratchaburi, and Nakhon Pathom, covering a combined area of more than 2,696 km². Historically, there are two methods of harvesting sugarcane: the first is harvesting fresh sugarcane,

and the second is burning the sugarcane before harvesting, commonly known as 'burned sugarcane.' This latter method accounts for approximately 60% of the total sugarcane harvest. Burning sugarcane creates air pollution due to the emission of carbon monoxide and volatile organic compounds, which disperse into the air, reducing

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visibility (Khatjoi *et al.*, 2021; Kanjanaphan and Rachniyom, 2024). In burned sugarcane leaves, the most prevalent chemical component is silica (SiO_2), a major component that can be used in the glass industry (Ibrahim *et al.*, 2022).

Glass is an amorphous solid with a homogeneous structure. It has a fine texture consisting of crystals with a free structure and small size, which reduces the likelihood of glass fractures and results in a low thermal expansion coefficient. Today, glass materials are used in various fields, such as packaging, windows, and laboratory glassware, among others (Singkiburin *et al.*, 2023; Singkiburin *et al.*, 2024). Furthermore, previous research has utilized ash from agricultural waste, which primarily consists of silica, in the glass melting process (Wahab *et al.*, 2020, Ravangvong *et al.*, 2022). Copper doped glasses have been of interest for various applications such as nonlinear optics, lasers, lighting, spectral conversion in solar cells. The optical properties of the copper-containing glasses are known to depend ultimately on the oxidation and aggregation states of the metal (Jiménez, 2021).

Copper is transition to possess a rich redox chemistry and exquisite coordinative flexibility, a neutral copper atom has an electronic configuration of $3d^{10} 4s^1$. The Cu^+ (cuprous) ions produce no colouring to the glass resulting from a complete 3d shell with no d-d transition taking place, whereas the Cu^{2+} (cupric) ions gives colouring to glasses due to the presence of a d-d electronic transition from absorption of light in visible region in the incomplete d electronic shell. Thus, the presence of Cu^{2+} in glass system gives them a range of colour between skyblue and green colouration, depending on the glass's compositional base which are useful in numerous applications stretching from advanced materials (Tsaturyan Arshak *et al.*, 2019; Umar *et al.*, 2023). This enhancement in the glass structure and its physical properties caused by adding CuO make it suitable for many technological applications (Sh. Heikal *et al.*, 2023).

Given the importance mentioned above, the researchers are interested in using sugarcane leaf ash as a component in glass preparation. Therefore, they studied the chemical composition of sugarcane leaf ash subjected to different temperatures using X-ray Fluorescence Spectrometry (XRF) for glass production. Subsequently, the sugarcane leaf ash was incorporated into glass to investigate its physical and optical properties, including density, molar volume, refractive index, light absorption, and color, with CuO doping at concentrations where x is 0.00, 0.10, 0.20, 0.30, 0.40, and 0.50 mol%. This study aims to provide guidelines for the further

development of glass products across various applications.

Materials and Methods

In this research simulated the burning of sugarcane leaf at different temperatures using an electric furnace mixed with oxygen. 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. It was noted that increasing the sintering temperature does not affect the amount of silica (SiO_2) (Kanjanaphan and Rachniyom, 2024), 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

The glass formula was developed based on the ratio $(50-x)\text{SiO}_2$ (with sugarcane leaf ash substituting for SiO_2 and CaO): $25\text{B}_2\text{O}_3$: $10\text{Na}_2\text{O}$: 8CaO : 7SrO : $x\text{CuO}$, where x represents the concentration of CuO (0.00, 0.10, 0.20, 0.30, 0.40, and 0.50 mol%). The glass formula design was based on the volumes of SiO_2 and CaO measured by the XRF instrument, resulting in the following ratio: $\text{SiO}_2 = 69.70$ wt% and $\text{CaO} = 17.88$ wt%. The amount of SiO_2 is three times that of CaO . In the process of weighing the substances, the amounts of SiO_2 and CaO were calculated based on their percentage by weight. According to mathematical principles, weighing of $\text{SiO}_2 = 11.4878$ g, and weighing of $\text{CaO} = 3.0615$ g from sugarcane leaf ash. The components were weighed, mixed 5 minutes, and then melted in an electric furnace at 1,200°C for 3 hours. The molten glass was poured into graphite molds and annealed at 500°C for

3 hours, followed by cooling at room temperature. After the glass is annealing at 500°C, the cooling process is important. The cooling process should be gradual, cooling about 0.55°C per 1 minute to reduce heat stress and improve the quality and strength of the glass. The cooled glass samples were cut to dimensions of $1.0 \times 1.5 \times 0.3 \text{ cm}^3$. These samples were then analyzed for physical and optical properties, including density (Densitometer, A&D HR-200), molar volume, refractive index (Abbe refractometer, Atago), Absorption spectrum (UV-VIS-NIR Spectrophotometer, Shimadzu UV-3600), and color coordinate (UV-VIS spectrophotometer, Cary-50).

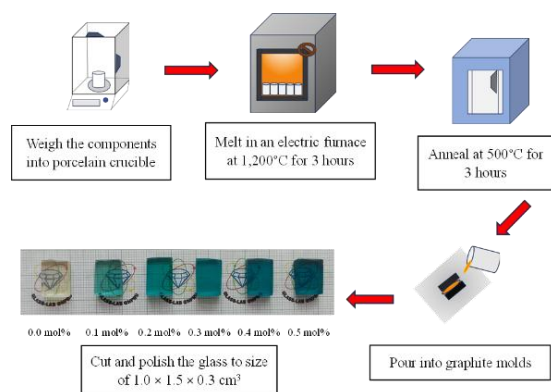


Figure 1. Preparation of glass with sugarcane leaf ash substituting for SiO_2 and CaO doped CuO

Results and Discussion

In the experiments on melting glass doped with CuO , prepared using the glass formula based on the ratio $(50-x)\text{SiO}_2$ (with sugarcane leaf ash replacing SiO_2): $25\text{B}_2\text{O}_3$: $10\text{Na}_2\text{O}$: 8CaO : 7SrO : $x\text{CuO}$, with CuO concentrations of 0.00, 0.10, 0.20, 0.30, 0.40, and 0.50 mol%, it was observed that the glass without CuO was clear with a slight yellow tint due to iron ions. After doping with CuO , the glass displayed a blue-green color, and as the CuO concentration increased, the glass became a more intense blue-green, as shown in Figure 2.



Figure 2. Glass doped with CuO at various concentrations

Analysis of the density values of CuO -doped glass with concentrations of 0.00, 0.10, 0.20, 0.30, 0.40, and 0.50 mol%, conducted using Archimedes' principle, revealed that density tends to increase with higher CuO concentrations, as shown in Table 2. The measured density values ranged from 2.6615 ± 0.0040 to $2.8242 \pm 0.0053 \text{ g/cm}^3$, reflecting the increasing incorporation of CuO into the glass matrix. This increase in density can be attributed to the higher molecular weight of CuO compared to SiO_2 . As CuO replace SiO_2 in the glass formulation, the overall molecular composition becomes denser. This substitution alters the atomic arrangement within the glass structure, enhancing packing efficiency. The presence of heavier copper ions (Cu^{2+}) significantly contributes to this density increase, as they occupy positions in the glass network that would otherwise be filled by lighter silicon ions (Si^{4+}). When plotting the relationship between density and CuO concentration, the resulting graph demonstrates a strong linear relationship (Singkiburin *et al.*, 2023) with a correlation coefficient (R^2) of 0.9670, as shown in Figure 3. This high correlation coefficient indicates a robust linear dependence of density on CuO concentration, reinforcing that precise control over CuO doping can effectively tailor the density of the glass. The linearity of this relationship also suggests that each incremental addition of CuO consistently impacts the overall density, making it a valuable parameter for predicting the material's properties based on its composition.

Table 2. Density, Molar volume, and Refractive index of glass doped with CuO at various concentrations

Concentrations of CuO (mol%)	Density, ρ (g/cm^3)	Molar volume, V_M (cm^3/mol)	Refractive index
0.00	2.6615 ± 0.0040	25.9972	1.6482 ± 0.0006
0.10	2.6930 ± 0.0015	25.7370	1.6510 ± 0.0005
0.20	2.7087 ± 0.0011	25.6323	1.6526 ± 0.0001
0.30	2.7477 ± 0.0021	25.3121	1.6533 ± 0.0001
0.40	2.7828 ± 0.0016	25.0357	1.6556 ± 0.0003
0.50	2.8242 ± 0.0053	24.7110	1.6574 ± 0.0005

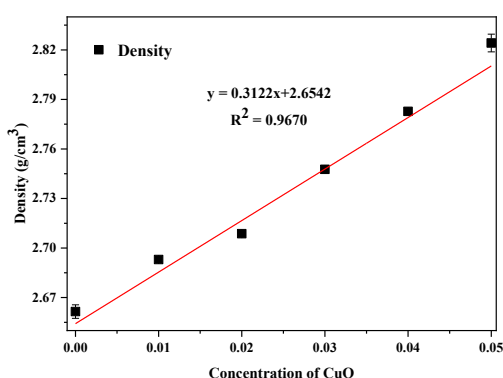


Figure 3. The density of glass doped with CuO at various concentrations

The molar volume, calculated based on the ratio of molecular weight to density, indicates a decreasing trend with increasing CuO concentration (Issever *et al.*, 2021). This calculation is critical understanding how the incorporation of different components affects the glass structure. As shown in Table 2, the molar volume values ranged from 25.9972 to 24.7110 cm³/mol. This reduction in molar volume suggests that as CuO concentration increases, the glass network becomes more compact. The observed decrease in molar volume can be attributed to the interaction of CuO with bridging oxygen atoms in the glass matrix. When CuO is introduced, copper ions replace some of the silicon ions typically present in the silicate network. This substitution alters the network's connectivity, allowing for stronger bonding interactions with bridging oxygens, thereby reducing the available space within the glass structure. Furthermore, this phenomenon indicates that higher CuO concentrations promote the formation of non-bridging oxygen species, further stabilizing the glass network. Consequently, the overall density of the glass increases, while the molar volume decreases, illustrating the intricate balance between composition and physical properties. The resulting graph demonstrates a strong linear relationship with a correlation coefficient (R^2) of 0.9441, this trend is visually represented in Figure 4, which graphically depicts the correlation between CuO concentration and molar volume, highlighting the significant impact of CuO on the structural characteristics of the glass.

Analysis of the refractive index values of CuO-doped glass with concentrations of 0.00, 0.10, 0.20, 0.30, 0.40, and 0.50 mol%, conducted using an Abbe refractometer, found that the refractive index tends to increase with higher CuO concentrations, as shown in Table 2. The refractive

index values ranged from 1.6482 ± 0.0006 to 1.6574 ± 0.0005 , indicating a consistent increase as the CuO content was elevated. This trend is similar to that of density and aligns with traditional dielectric theory, which posits that the refractive index is influenced by both the density of the material and the polarizability of its constituent atoms (Singkiburin *et al.*, 2024). Specifically, the presence of CuO introduces heavier copper ions into the glass matrix, which increases the overall polarizability and density, thereby elevating the refractive index. The relationship between the refractive index and CuO concentration, the resulting graph demonstrates a strong linear correlation, with a coefficient (R^2) of 0.9619, as shown in Figure 5. This high correlation indicates a reliable relationship between CuO concentration and the optical properties of the glass, suggesting that careful control of CuO doping could be employed to tailor the refractive index for specific applications in optics and photonics.

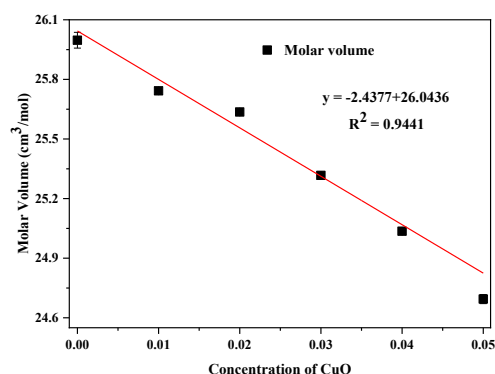


Figure 4. The density of glass doped with CuO at various concentrations

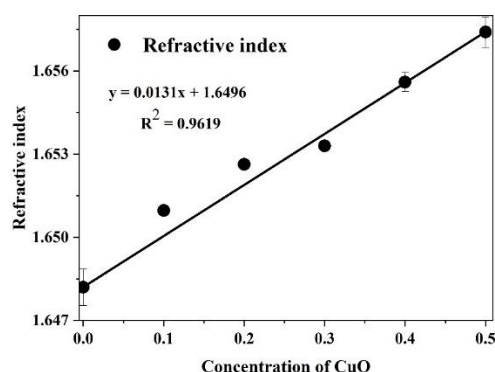


Figure 5. The refractive index of glass doped with CuO at various concentrations

X-ray diffraction (XRD) analysis of the CuO-doped glass revealed that the XRD pattern consisted of fluctuations and no diffraction peaks are observed confirming the non crystalline nature of the glassy system (Attallah *et al.*, 2018, Jiménez, 2021). Therefore, no significant changes were observed before and after the sequential addition of copper oxide, as shown in Figure 6.

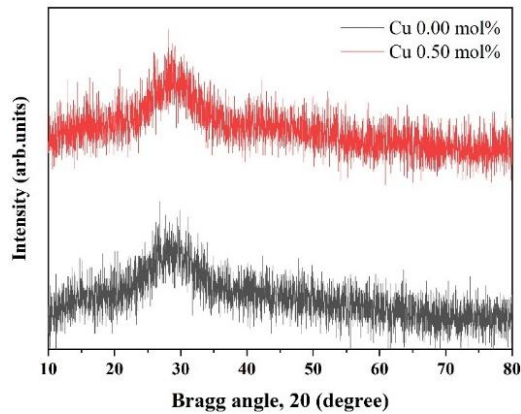


Figure 6. X-ray diffraction (XRD) of glass doped with various concentrations of CuO

The absorption spectra of CuO-doped glass with concentrations of 0.00, 0.10, 0.20, 0.30, 0.40, and 0.50 mol% were measured across the wavelength range of 350-1,100 nm. The spectra exhibited a distinct absorption peak around 775 nm, corresponding to the $^2B_{1g} \rightarrow ^2B_{2g}$ electronic transition of Cu^{2+} ions (Yonphan *et al.*, 2021; Ibrahim *et al.*, 2023), which is characteristic of the d-d transitions in copper complexes. As the concentration of CuO increased, the absorption intensity at this peak also increased, indicating a greater presence of Cu^{2+} ions in the glass matrix. This behavior suggests that the incorporation of CuO not only influences the optical properties but also enhances the interaction of the glass with light in this wavelength range, as shown in Figure 7. The increase in absorption with higher CuO concentrations is consistent with the elevated presence of copper ions in the glass, which contributes to the observed blue-green coloration.

Analysis of the CIE Lab color space* for CuO-doped glass with concentrations of 0.00, 0.10, 0.20, 0.30, 0.40, and 0.50 mol% revealed that brightness (L^* value) decreases as the CuO concentration increases. The L^* values ranged from 73.9537 to 56.4379, indicating a shift towards darker shades

with increasing CuO content in the glass matrix. In addition to brightness, the a^* and b^* color coordinates were significantly affected by CuO concentration. The a^* values, representing the green-red axis, ranged from -28.7771 to -0.8374, while the b^* values, representing the blue-yellow axis, varied from -17.2961 to 5.1837, as shown in Table 3. The negative a^* values indicate a predominant green hue, while the negative b^* values suggest a tendency toward blue. A combined analysis of the a^* and b^* coordinates shows that the glass samples' color values are located on the negative axes, corresponding to the green and blue regions of the color spectrum. Notably, the higher negative values of a^* compared to b^* indicate a stronger green presence than blue, resulting in a distinctive blue-green coloration for the glass. This trend is visually represented in Figure 8, where the color shifts across different CuO concentrations are illustrated, emphasizing the significant impact of CuO on the aesthetic properties of the glass (Yonphan *et al.*, 2021; Mahingsa *et al.*, 2023).

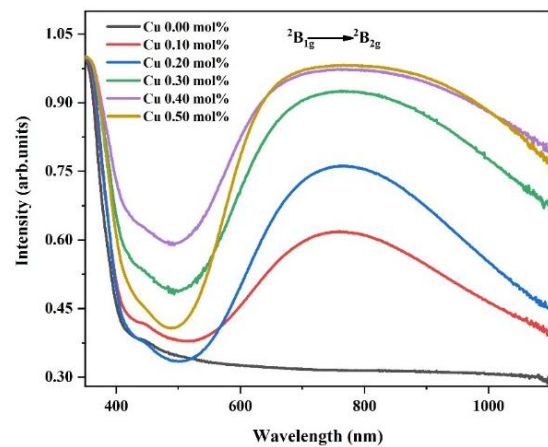


Figure 7. The absorption of glass doped with CuO at various concentrations

Table 3. Color value of CuO doped glass at various concentrations

Concentrations (% mol)	Color measurement		
	L^*	a^*	b^*
0.00	73.9537	-0.8374	5.1837
0.10	68.0900	-9.8681	-0.8326
0.20	69.6889	-17.5614	-5.6596
0.30	63.3727	-19.7111	-8.6911
0.40	57.8047	-24.8327	-14.0848
0.50	56.4379	-28.7771	-17.2961

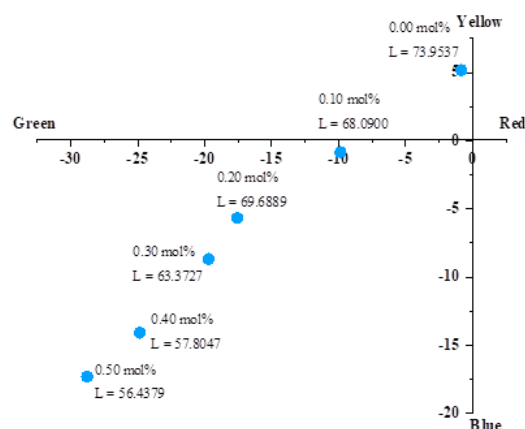


Figure 8. Glass doped CuO show CIE L*a*b*

Conclusions

In this study, glass was successfully synthesized using sugarcane leaf ash as a silica (SiO_2) replacement in the formula $(50-x)\text{SiO}_2: 25\text{B}_2\text{O}_3: 10\text{Na}_2\text{O}: 8\text{CaO}: 7\text{SrO}: x\text{CuO}$, with varying CuO concentrations (0.00, 0.10, 0.20, 0.30, 0.40, and 0.50 mol%). The results demonstrated that doping with CuO significantly influenced both the physical and optical properties of the glass. Specifically, the density and refractive index were observed to increase with higher CuO concentrations, while the molar volume decreased, indicating a more compact structure. It was also found that the glass obtained had an amorphous structure before and after doping with CuO. The absorption spectra showed enhanced absorption intensity, especially at 775 nm, corresponding to the Cu^{2+} ion transitions, with increasing CuO concentration contributing to the characteristic blue-green color. The CIE L*a*b* color analysis confirmed that the glass samples displayed a blue-green hue, with the intensity of color increasing as more CuO was added. These findings suggest that sugarcane leaf ash is a viable alternative to traditional silica sources for glass production, and CuO doping provides a useful method for tuning the glass's optical properties, particularly in terms of color and refractive index. Overall, this research offers valuable insights into the potential use of agricultural waste in glass manufacturing, promoting sustainability while maintaining high-quality glass properties. Further studies could explore the application of this glass in various industries, including optics, photonics, and decorative materials.

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