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Effects of CuO on glass prepared from local sand in Nakhon Pathom **Province**

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Abstract

In this work, sand was obtained from pit sand in district Kamphaeng Saen, Nakhon Pathom province. This sand was investigated the chemical compositions and the crystal structures before use it as raw material to prepare glass samples. The glass samples were prepared in compositions of (49-x)SiO₂: 7.5B₂O₃ : 8CaO : 10SrO₂: 0.5Sb₂O₃: 25Na₂O : x CuO (where x = 0.0 , 0.2 , 0.4 , 0.6 , 0.8 , 1.0 mol%) by melt quenching technique. After that, all glasses were cut and polish in proper shape for measurement in density, refractive index, hardness and optical properties. The results of sand sample observed major components with SiO_2 (wt.%), K₂O (wt.%) and Fe₂O₃ (wt.%). The crystalline structure of sand has shown large of quartz phase. For the results of glass sample, the density values of glasses were between 2.818–2.834 g/cm³. The refractive index values were in range 1.6523-1.6532. The Vicker's hardness values were widely around 319.28-410.94 without relative with the increasing of CuO concentration. The absorption spectra of glasses were occurred the contamination of Co^{2+} , Fe^{2+} and Fe^{3+} from crucibles and raw material. In addition, the effects of CuO were shown board peaks of absorption spectra around 780 nm., and they were increased with increasing of CuO concentrations. The colors of glasses were shown in green and yellow color which stronger and brightness with increasing CuO concentration.

Keywords: glass, local sand, quartz

1. Introduction

Current, a lot of sand mining occurs widely in Nakhon Pathom province. Especially, sand mining located in district Kamphaeng Saen, Bang Len and Nakhon Chai Si [1]. These sands were often utilized in constructions, building, road and land field. The price of local sands in Nakhon Pathom were low because they are considered low grade. In fact, local sands can add value to use in other industries such as glass production and industrial iewellery.

Silica or glass sand is a type of quartz sand which good for glass production [2]. Silica sand has various levels of purity, but small proportion in Thailand which suitable for glass production or high-grade industrial applications [3]. Glass sand that suitable for glass production should be consists with high silica content, low iron, transition metal and another compound. The iron oxide or transition metal iron caused be effect on color of glass and other physical properties such as density, refractive index, durability, etc. Sand is a natural raw material that has various components according to the geology of source. So,

this work was examined properties of local sand in Nakhon Pathom for utilization in glass production

This work, we analyzed sand sample from pit sand in Amphoe Kamphaeng Saen by energy dispersive xray fluorescence (EDXRF) for investigating chemical composition and by X-ray Diffractometer (XRD) for crystalline structures. The glass samples were prepared with local sand as SiO₂ content. The CuO was varied to replaced SiO₂ and investigated properties in density, refractive index, hardness, absorption spectra and CIE L*a*b.

2. Materials and methods

Sand in this work is used as SiO₂ for preparing glass. It was procured from sandpit in Nakhon Pathom Province. The chemical compositions of it were analyzed with an energy dispersive x-ray fluorescence (EDXRF), model Panalytical Minipal-4 spectrometer. The crystal structures of sand were investigated by X-ray Diffractometer, model Shimadzu 6100 with CuK_a radiation $(\lambda = 1.5406 \text{ Å})$ over the 2 θ range 10°–80°.

The glass samples in this work were prepared in formula: (49-x)SiO₂ : 7.5B₂O₃ : 8CaO : 10SrO₂:

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Chemical compositions (% by weight)								
SiO ₂	K ₂ O	CaO	TiO ₂	MnO	Fe ₂ O ₃	CuO	Rb ₂ O	SrO
85.465	8.223	1.554	0.891	0.102	3.621	0.008	0.077	0.02

Table 1 Chemical compositions of sand sample



Figure 1 XRD pattern of sand sample

 $0.5Sb_2O_3$: 25Na₂O : x CuO (where x = 0.0, 0.2, 0.4) 0.6, 0.8, 1.0 mol%) by melt quenching technique. All chemicals: B2O3, CaO, SrO, Sb2O3, Na2O and CuO used in this work were of analytical reagent grade. The raw materials were mixed in alumina crucible and placed in an electrical furnace for melting at 1,200 °C. These melts were soaking at this temperature for 4 hours and casted in stainless steel mould at the room temperature. After that, the glasses were annealed 500 °C for 3 hours and leave it to cool down slowly to room temperature. The glasses were cut and polish in same proper shape and size for characterization the density was determined by the Archimedes method and xylene was used as an immersion fluid. The reflective indices at room temperature were measured by the abbe refractometer (ATAGO-3T) with using mono-bromonaphthalene as an adhesive coating. The UV-Visible-NIR spectrophotometer (Shimadzu UV-3600) was used for analyzed optical spectra of glass samples in the range 200-2000 nm. The Vicker's hardness (Enkay DHV-1000) was measured by indentation method with selected load of 0.1 kg and fixed time of indentation as 10 s.

3. Results and discussion

The EDXRF reported the chemical compositions according to Table 1. It can observe that SiO_2 are major content with 85.465 wt.% of sand. Also found

large amount of K_2O and Fe_2O_3 around 8.223 wt.% and 3.621 wt.%, respectively. The XRD pattern of sand was presented in figure 1. included Quartz phase. This sand has been shown major crystalline structure in quartz phase.

For the glass samples, the addition of various CuO has been effect on physical properties of glasses. The density properties were in range 2.818–2.834 g/cm³. The density properties of glasses are shown in Figure 2. It cannot be found relation between the increasing of CuO concentrations and density properties. Probably due to sand in this experiment contained large of Fe₂O₃ content. So, the replacement of CuO in sand may be like the replacement of Fe₂O₃ with CuO. The molecular weight of Cu 63.546 (g/mol) is close to Fe (55.847 g/mol) same with results of density values were similar. The refractive index properties of glass samples were same trend with density properties. These results are corresponding with the classical dielectric theory, the refractive index values were depended with density [4]. Figure 3 show the refractive index values of glasses in range 1.6523-1.6532. The hardness values of glasses are shown in figure 3. The glass with 0.2 mol% of Cu has low hardness value at 319.28 HV and the best in this experiment is 0.8 mol% concentration of Cu with 410.94 HV.



Figure 2 Density and refractive index properties of glass samples



Figure 3 Vicker's hardness values of glass samples



Figure 4 Absorption spectra of glass samples



Figure 5 CIE L*a*b* color scale of glass samples

The absorption spectra of glasses are shown in figure 4. It can observe that the absorption band of all glass samples were occurred the pattern of absorption bands of Co²⁺ with very intense absorption band centered about 600 nm [5]. It is possible that they were caused by contamination from the alumina crucible. However, all glass samples were showed the shoulder absorption bands at wavelength of about 440 nm which are due to $\operatorname{Fe}^{3+}({}^{6}\operatorname{A}_{1g}(S) \rightarrow {}^{4}\operatorname{T}_{2g}(G))$ (Fig.4a.) and single board peaks around 1,050 nm due to Fe^{2+} $({}^{5}E_{g} \rightarrow {}^{5}T_{2})$ (Fig.4b.). Ferric iron gives a yellow to brown color of glass samples [6]. Ferrous and ferric ions were contaminated from sand as a result of EDXRF (table 1). In part of absorption band were caused by various concentration of CuO. The glasses were showed the shoulder peaks about 780 nm due to $Cu^{2+} ({}^{2}B_{1g} \rightarrow {}^{2}B_{2})$ [4]. The absorption band of Cu^{2+} were increased with increasing concentration of CuO.

For the colorants of glasses, the glass without additive CuO is yellow color and green. This caused by effect of Fe_2O_3 which obtained from raw material. It is corresponding with the EDXRF results in table 1. The increasing of CuO concentration have been affected to more yellow and green as shown in figure 5. The brightness, L* were decreased with increasing concentrations of CuO.

4. Conclusions

Local pit sand in Kamphaeng Saen are consist large amount of SiO₂ (85.456 wt %), K₂O (8.223 wt.%) and Fe₂O₃ (3.621 wt.%). This sand has quartz phase with major crystalline structure. For the glass sample, the density values and refractive index values were similar trend with increasing concentration of CuO. The hardness values were in range 319.28410.92 HV. The increasing of CuO concentration cannot be found the relation between hardness values and CuO content. The absorption spectra of glasses were found the contamination of Co^{2+} from crucible, and Fe²⁺ and Fe³⁺ from raw material. The absorption spectra od Cu²⁺ were observe board peak at 780 nm. and increased with increasing concentration of CuO. The colors of glass samples were in green and yellow. They were more strong color and brightness when increased CuO contents.

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