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LUMINESCENCE PROPERTIES OF (70-X) $ZN_2$30SIO_4DOPED WITH Sm^{3+}via CONVENTIONAL SOLID-STATE TECHNIQUE

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ABSTRACT

In this work optical properties of zinc silicate (willemite) glass ceramics doped with samarium oxide at different weight concentrations were determined. The melt quench technique was used to synthesis the samples, with waste rice husks as the silicate source. The energy band gap of samarium doped willemite glass ceramics decreased with increase of samarium dopant concentrations. This indicated the absorption of ultraviolet radiation by the samples.The photoluminescence measurements revealed the red emission which corresponded to the $^4G_{5/2} \rightarrow {}^6H_{11/6}$ (646.71 nm) under the blue excitation.

INTRODUCTION

Amidthe frequent host luminescent materials, zinc silicate (Zn_2SiO_4) is known as a suitable host matrix to integrate with rare earth ions and transition metal ions developing in a first luminescence region in blue, green and red spectra (Reddy et al., 2014). In modern technology, rare earth ions are better dopants for light activators in the host matrix materials than transition metals. The outermost orbitals of $^5S_2$ and $^5P_6$ completely sheltered the filled 4f shells. The emission changes supply high-pitched lines in the optical spectra accommodating phosphors with excessive luminescence efficiency. Moreover, the rare earth (RE) ions have drawn attention to substituting as an efficient ion in many optical materials due to their large number of absorption and emission bands surfacing from the transitions within the energy levels. Given these factors, many scientists focused their interest on trivalent samarium ions as luminescence centers in deep red emission arising from conduction band $^4G_{5/2} \rightarrow {}^6H_{9/6}$ transition (Sánchez-Royo et al., 2014). Currently, the samarium (Sm^{3+}) ion is one of the most attractive and Valuable rare-earth dopants considering Sm^{3+}-doped phosphors are common to be favorable materials for optical amplifiers, lasers, and electroluminescent devices. Several approaches have been grown to prepare Zn_2SiO_4: Sm^{3+} such as sol-gel synthesis, spray pyrolysis, co-precipitation synthesis, conventional solid-state reaction and hydrothermal method. As a contrast, the stable states technique aims a better clarity in the industrial application than sol-gel method. However, this technique regularly used very cost in SiO_2 source to synthesize the zinc silicate. To overwhelm the disadvantage, it requires changing new silicate sources from waste rice husks silica (WRHS) glasses for expense saving.

The husks silicate glass established to be an appropriate optical material with prominent transparency, excellent rare-earth ion solubility, high thermal stability, and low melting point. Hence, this paper intends to study the synthesis and optical properties of Sm^{3+} doped Zn_2SiO_4host matrix by adopting an inexpensive solid-state reaction method. Effects of Sm_2O_3 variation concentration in Zn_2SiO_4 (willemite) on the structural and optical properties were investigated.

METHODOLOGY

The conventional solid-state technique was used to produce (70-X) $Zn_2XSm30SiO_4$ based glass ceramics. The doping proportions were 1, 3 and 5wt% of the samarium oxide REacton 99.9 %. Zinc oxide 20 nm particle size with 99.5% purity was mixed with 63-micron mesh of silica. The silica was acquired from waste rice ash. The mixing procedure was carried out for 24 hours with low energy ball milling. The mixture was heated in a furnace at 1500 ℃ for six hours. The molten mixture was quenched in water and formed a glass fritz at room temperature(Syamimi et al., 2014). Then the glass fritz was crushed and sieved with 63 microns mesh. Polyvinyl alcohol (PVA) acted as a binder to the samples powder. The sample powder was turned to pellets, with 3.5 tons of pressure for 5 min and sintered at temperatures of 1000℃. At room temperature, Shimadzu UV– visible spectroscopy performed absorption spectra measurements in the range of 217.49–800 nm wavelengths. The optical band gaps of the material was analyzed the sample absorption edges. At room temperature, Perkin Elmer LS 55 Fluorescence spectrometer was used for photoluminescence excitation and emission spectra measurements.

RESULTS AND DISCUSSION

Optical absorption measurements

The optical absorbance of (70-X) $Zn_2XSm30SiO_4$ was characterized by UV–Vis spectroscopy in Figure 1 revealed the absorption spectra of samarium oxide doped Zn_2SiO_4 in the spectral zone of 220 to 800 nm at room temperature. The shifting of the absorption band to the lower wavelength agreed to the decrease in non-bridging oxygen preparing the structure to be more compact. The absorption spectra of the samples were improved due to dispersing of Zn_2SiO_4 in the crystals lattice(Amjad et al., 2012).

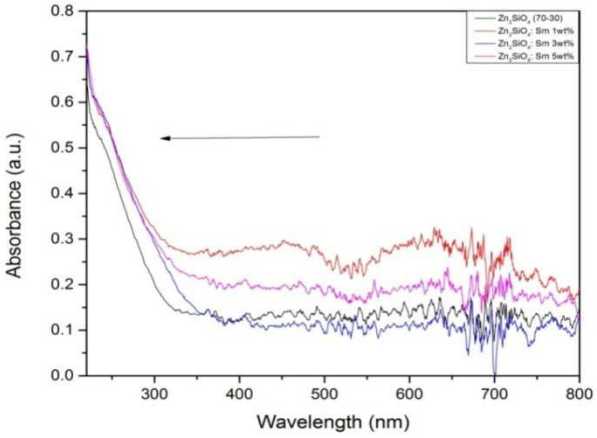

Figure 1. Absorption spectra of(70-X) $Zn_2XSm30SiO_4$ at X =0, 1, 3 and 5 wt%

Optical band gap

The valence and conduction bands transition of samarium doped Zn_2SiO_4 absorption spectra were accomplished to calculate the band gap(Sahu et al. 2015). The UV–Vis spectrum of the pure Zn_2SiO_4 crystal was predicted to have a band gap of 5.5 eV. Figure 2 showed that the undoped Zn_2SiO_4 had the band gap energy (Eg) of 3.88 eV before the samarium addition. In the event of Zn_2SiO_4,

the indirect transition connection between incident photon energy (hυ) and absorption coefficient (α) had a typical relation[25]:$(\alpha h\nu)^{1/2} = \mathrm{k}\left[\mathrm{h}\nu - E_g\right]$.Where k is proportionality constant and E_g was the material optical energy band gap. The direct band gap values were extrapolated from the linear portion of the absorption curves. The variations of dopants concentrations of 1, 3 and 5 wt% yielded the energy band gaps of 3.29, 3.48 and 3.20eV respectively.

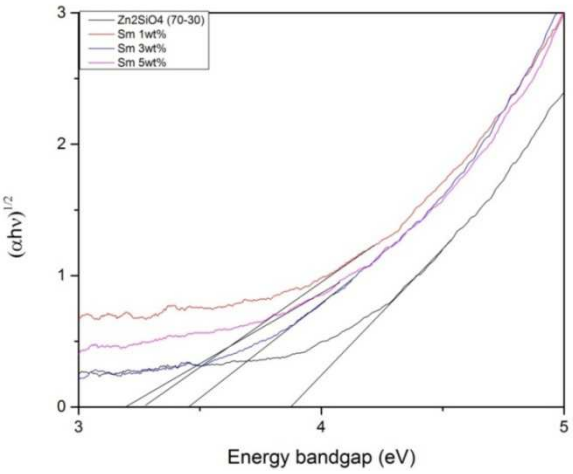

Figure 2. Optical band gap of (70-X) $Zn_2XSm30SiO_4$ at X =0, 1, 3and 5 wt%

Figure 3 exhibited the differences of the energy band gap, with different dopant concentrations of (70-X) $Zn_2XSm30SiO_4$. The energy band gap decreased linearly as the concentration of samarium oxide increased to the pure Zn_2SiO_4. The higher the dopant concentrations led to the increase of non-bridging oxygen and reduced the bridging oxygen. The absorption band tended to move toward the higher wavelength. In this regard, the sample navigated toward the lower frequency and energy respectively. The increase of the dopant concentrations led to the broadening of impurity band and reduced the energy band gap(Sharma & Bhatti, 2009). The broadening impurity band was simply the band that was located in between valence and conducting band(Erdinc et al., 2010).

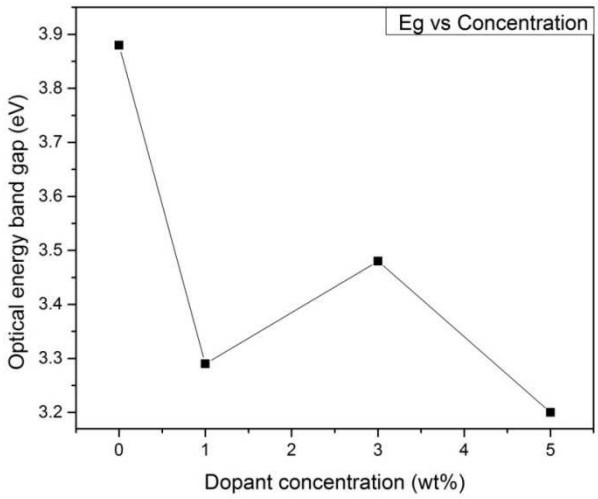

Figure 3. Variations samarium oxides dopant concentrations against optical energy band gap

Photoluminescence

The photoluminescence excitation results for (70-X) $Zn_2XSm30SiO_4$were shown in Figure 4. The excitation spectra demonstrated four important peaks at 362.26, 376.80, 389.61 and 406.80 nm with the ground state ($^4G_{5/2}$) transitions indicating excited levels. The excitation of 4f-f levels of Sm^{3+} were occupied because of energy transfer from the excited level to the triplet level, which was employed by rapid intersystem crossing(Bakier 2005). The major peak was positioned at 406.80 nm because of the transitions from $^6H_{5/6} \rightarrow {}^4G_{5/2}$. The samples experienced high reduction of Luminescence intensity due to quenching effect that occurred in the glass matrix. The addition of Sm^{3+} ions led to the increase of the phonon energy.

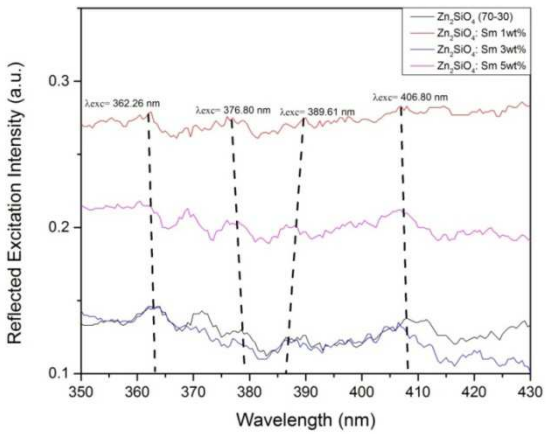

Figure 4.Excitation spectra of (70-X) $Zn_2XSm30SiO_4$ at X =0, 1, 3and 5 wt%

The peak verified Sm^{3+} emission spectra (Fig.5) The blue light (406.80 nm) excitation disclosed four emission spectral bands at 570.50, 595.11, 622.09 and 646.71 nm that were accredited to ($^4G_{5/2} \rightarrow {}^6H_{5/6}$), ($^4G_{5/2} \rightarrow {}^6H_{7/6}$), ($^4G_{5/2} \rightarrow {}^6H_{9/6}$) and ($^4G_{5/2} \rightarrow {}^6H_{11/6}$) respectively. The red shift emission peak originated at 646.71 nm ($^4G_{5/2} \rightarrow {}^6H_{11/6}$) which was ascribed to the conscious electric dipole transitions of Sm^{3+}. Hence, the peak at 570.11 nm ($^4G_{5/2} \rightarrow {}^6H_{5/6}$) was accredited to magnetic dipole transitions(Marzouk et al., 2015). As shown in Figure 5. , the emission intensity of ($^4G_{5/2} \rightarrow {}^6H_{11/6}$) transition was decreased due to broken effect of the sample(Brunold, C.T., Gudel, H.U., Cavalli, 1996).

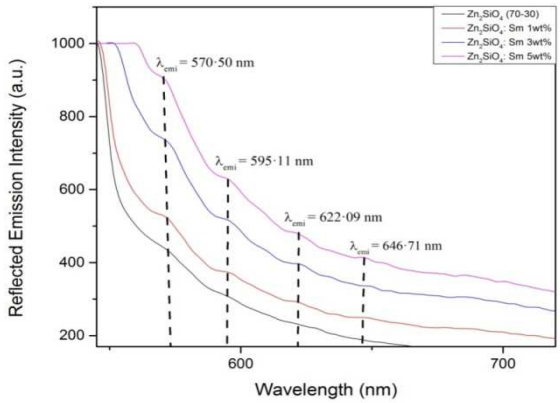

Figure 5. Emission spectra of (70-X) $Zn_2XSm30SiO_4$ at X =0, 1, 3and 5 wt%

CONCLUSION

A melt quench technique was successfully used to synthesize Sm^{3+} doped Zn_2SiO_4 with different weight concentrations. The photoluminescence revealed a red emission at 646.71 nm because of the Sm^{3+} intra ionic transitions at 406.80 nm excitation. The broadening figures of the excitation and emission spectra were showing the high quenching yield of the sample, which led to the broken effect.

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