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Low cost phosphors: Structural and photoluminescence properties of Mn^{2+}-doped willemite glass-ceramics

Nur Farhana Samsudin [a], Khamirul Amin Matori [a,b,*], Zaidan Abdul Wahab [b], Josephine Ying Chi Liew [b], Yap Wing Fen [a,b], Sidek Hj. Ab Aziz [b], Mohd Hafiz Mohd Zaid [b]

[a] *Materials Synthesis and Characterization Laboratory, Institute of Advanced Technology, Universiti Putra Malaysia, 43400 UPM Serdang, Selangor, Malaysia*
[b] *Department of Physics, Faculty of Science, Universiti Putra Malaysia, 43400 UPM Serdang, Selangor, Malaysia*

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ABSTRACT

In this study, Mn^{2+}-doped willemite glass-ceramics were prepared from conventional melt-quenching method derived MnO-ZnO-SLS precursor glass by an isothermal heat-treatment process where waste SLS glass bottle are used as a silicon source. The structure, phase morphology and luminescent properties of the Mn^{2+}-doped willemite phosphors were characterized using X-ray diffraction (XRD), field emission scanning electron microscopy (FESEM) and photoluminescence (PL) spectroscopy. The XRD pattern revealed the phase transformation of zinc orthosilicate (α-Zn_2SiO_4) with the progression of heat treatment temperature. The FESEM images showed the aggregated and irregularity in shape morphology of the samples. Meanwhile, the emission intensity of the samples exhibit a green emission centered at about 527 nm while the yellow emission centered at 585 nm and red emission centered at 600 nm resulted from $^4T_1$–$^6A_1$ energy transition of Mn^{2+} ions, respectively.

1. Introduction

In recent years, a number of scholars proposed an interest on synthesizing method and various properties of variety oxide based phosphors [1–3]. Among them, Manganese doped willemite (Zn_2SiO_4:Mn^{2+}) has been distinguished as a competent host matrix for numerous transition metal and rare earth dopant ions for efficient luminescence in the red, yellow and green spectral zones [4,5]. Zn_2SiO_4:Mn^{2+} is a significant material that has been used in Cathode Ray Tubes (CRT), Plasma Displays Panels (PDP) and lamps due to its high saturated color, strong luminescence, long life span, lack of moisture sensitivity and chemical stability [6–8]. Zn_2SiO_4:Mn^{2+} is a polymorphic that exist α, β and other phases where stable α-Zn_2SiO_4 emits green luminescence, while metastable β-Zn_2SiO_4 emits yellow emission under photoluminescence measurements [9–11]. The yellow and green emission come from the same sources, which the energy transition from $^4T_1$-$^6A_1$ of the Mn-doping ions, substituted into the Zn sites of the Zn_2SiO_4 host lattices [12]. It appears to be that β-Zn_2SiO_4 can only occur

* Corresponding author at: Materials Synthesis and Characterization Laboratory, Institute of Advanced Technology, Universiti Putra Malaysia, 43400 UPM Serdang, Selangor, Malaysia.
E-mail addresses: nfarhanasamsudin@gmail.com (N.F. Samsudin), khamirul@upm.edu.my (K.A. Matori), zaidan@upm.edu.my (Z.A. Wahab), josephine@upm.edu.my (J.Y.C. Liew), yapwingfen@gmail.com (Y.W. Fen), sidek@upm.edu.my (S.Hj.A. Aziz), mhmzaid@gmail.com (M.H.M. Zaid).

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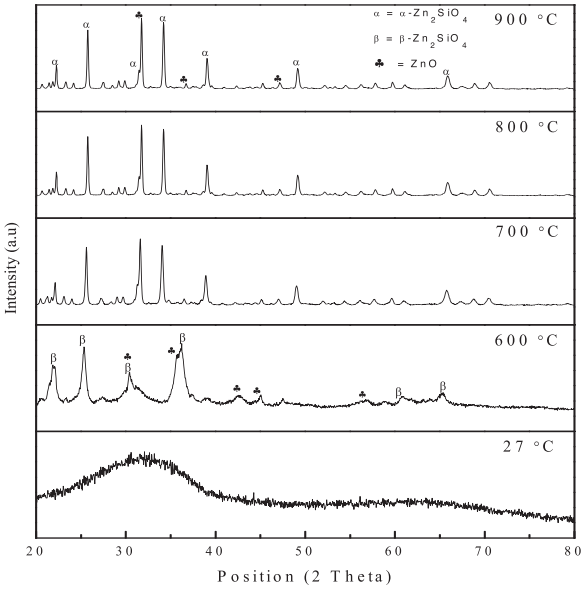

Fig. 1. XRD pattern of 1 wt.% Mn-doped Zn_2SiO_4 sintered at different temperatures.

under certain condition. Previous researcher synthesized Mn-doped β-Zn_2SiO_4 by melting and rapidly cooling mixtures of Zn_2SiO_4 composition from 1500 °C [13]. It is noted that the preparation condition were being responsible for the formation of α-Zn_2SiO_4 and β-Zn_2SiO_4 [14].

Over the time, numerous researchers have reported that Zn_2SiO_4:Mn^{2+} shows strong green emission under an ultraviolet light [15–19]. Surprisingly, very little information is observed on the effects of transition metals doped zinc silicate towards soda lime silica (SLS) waste bottle as a source of silica, SiO_2. Most of them use pure SiO_2 as a starting material in the synthesis process. Instead of using pure SiO_2 as a source of silica, the using of SLS in the production of Zn_2SiO_4 can reduce the cost of production and also has advantages as an attractive host matrix for transition metal ions because of its fine optical and mechanical properties, such as good chemical stability, high transparency, low melting point and high thermal stability [20]. In the present study, Zn_2SiO_4:Mn^{2+} have been prepared from conventional melt-quenching method derived MnO-ZnO-SLS precursor glass by an isothermal heat-treatment process and the effect of sintering temperature towards its structural and photoluminescence properties also been investigated.

2. Experimental

1 wt.% Mn-doped willemite glass ceramics were prepared from a 30 g batch with the starting materials, i.e. ZnO (99.99%, Alfa Aesar), SLS waste glass powder and MnO (99.99%, Alfa Aesar) by using the conventional melt-quenching method. All chemicals were mixed via milling process up to 24 h using the ball milling jar to obtain the homogenous batch. Then, the chemical batch was melted in an alumina crucible at 1400 °C in air with heating rate of 10 °C min^{-1} for 2 h in an electrical furnace. Later, the molten mixture was poured into the water to get the transparent of glass fritz. The glass frits were cooled to room temperature and then was crushed and sieved into fine powder about 45 μm. Next, the fine powders with an addition of 1.75 wt.% Polyvinyl Alcohol (PVA) as the binders, have been pressed at a pressure of 5 tons to form the disk pellets. After that, all the disk pellets were sintered at various sintering temperature in the electrical furnace with duration of 2 h to form the glass-ceramics for further characterization.

X-ray diffraction (PANalytical (Philips) X'Pert Pro PW 3050/60) with Cu Kα radiation was used to confirm the amorphous state of the glass samples and to identify the crystalline phase formed in the sintered samples. The x-ray beam diffracted on the samples through an angle of range of $2\theta = 20^\circ - 80^\circ$ using 0.02 steps. Field Emission Scanning Electron Microscopy (FESEM, FEI, Nova Nano SEM 30) was employed for the microstructural observation on the top surface and cross-section area of the glass and glass-ceramic samples. The photoluminescence excitation and emission spectra were measured by (PL, Perkin Elmer LS 55 Fluorescence) with a 450 W xenon lamp at room temperature at the excitation wavelength of 260 nm, respectively.