Influence of Microstructural Effect on Microvickers Hardness Properties of SiO_2-Na_2O-CaO (SNC) Waste Based Glass-ceramic

IOP Conf. Series: Journal of Physics: Conf. Series **1083** (2018) 012008 doi:10.1088/1742-6596/1083/1/012008

Influence of Microstructural Effect on Microvickers Hardness Properties of SiO_2-Na_2O-CaO (SNC) Waste Based Glass-ceramic

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Abstract. There are a lot of waste materials consist of silicate based such as coal combustion ash, slag from steel production, fly ash, mud, as well as glass cullet or mixtures to produce glass-ceramics. This research work using clam shell (CS) ash and soda-lime-silica (SLS) waste glass powder for fabricating novel SiO_2-Na_2O-CaO (SNC) glass-ceramic. The samples were composed of SLS (50%), Na_2CO_3 (30%), and CS (20%) in weight percentage via conventional melt-quenching technique and solid-state sintering technique. The samples were investigated via X-Ray Diffractometer (XRD), Field emission microscope (FESEM), and microvickers hardness tester. The samples were sintered at 550-950 °C to investigate the influence of microstructural effect on microvickers hardness properties at applied force 0.5 and 1.0 kgf. The optimal Vickers hardness properties at sintering temperature 850 °C due to high crystallization of SiO_2 phase from the residual glass and CaO content enhanced the viscosity flow, high compactness of particles arrangement and densification of sample.

1. Introduction

Glass-ceramics are composite materials consist of combination of residual amorphous phase and crystalline phase. Since the glass-ceramic is used in a large scale application, the materials technology is evolving rapidly. In this study, the mechanical properties of polycrystalline SNC glass-ceramic were determined by phase crystallization and microstructural properties at different sintering temperature. Generally, the mechanical properties of SNC system are less studied by previous researchers. The mechanical behaviour on evolving microstructure of the parent glass with fully crystallized phase have been explained and compared in previous published paper [1-3]. The crystal size plays an important role in preventing the propagation of the cracks in the structure of silicate glass-ceramics with fine

grained microstructure increasing the hardness values [4]. The decrement in glass transition temperature influenced the decrement in microvickers hardness properties of bioactive glass [5]. The microhardness of glass-ceramics generally increased with the crystallization amount, smaller crystalline grains as well as formation of fine microstructure [6]. The mechanical properties of the glass-ceramic are influenced mainly by the evolution changes in microstructure properties [7]. The aim of present research is to investigate the phase composition, crystal system, morphological properties and mechanical properties with the low production cost by using SLS waste glass and CS in the SNC glass-ceramic. All the investigations were carried out to understand the microstructural-mechanical relationships at different applied loads dependence of sintering temperature.

2. Materials and methods

The SNC compositions used SLS (SiO_2), Na_2CO_3, and CS ($CaCO_3$) as the starting materials. The SLS waste glass and CS were plunged and milled by using mill jar for 24 hours to obtain fine powder. The glass-ceramic compositions were composed of 50 wt% SLS, 30 wt% Na_2CO_3, and 20 wt% CS was prepared using the melt-quenching and solid-state sintering. The powders were weighed accurately using an electronic balance and are mixed together by ball milling process at 700 rpm for 24 hours to improve homogeneity of the powders. The batch formulation of SNC composition was melted at 1400 °C. Then, the melted SNC composition was rapidly quenched in water to obtain the glass frits. The SNC glass frits were dried for one day at room temperature. After that, the SNC glass frits were crushed, dry milled for 24 hours and sieved at 45 μm. Finally, the prepared SNC glass-ceramic sample was pressed at 3 tonnes and sintered in air at 550-950 °C for 3 hours. The phases crystallization were investigated through XRD diffractometer (PANalytical X'Pert Pro PW3050/60) with Cu-Kα radiation (λ = 1.54060 Å) in the range of 20° - 80° by a 2θ scan mode and the XRD results were analyzed by using PANalytical X-Pert High ScorePlus Programme. The micrograph of samples that were coated with gold (Au) was observed by Nova Nano 230 Field Emission Scanning Electron Microscope (FESEM). The microvickers hardness tester was used to measure Vickers hardness (HV) values from the symmetrical indentation diagonal length of samples at applied force 0.5 kgf and 1.0 kgf.

3. Results and discussion

Figure 1 shows XRD pattern of the synthesized SNC glass–ceramic samples at different sintering temperatures. The broad peak in XRD pattern corresponds to low crystalline structure. Before sintering, sample was observed to have amorphous phase coexist with some crystalline phases at room temperature. The main phase formed before sintering belongs to $Ca_2Na_2Si_2O_7$ (Ref. code: 98-006-9558) which is in the monoclinic crystal system and SiO_2 phase formed as the secondary peak. Segnit (1953) obtained $Ca_2Na_2Si_2O_7$ compound in the parts of ternary system which is richer in CaO and Na_2O [8]. The compositions of $Ca_2Na_2Si_2O_7$ in SiO_2-Na_2O-CaO system were discovered by Kahlenberg and Hösch (2002) [9]. When sintering temperature at 550 °C, new phases were detected which were belong to Na_2CaSiO_4 and Ca_2SiO_4. The existence of SiO_2 phase from the residual glassy phases together with $Ca_2Na_2Si_2O_7$ phase remains as the temperature increased. When sintering temperature increases at 650–950 °C, the intensity increases and 2θ became broader with the appearance of new $Ca_3Na_6Si_6O_{18}$ (Ref. code: 98-002-1442) crystalline phase. These phase to be a mixture of a solid solution with Na_2CaSiO_4 and Ca_2SiO_4 phases transformed to individual compound, $Ca_3Na_6Si_6O_{18}$ phase which is hexagonal crystal system.

Figure 2 shows unit cell volume (Å^3) of SNC at different sintering temperature. The unit cell volume was calculated manually by multiplication of lattice parameters *a*, *b*, and *c* value. The lattice parameters *a*, *b*, and *c* value were obtained from the XRD programme analysis. The highest unit cell volume indicated un-sintered sample at room temperature. When SNC was sintered from 550-950 °C, unit cell volume increases from 415.161 - 1453.323 Å^3. The lowest unit cell volume was corresponding to sample at 550 °C due to decrement of crystallization phase and low intensity of crystallization peak as observable in Figure 1. The unit cell volume decreased at 850 °C and 950 °C

correlated with increasing intensity peak of $Ca_2Na_2Si_2O_7$ phases which corresponding to high content of CaO and residual glassy phase of SiO_2. High content of CaO and SiO_2 induced high viscosity in the SNC system which leads towards high densification of sample as shown in Figure 3(d) and (e).

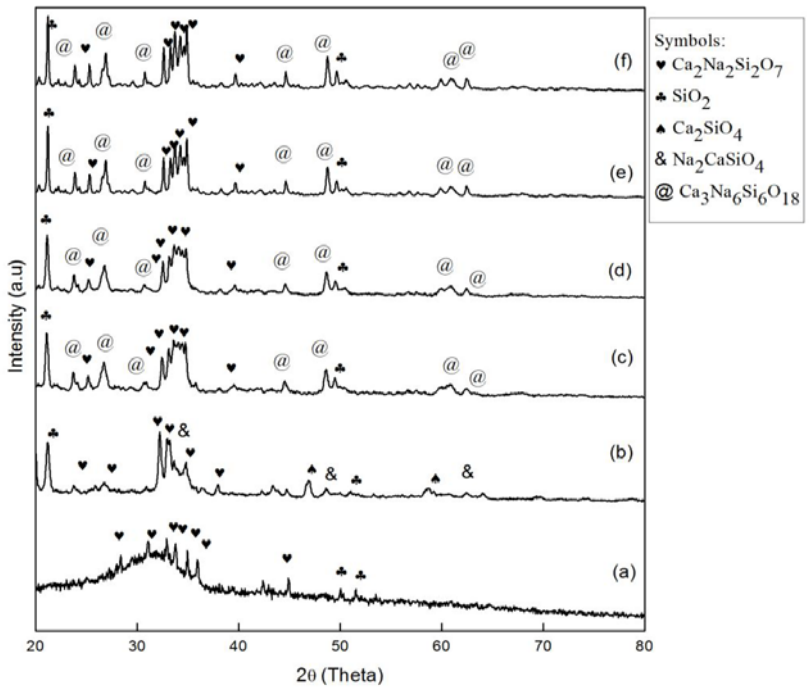

Figure 1. X-ray diffraction pattern of SNC samples (a) before sinter, (b) 550 °C, (c) 650 °C, (d) 750 °C, (e) 850 °C and (f) 950 °C.

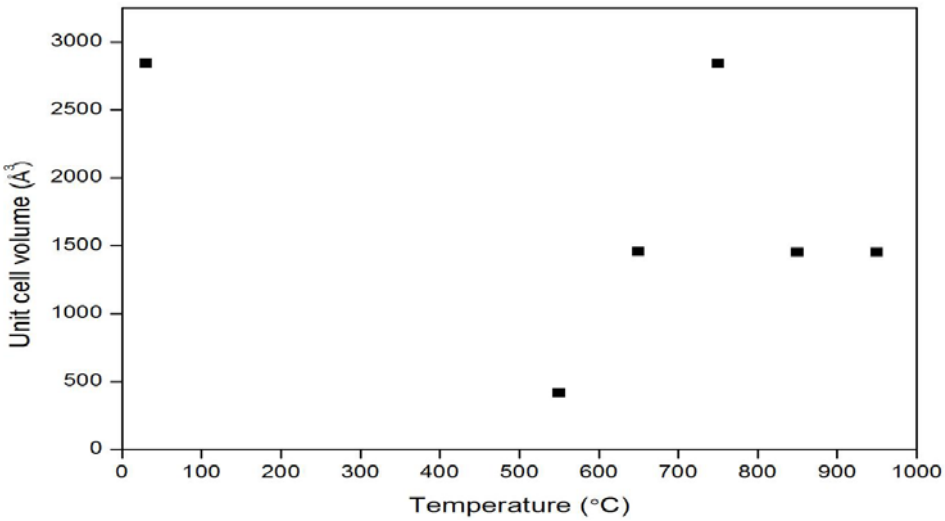

Figure 2. Unit cell volume of SNC versus sintering temperature.

Figure 3 shows the results of SNC FESEM microstructural at different sintering temperatures by using magnification 2000×. Generally, microstructural evolution was caused with increasing the sintering temperatures. The microstructures observed at 550–750 °C showed the increment of grain growth, while the grain boundary and porosity decrease. Samples at 850 °C and 950 °C showed the increment

of grain, while grain boundary decreases induced the grain to combine each other to form agglomerate, less coalescence and porosity decreased. Sintering temperature increases affect the decrement in level of porosity of SNC microstructural. The grain in samples 3 (a) and (b), were appeared with small white spot interstitial remaining glassy phase of SiO_2 which is not fully homogeneous below 750 °C. Meanwhile, in samples 3 (c)-(e) the white spot decreased when it was sintered at 850-950 °C. All SNC samples show the presence of SiO_2 phase from the residual glass as detected by XRD analysis as shown in Figure 1 could enhance the viscous flow and densification of sample at high crystallization temperature. The grain increased as the sintering temperature increased due to the grain growth of major phase crystallization and agglomeration of particles at 850-950 °C. Therefore, SNC produced fine and uniform grain due to high crystallization of SiO_2 and high Na_2O content that act to homogenize the SNC composition as depicted in Figure 3(d) and (e).

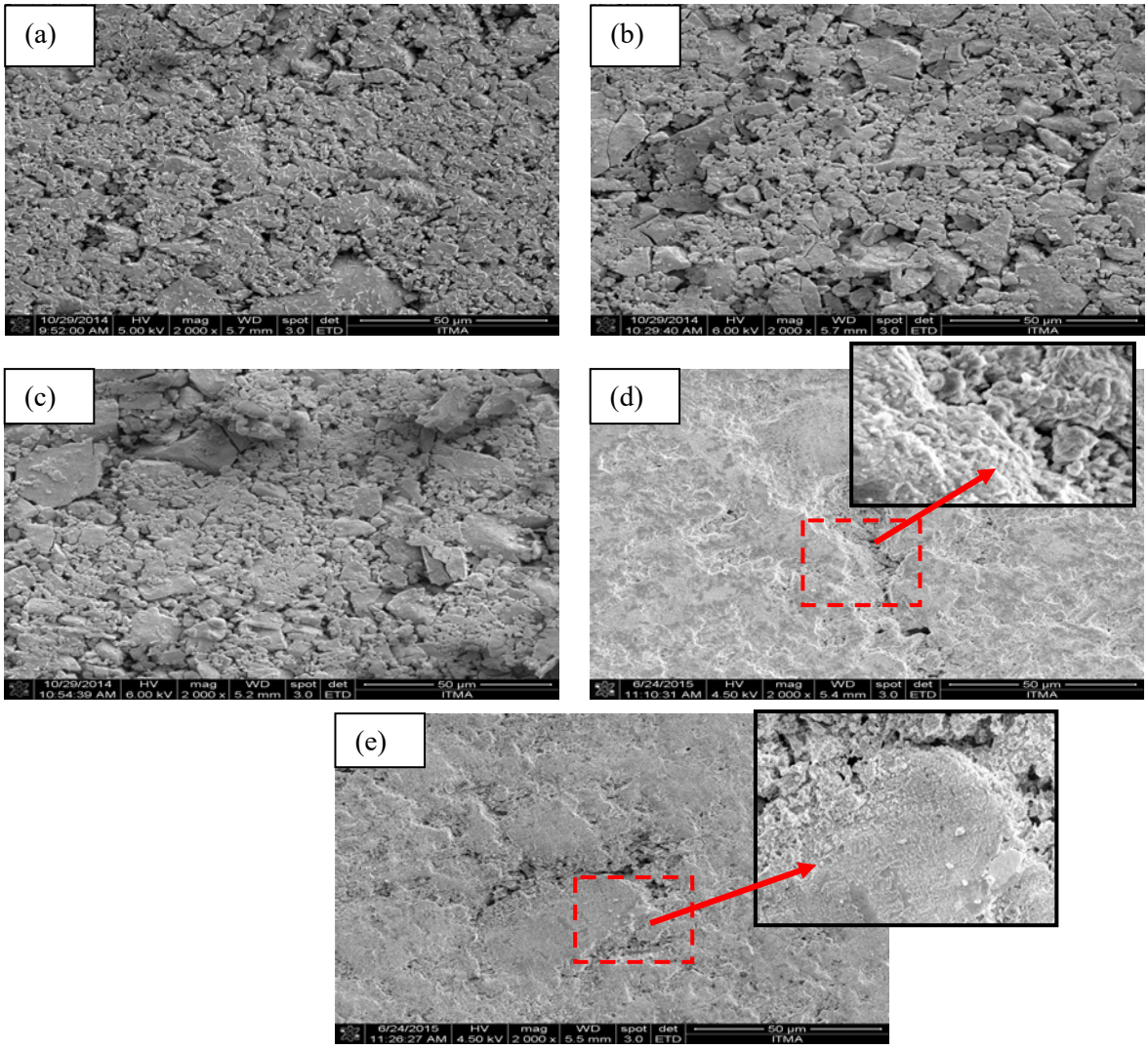

Figure 3. SEM micrographs of SNC at (a) 550 °C, (b) 650 °C, (c) 750 °C, (d) 850 °C, and (e) 950 °C

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Microvickers hardness tester is used to measure hardness and the symmetrical indentation diagonal length of samples [10]. The applied force subjected to loads at 0.5 kgf and 1.0 kgf was chosen to evaluate the sample hardness that can withstand with the microvickers indentation at high and low applied force. Figure 4 demonstrates microvickers hardness (HV) value of SNC at different sintering temperature. It is apparent that both loads resulted in different shapes of Vickers hardness were corresponding to decrement of symmetrical indentation diagonal length on the sample at applied force 1.0 kgf while at 0.5 kgf the symmetrical indentation diagonal length on the sample increased. The HV graph trend at 0.5 kgf showed an increment from 189 HV to 322 HV. On the contrary, when sintering temperature was elevated at 950 °C, the hardness value dramatically decreased to 228 HV. The HV value decreased as the sintering temperature increased at 950 °C possible related with high intensity peak of SiO_2 phase as can be seen in Figure 1. The sample hardness properties at applied force 1.0 kgf obtained the highest value, 277 HV at 550 °C subsequently, drop to 238 HV at 950 °C. Both applied force have the highest HV value at the same sintering temperature, 850 °C. The HV value of SNC samples at 550 – 950 °C from waste-based glass-ceramic not achieved the hardness of SNCP system in the range of 458–600 HV [11-12].

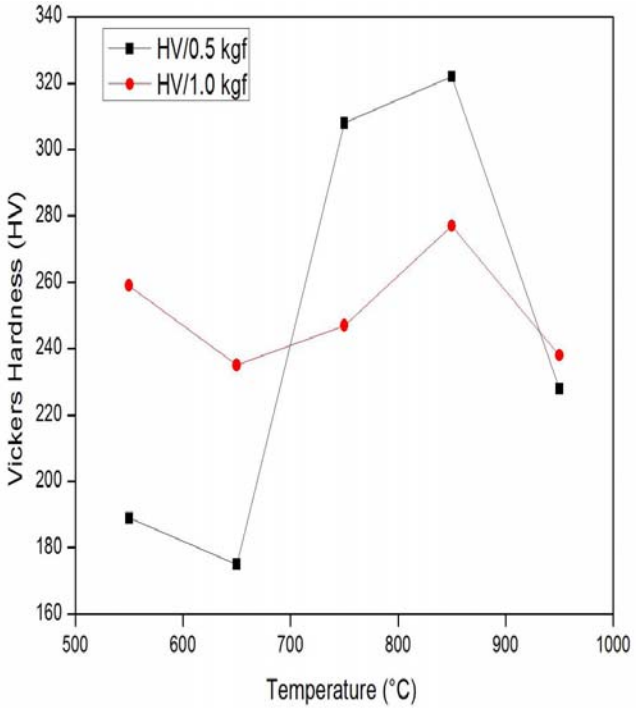

Figure 4. Microvickers hardness of SNC at 0.5 kgf and 1.0 kgf dependence of sintering temperature

4. Conclusion

In this study, the XRD characterization indicated the intensity phase crystallization of $Ca_2Na_2Si_2O_7$, SiO_2, and $Ca_3Na_6Si_6O_{18}$, was increased in correlated with agglomeration of particles and grain growth of major phase crystallization and at 850-950 °. It can be concluded that the SNC microstructural properties at sintering temperature 850 °C had high content of CaO and SiO_2 from the SNC phases crystallization induced high viscosity which leads towards high densification, reduction of porosity, and high compactness of particles arrangement influenced to the highest mechanical microvickers hardness value, 322 HV at applied force 0.5 kgf. The microvickers hardness properties above 950 °C affect to poor mechanical properties which was above high glass crystallization temperature.

Acknowledgements

The authors acknowledge the financial support from Universiti Putra Malaysia and Ministry of Education Malaysia (MOHE) for MyBrain15 sponsorship.

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