GLASS-CERAMIC BASED ON SODA LIME SILICA GLASS SYSTEM

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Glass-ceramics are a group of materials that takes advantage of the various glass-forming methods before they are subsequently heat-treated in a controlled manner to effect nucleation and crystallization to produce crystalline materials. The production of glass-ceramic materials is to overcome the low mechanical strength in pure glassy materials. In this work, a study on the crystallisation of a soda-lime-silica glass was undertaken to ascertain how the processing parameters affect the crystallization of such glasses, viz. either via a single or two-step heat-treatment procedure, as well as the effect of soaking duration at the heat-treatment temperature. A soda-lime-silica glass system was chosen because the raw materials for producing such glasses are readily available and can be considered to be the cheapest. The glass produced was examined by thermal analysis to determine the nucleation and crystallization temperatures before they were heat-treated using a single-step and a two-stage heat-treatment procedures. The resultant glass-ceramics produced were characterized using x-ray diffraction as well as by scanning electron microscopy. The results thus obtained showed that a two-stage heat-treatment procedure is more successful in producing a well-crystallized glass-ceramic product.

Keywords: glass-ceramics, heat-treatment, crystallization, nucleation

INTRODUCTION

It is well known that glasses in practice exhibit low mechanical strength due to surface defects in the form of cracks, pores, or inhomogeneity [1, 2]. This is due to the fact that, once a crack is initiated, the crack will propagate to failure when stress is applied. The strategy in enhancing the strength of glasses include the incorporation of crack will propagate to failure when stress is applied. The strategy in enhancing The strength of glasses include the incorporation of crack barriers. This can be in the form of lamination with a polymeric layer to arrest crack propagation (laminated glass), or by introducing crystalline phases in the glass to act as crack barriers (glass-ceramics) [3,4].

Glass-ceramics are produced by a controlled heat-treatment process, at a slow heating rate of 5°C /min., to effect the crystallization of fine crystals in the glass [1,4]. The crystallization can either be carried to full completion or to various degrees of crystallization, by controlling the duration at the heat-treatment temperature, resulting in a variety of glass-ceramic materials with different properties [2, 3].

A fast heating rate can lead to the instability of the crystalline phases formed or a defective glass-ceramic product [5]. The heat-treatment process can also be carried out using a single- or two-stage heat-treatment protocol [2,3]. Many glass-ceramic systems have been studied [6,7] to produce different glass-ceramics with specific outstanding properties, such as high strength, excellent thermal shock resistance,

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machinability, etc. In this study, a soda-lime-silica glass was chosen based on the easily available raw materials that can be procured at a relatively low cost. Moreover, about 80% of the world's production of glass are based on soda-lime-silica glass and not much literature are found on soda-lime-silica glass-ceramics systems.

MATERIALS AND METHODS

The raw materials used in this study were chemical grade powders (Merck), viz. silica (SiO_2), calcium carbonate ($CaCO_3$) and sodium carbonate (Na_2CO_3). The raw materials were weighed-out based on a soda-lime-silica glass of composition 19:14.5:66.5. After manual mixing in a beaker, the batch (or mixture) was placed in an alumina crucible before being loaded into a bottom-loading glass-melting furnace (Lenton EHF 1700). The furnace was heated up to 1200°C at 10°C/min and soaked for 3 hours before casting the molten glass in a steel mould. As the temperature decreased, the mould with the glass was transferred to an annealing furnace set at 600°C for 2 hours. After cooling, a small piece of the glass was taken and ground in an agate pestle and mortar to a fine powder. The powder was characterized by thermal analysis using a simultaneous DSC/TG (Netsch STA 409). Small pieces were then cut from the cast glass and were heat-treated under various conditions of temperature and duration as shown in Table 1.

The heat-treated specimens were characterized by XRD (Bruker AXS D8 Advance) and SEM (Gemini Supra 55 VP).

Table 1. *Codes and treatment of specimens*

Code	Temperature °C	Duration, hours	Heating Rate, °C/min
A1	770	4	5
A2	770	5	5
B1	770	2	5
	850	2	
B2	770	2	5
	850	3	
C	770	2	5
	1000	2	

RESULTS AND DISCUSSION

(a) XRD Results

The results from x-ray diffraction for the untreated glass and all of the heat-treated glasses are as shown in Table 2. The untreated and A1 glasses do not show any crystalline peaks except for a diffuse halo at a diffraction angle of 30° 2θ which is typical of any amorphous phase. The crystalline phase in all the other heat-treated glass consists of a crystalline sodium calcium silicate ($Na_2Ca_3Si_6O_{16}$) phase. Apart from that, specimen C shows a small presence of a crystalline SiO_2 phase (crystobalite).

Table 2. *Phases upon heat-treatment*

Specimen	Crystalline phase
Untreated	none
A1	none
A2	Na-Ca silicate
B1	Na-Ca silicate
B2	Na-Ca silicate
C	Na-Ca silicate, crystobalite

Specimen A1 do not show any crystalline phase because the heat-treatment schedule is insufficient to effect any nucleation and crystal growth in the glass. In the other specimens, the temperature and duration of heat-treatment are sufficient to cause nucleation and crystal growth [2,3,4]. By raising the temperature to 1000°C, another new crystalline phase based on SiO_2 starts to form.

(b) Morphological observation by SEM

Figure 1 shows the morphology of fracture surface of specimen A1 (single-stage heat-treatment at 770°C for 4 hours). The surface shows a glassy surface fracture with no signs of crystals formed. This confirms the result obtained by XRD (Table 2). Figure 2 shows the fracture surface of specimen A2, which had been heat-treated at 770°C for 5 hours. A small amount of short flaky crystals are observed as the soaking duration was increased from 4 hours (as in A1) to 5 hours. Specimen B1 (Figure 3) shows that the crystalline phase has become more numerous.

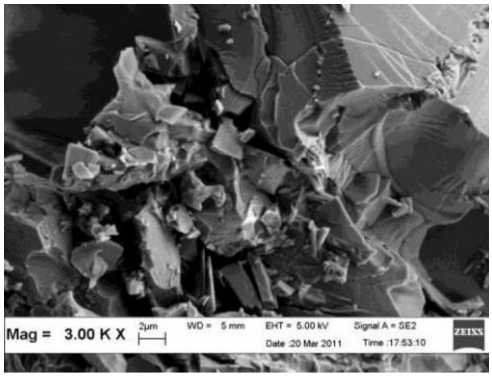

Fig. 1 *SEM micrograph: fracture surface glass ceramic A1 (770°C, 4 hours).*

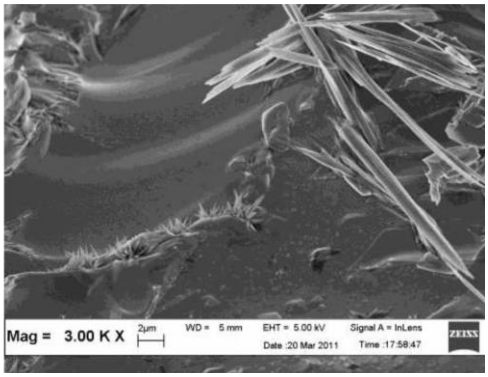

Fig. *2 SEM micrograph of fracture surface A2 glass ceramic (770°C, 5 hours)*

Specimen B1 (Figure 3) shows that the crystalline phase has started to form even at 2 hours soaking duration using a two-stage nucleation and crystal growth heat-treatment Schedule (770°C, 850°C, 2 hours). Subsequently, as the soaking duration was increased to 3 hours, it can be seen (Figure 4) that the crystals have increased in numbers and are distributed much more homogeneously. This is ascribed to the increase in the number of nucleus formed, which later led to crystal growth [8,9]. This proves that a two-stage heat-treatment procedure is much more effective in promoting the nucleation and crystal growth in this glass system. Subsequently, as the growth temperature was increased to 1000°C whilst the duration of soaking still being maintained at 2 hours, it is observed that the crystals have grown into elongated fibres (Figure 5).

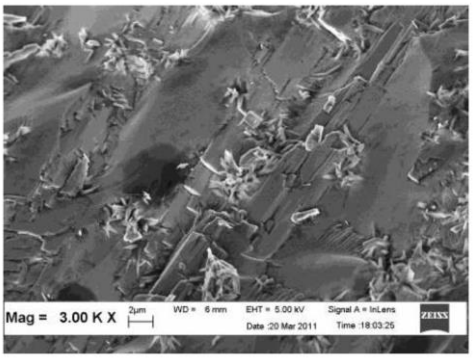

Fig. 3 *SEM micrograph: fracture surface glass ceramic B1 (770°C, 850°C, 2 hours)*

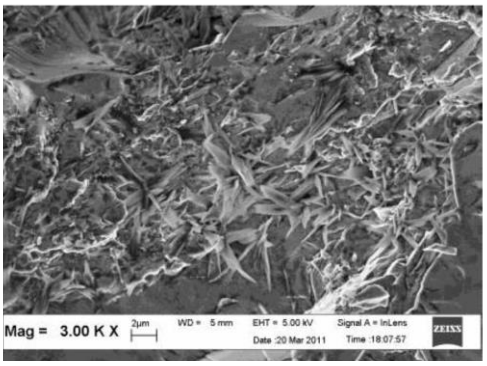

Fig. 4 SEM micrograph: fracture surface glass-ceramic B2 (770°C, 850°C, 3 hours)

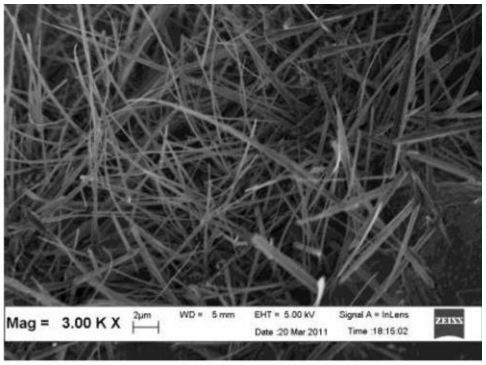

Fig. 5 *SEM micrograph: fracture surface glass ceramic C (770°C, 1000°C, 2 hours)*

CONCLUSIONS

In conclusion, it can be confirmed that soda-lime-silica glass system can be converted into a glass-ceramic with crystals of different morphologies dependent on the heat-treatment temperature and the duration it is soaked at that temperature. This work has also ascertained that a two-stage nucleation and crystallization heat-treatment regime is successful in controlling the amount and size of the crystals being formed.

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REFERENCES

[1] McMillan, P.W. (1964), Glass-Ceramics, Academic Press Inc. (London), 7-156.

[2] Lewis, M.H. (1989), Glasses and Glass-Ceramics, Chapman & Hall Ltd., London, 226-246.

[3] Shelby, J.E. (2005), Introduction to Glass Science and Technology, The Royal Society of Chemistry, London, 1-47.

[4] Strnad, Z. (1986), Glass-Ceramics: Liquid Phase Separation, Nucleation and Crystallization in Glasses, *Elsevier Science Publ. Co.*, 114-120.

[5] Montoya, B.G., Torres-Martinez L.M., Ibarra J. (2003), Alternative batch composition in the glass-forming region of the Na2O-CaO-SiO2 system, *Journal of Non-Crystalline Solids*, 329, 22-26.

[6] Zanotto, E.D. (2010), A bright future for glass-ceramics, *Am. Ceram. Soc. Bull.*, 89,(8) 19-27.

[7] Li, S., Jianfang, W., Zhen, L., Xiaofei, H., Yongsheng, Y. (2015). Crystallization mechanisms and properties of α-cordierite glass–ceramics from K2O–MgO–Al2O3–SiO2 glasses, Journal of Non-Crystalline Solids, 419, 16–26.

[8] Jing, W., Chao, L., Gaoke, Z., Jun, X., Jianjun, H., Xiujian, Z. (2015), Crystallization properties of magnesium aluminosilicate glass-ceramics with and without rare-earth oxides, *Journal of Non-Crystalline Solids*, 419 (2015) 1–5.