Egypt. J. Chem. **Vol. 65,** No. SI 13B pp. 1113 - 1117 (2022)

Egyptian Journal of Chemistry

http://ejchem.journals.ekb.eg/

Producing mosaic glass-ceramic from cullet of soda-lime silica glass

Aya Emad El Din[1] , Hanaa A El Kazazz[1] Rasha Mohamed Ali[1] Esmat M A Hamzawy[2]

[1]Faculty of Applied Art, Glass Dep. , Dokki P.C., Helwan University, Cairo, Egypt

[2]Glass Research Dep. National Research Cennter , Dokki Cairo P.C. 12622 , Cairo, Egypt

Abstract

Mosaic glass-ceramic was prepared from external glass cullet (waste glasses). The collect external glass cullet include float glass , green, blue and brown glass bottles which undergo milling (<0.063 mm) then shaping by uniaxial pressure and finally sintered at different temperatures. However, the powder of float, brown, green and blue glasses were shaped in the form of mosaic pieces and sintered at 650, 700 and 750 ˚C. The developed phases were low quartz and wollastonite. The microstructure shows submicron and nano size particles spread in glassy matrix. The densities and hardness of the sintered glass-ceramic samples were between 2.22 and 2.45 g/cm^3 and 650 kg/mm^2 respectively. Also the coefficient of thermal expansion CTE of the sintered sample was 50 x 10^{-7} ˚C^{-1} (25-500 ˚C) and is better than the parent bulk glass. The chemical resistance of the sintered glass powder in water and acidic medium were better than in the parent bulk glass. The final products have good properties and can used in decorative and ornamental purposes.

Keywords: glass cullet, sintered glass-ceramic, mosaic.

1. Introduction

Glass cullet is a glass wastes, especially soda lime glass, is produced from containers or flat glass. This glass was produced after the gathering of raw materials and energy, and it must be re-used. Therefore it is necessary to find a suitable technical way to take advantage of these glass wastes by recycling them. In consequence, the glass wastes were crushed and sifted in suitable grain size where this powder was used in the production of mosaics of ceramic glass that can be used in covering the walls and decorating too.

Recycling of glass wastes (external glass cullet), other than reusing it in the glass industry, many authors are tested it in different materials. It was easy to put the grind glass cullet (in micro-scale size) and use it in the production of concretes [1-4] . The later glass cullet was also used in autoclaved materials (silica sand –limestone) . Glass-ceramic containing wollastonite, diopside and orthoclase phases was produced from cullet of cathode ray tube (CRTs , containing high ratio of SrO and BaO) with the additive of iron slag, flyash and Calicglass (containing high ratio of CaO) [5] . The high heat-treatment (1050 ˚C) of a well-mixed mixture of oil shale ash with glass cullet gave a ceramic materials with low water absorption and high bending strength [6] . Increase the paste strength in the concert mixture with the addition of glass cullet (composed of glass cullet ,cement, and quick lime) since the compression strength increase with time [7]. In the countries that lack natural silica sand, glass cullet in sand size can be used in concert instead of silica sand [8] .

Applications of glass cullet in decorative materials are rare. In the un-referenced work, polymer or wax have un-coloured / coloured glass pieces were produced in different decorative articles such as vasa candle kits, or also in the swimming pool. However, self-re-use of the glass cullet a mosaic for decorative purposes was rare. The present research deals with the re-use of the glass cullet in ornamental mosaic materials. For the later purpose, characterization of the glass and the corresponding sintered glass-ceramic was considered using x- ray analysis and scanning electron microscopy and some properties were measured such as density, hardness, coefficient of thermal expansion, and chemical durability.

2. Experimental and Methods

Glass cullet of different coloured (white , green, brown and blue) was ground using grinding with quartz balls (Agate - Retsch - Germany) to obtain a homogeneous powder in micron size (< 0,063 mm) . Different steel moulds are used to shape the glass powders. The glass powder was mixed with little binder solution (polyvinyl alcohol -7%) then shaped in a mould through uniaxial compacted (Paul Weber – Germany) at 20 KN .

The heat-treatment regime was between 200 ˚C and 750 ˚C with 10 ˚C/min heating rate. The developed phases were identified using x-ray diffraction

*Corresponding author e-mail: ehamzawy9@gmail.com.;
Receive Date: 11 August 2022, Revise Date: 02 September 2022, Accept Date: 18 September 2022
DOI: 10.21608/EJCHEM.2022.155701.6735

analysis ((XRD-model-Bruker AXS D8 advance using Cu Kα - radiation) whereas the microstructure was examined using scanning electron microscopy attached with x-ray microanalysis (FE-SEM, Model FEI, QUANTA, FEG, 250, Holland). For SEM analysis, the fresh fracture glass-ceramic surface was etched chemically (using 1% HF+1%HNO_3 for 30 seconds) and then washed with distilled water. The sintered glass-ceramic sample must be coated with carbon film before subjecting to SEM analysis.

Density was measured by the Archimedes (g/cm^3) method and the microhardness was determined using a Shimadzu microhardness tester (Kg/mm^2, type –M, 100 g with time 15 second) . The coefficient of thermal expansion CTE (α x 10^{-7} $˚C^{-1}$) was measured using a rod (with dimension 2 cm x 2 mm x2 mm) using a dilatometer (CTE- NETZSCH DIL402 PC, Germany). The chemical resistance of the sintered glass-ceramic was done by calculating the surface area of the samples after soaking the sample in water and acidic medium (1M) at 90 ±2 for 1h) . The weight loss % in the sintered sample was measured after washing and drying. For accuracy, two samples were measured in chemical resistance.

3. **Results and discussion**

A. Characterization of the samples

The sintering process between 650 ˚C to 750 ˚C produces a different nature of the samples from opaque to vitreous state respectively. However, the best mosaic glass-ceramic were formed at 750 ˚C /2h (Fig 1). Figure 1 shows the developed mosaic glass-ceramic samples with mix between the glass powders in ornamental shape. The following characterization and properties tests were done on the float glass which represents the major constituent.

The crystalline phases formed after sintering the glass samples at temperatures 650, 700 and 750 ° C (Fig. 2) show the formation of quartz (ICDD Card No. 96-900-0776. SiO_2) and a little wollastonite (ICDD Card No. 96-900-5778. $CaSiO_3$). The presence of a large proportion of silica (<70%), as it is the main component of the irregular (amorphous) composition helps the crystallization of the low quartz. Also, wollastonite crystallizes in the presence of calcium (8-13%) and silica helps its formation. The energy of formation both quartz and wollastonite in the current glass system (Na_2O-CaO-SiO_2) increases with increasing temperature and also increases the surfaces of glass grains.

This SEM micrographs of the crystalline microstructures formed at 650, 700 and 750 ˚C temperatures show different textures. At low magnification, some parallel lines reflect the shape of the fracture at 650 ˚C. Some scattered crystals and some holes also appear in the glass, which represents a large percentage of non-crystallization. At 700 ˚C, the crystals appear granular (and their proportion is more than the sample that was treated in 650 ˚C. Hexagonal crystals appear, which refers to quartz. At 750 ˚C, scattered crystals appear and are larger in size, as well as some larger holes than in the previous temperatures. However, the crystals at a temperature of 700 ˚C were between 200 and 500 nanometers and the crystals formed at a temperature of 750 C were between 200 and greater than 1000 nanometers (Fig. 2) .

The EDX microanalysis of the sintered samples 650, 700 and 750 ˚C are shown in **figure 3**. The microanalysis shows more or less the components of the glass, i.e. soda –lime-silica glass. Although there are few differences in the proportions and also in the presence of some elements with small proportions in the glass, which either do not appear in the analysis or may be Interfering (wavelength) with other elements. Table 2 shows the chemical compositions of the commercial glass , in comparison with the corresponding sintered glass at temperatures 650, 700 and 750 ˚C.

B. Some properties of the sintered glass samples at 750 ˚C.

The values of densities, hardness, coefficient of thermal expansion and chemical resistance for the bulk glass and corresponding sintered glasses at 750 C are listed in Table 1 . The density of the bulk glass and sintered glass were 2.530 and 2.456 respectively since the decrease of density in glass-ceramic is related to the residual pores after the sintering process. However, the recorded density of soda-lime silica glass was 2.550 g/cm^3 whereas the density of low quartz was 2.650 g/cm^3 whereas wollastonite density was 2.900 cm^3 [9-12].

The microhardness (HV) values of the bulk glass and the corresponding sintered glasses were 537 and 650 kg/mm^2 respectively. The HV recorded values of low quartz and wollastonite glass-ceramic were 635 and 544 Kg/mm^2 [13-14] whereas the hardness of soda-lime-silica glass was 540 Kg/mm^2 [15] . The hardness results of the sintered glass-ceramic is higher than the bulk glass-ceramic.

The CTE of bulk glass was between 102 and 120 x 10^{-7} $˚C^{-1}$ while it was 50 (35-300 ˚C) or 76 (35-500 ˚C) x 10^{-7} $˚C^{-1}$ for the sintered glass-ceramic. The recorded CTE of wollastonite glass-ceramic is 76.1 x 10^{-6} / K (30−380 °C) [16]. For low quartz (hexagonal) , the CTE is different according to the axis position since it is low at a - axis and high along c-axis from negative value to > 100 x 10^{-6} / K [17 and 18] .Therefore the CTE was low in the sintered samples than the bulk glass.

The chemical resistance in water and HCl (1M) was better in sintered glass-ceramic than in bulk glass. Crystallization of both low quartz and wollastonite resist the attack of the solutions than in amorphous bulk glass (Table 1) .

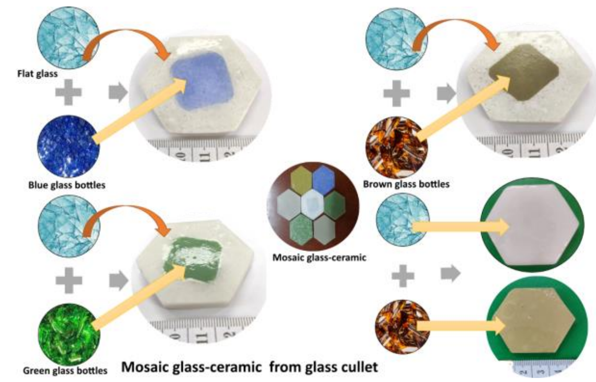


Fig 1. Mosaic glass-ceramic samples from external glass cullet .

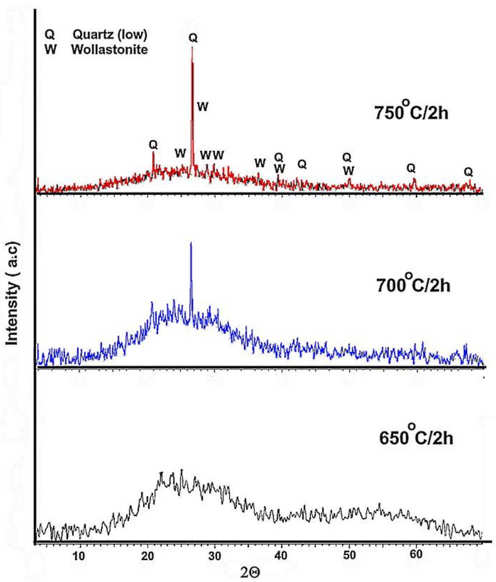
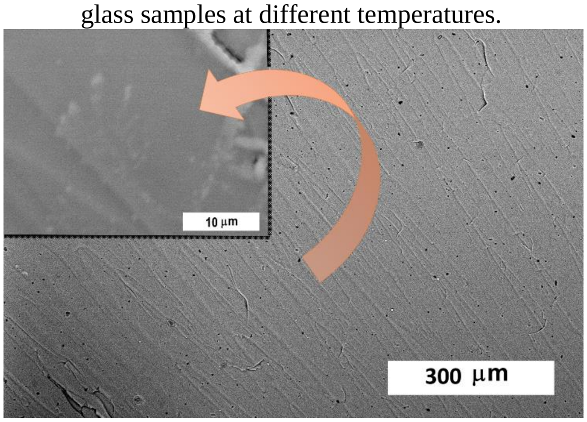


Fig . 2. X-ray diffraction analysis of the sintered float glass samples at different temperatures.



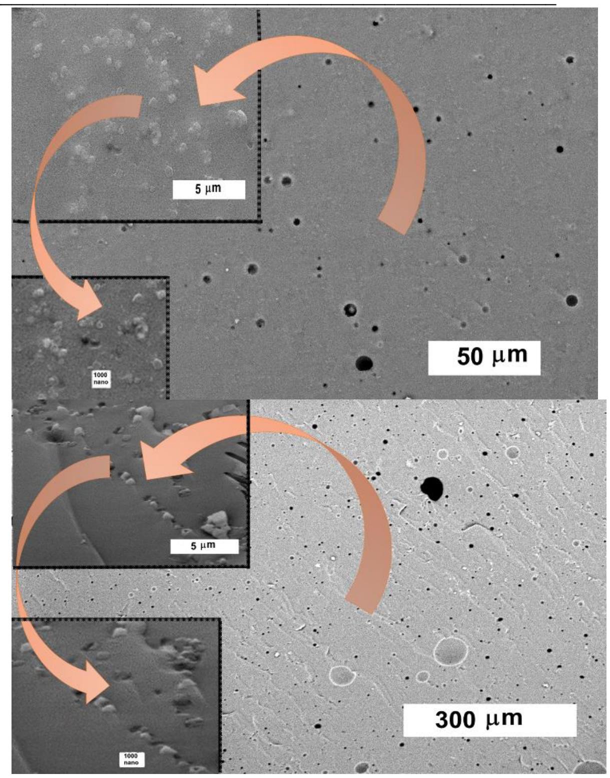


Fig 3. SEM micrographs of the float glass samples sintered at : A.650 ˚C, B.700 and ˚C.750 ˚C .

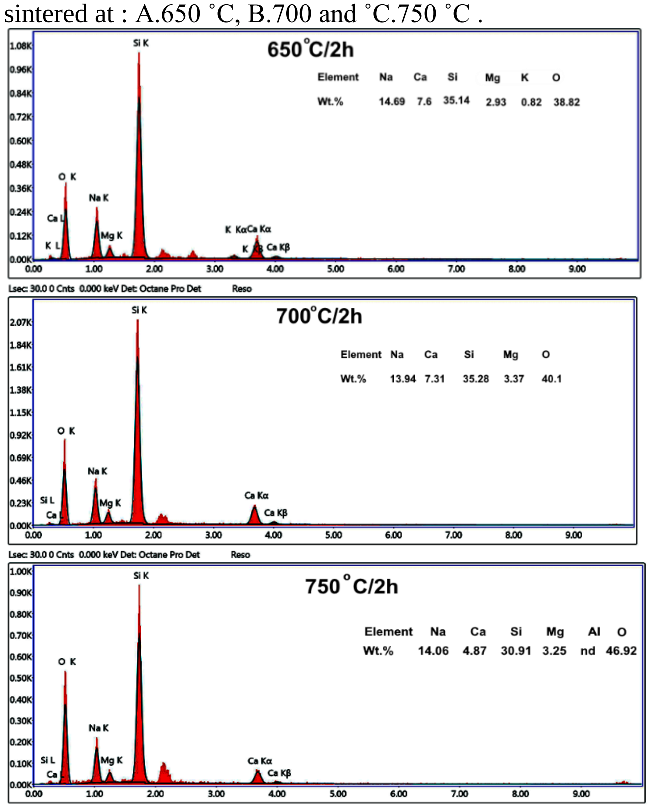


Fig 4. EDX microanalysis of the float glass samples sintered at : 650 ˚C, 700 and 750 ˚C

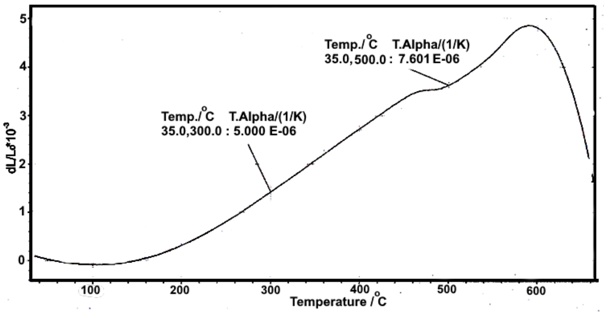


Fig. 5. CTE of sintered glass-ceramic sample

Table 1. Results of densities, hardness, thermal expansion coefficients and chemical resistance of the sintered float glass sample at 750 ˚C.

Property (unit) Sample	Results
Density, ρ (g/cm^3)	
Bulk glass	2.530
Sintered glass powder	2.456
Hardness, V'H (kg/mm^2)	
Bulk glass	537
Sintered glass powder	650
Thermal Properties	
Glass	
Transformation Temperature Tg°C	560
Softening point Ts°C	591
Coefficient of thermal expansion CTE = α x 10^{-7} ˚C^{-1}	
(bulk glass)	
20-300 ˚C	125.30
20-500 ˚C	102.00
Glass-ceramic	
Coefficient of thermal expansion CTE= α x 10^{-7} ˚C^{-1}	
(sintered glass powders)	
35-300 ˚C	50.00
35-500 ˚C	76.01
Chemical resistance (at 95 ˚C/1h) wt. loss ratio g/cm^2	
Distilled water	
bulk glass	0.001
Glass-ceramic (sintered)	0.0001
HCl (1M)	
bulk glass	0.00032
Glass-ceramic (sintered)	0.0001

Conclusion

Mosaic glass ceramics were prepared from different glass cullet including float glass , exhausted green, brown and blue bottles. The cullet undergoes pulverizing shaping then sintering process to get the mosaic product. The studied sintered float glass cullet shows the crystallization of low quartz and wollastonite which appear as scattered crystals , granular- or hexagonal-shape , in submicron and nano-scale size. The density and hardness of the sintered sample was 2.456 g/cm^3 and 650 Kg/mm^2. The coefficient of thermal expansion of the sintered mosaic sample was 50.00 and 76.01 x 10^{-7} ˚C^{-1} (20-300 and 35- 500 ˚C range respectively) . The solution resistance of the sinterd (either in water or HCl) is better than original glass cullet). The present mosaic glass-ceramic has suitable properties for using as ornamental wall cladding.

Conflicts of interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

References

1. Stepien A. , Potrzeszcz-Sut B., Kostrzewa P., Influence and Application of Glass Cullet in Autoclaved Materials , *Materials Science and Engineering* 471 , 032065 (2019).
2. Aliabdo A. A., Abd Elmoaty A. E. M., Aboshama A. Y. : Utilization of waste glass powder in the production of cement and concrete, *Construction and Building Materials* 124 (15) 866-877(2016).
3. Sadiqul Islam G. M. Waste glass powder as partial replacement of cement for sustainable concrete practice, *International Journal of Sustainable Built Environment* 6, 37–44(2017)..
4. Harbec,. Zidol A, Tagnit-Hamou A., Gitzhofer F., Mechanical and durability properties of high performance glass fume concrete and mortars, *Construction and Building Materials* 134, 142-156 (2017).
5. Reben M., Wasylak J., Kosmal M., Glass-ceramics from kinescope glass cullet. Processing, Properties, and Applications of Glass and Optical Materials: Ceramic Transactions Edited by Arun K. Varshneya, Helmut A. Schaeffer, Kathleen A. Richardson, Marlene Wightman and L. David Pye Copyright © 2012 *The American Ceramic Society*, p 151 - 159(2012) .
6. ALJBOUR S. H. : Production of ceramics from waste glass and jordanian oil shale ash. *Oil Shale*, 33 (3) 260–271(2016).
7. Deja J. and Gołek Ł. , Application of glass cullet in binder production . Article in Cement, Wapno, Beton ·. This work was supported by Polish Ministry of Sthe cience and Higher Education (project No. 11.11.160.938 November (2011).
8. Mavroulidou, M. and Ahmed M.U. : Geotechnical properties of glass cullet . *Proceedings of the 12th International Conference on Environmental Science and*

Technology , *Rhodes*, *Greece*, 4-8 September (2011).

9. Andrew, A., Densities of Common Rocks and Minerals., Thought Co, Feb. 16, , thought co.com/densities-of-common-rocks-and-minerals-1439119, (2021).
10. K. Lin, J. Chang, Y. Zeng, W. Qian : Preparation of macroporous calcium silicate ceramics. Mater Lett, 58 (2109–211) (2004).
11. A.B.Y. Hazar Preparation and in vitro bioactivity of CaSiO3 powders Ceram Int, 33 (687–69) (2007), p. 2
12. Karamanov A., Pelino M. , Induced crystallization porosity and properties of sintereds diopside and wollastonite glass-ceramics, J *Eur Ceram Soc*, 28 (3) , 555-562 (2008).
13. Teixeira S. R., Souza A. E., Carvalho C. L., Reynoso V.C.S., Romero M., Rincón J. Ma., Characterization of a wollastonite glass-ceramic material prepared using sugar cane bagasse ash (SCBA) as one of the raw materials. *Materials Characterization* 98, 209–214(2014).
14. Ainsworth L. The diamond pyramid hardness of glass in relationto the strength and structure of glass. Part II. Silicate glasses. *J SocGlass Technol* 38 : 501-35(1954).
15. Aktasa B., Albaskara M., Dogru K. and Yalcin S., Mechanical Properties of Soda-Lime-Silica Glasses Doped with Eggshell Powder , *Acta Physica Polonica Series a*, 132(3), (2017) 436-438 .
16. Holand, W., Beall, G.H.: Glass-ceramic technology. 2nd ed., Johan Wiley & Sons, Hoboken, New Jersey (2012), ISBN: 978-0-470-48787-7
17. Kosinski J. A., Gualtieri J. G., and Ballato A., Thermoelastic Coefficients of Alpha Quartz . IEEE TransactLons on Ultrasonics, Ferroele(Trics, and Frequency Control, 39, 4, (1992)
18. John P. Sutter, and Hasan Yava , Material properties of α-quartz that are relevant for its potential use in X-ray monochromators and analyzers . Physics Instrumentation and Detectors arXiv:, 1612.07049 (2017).