International Journal of Nanoelectronics and Materials
IJNeaM
ISSN 1985-5761 | E-ISSN 2232-1535

Processing of Porous Glass Ceramic using Silica Sand and Industrial Waste for Tiling Application

Mohd Hakim Ibrahim[a,*], Mohd Idham Mustaffar[a], Anis Nadhirah Ismail[b], Syarifah Aminah Ismail[a] and Nabihah Othman[a]

[a]*Mineral Research Centre, Department of Mineral and Geoscience Malaysia, Jalan Sultan Azlan Shah, 31400 Ipoh, Perak, Malaysia*
[b]*Faculty of Chemical Engineering Technology, Universiti Malaysia Perlis (UniMAP), Kompleks Pusat Pengajian Jejawi 3, Kawasan Perindustrian Jejawi,02600 Arau, Perlis, Malaysia*
**Corresponding author. Tel.: +605-5477051; fax: +605-5477185; e-mail: mohdhakim@jmg.gov.my*

ABSTRACT

The main goal of this research is to produce a porous glass ceramic from natural silica sources and industry waste to be used as tiling in building applications. Basically, silica sand is the raw material of natural silica while soda lime glass is the raw material of industrial silica. Both materials were mixed with carbonate foaming agent and clay binder. Then, the mixed samples were pressed into button shape. The button-shape samples were sintered at sintering temperatures of 800, 850, 900 and 950°C with natural cooling at room temperature for 24 hours. It was found that the highest bulk density and modulus of rupture (MOR) were obtained at 800°C with 1.91 g/cm^3 and 21.97 MPa respectively, while the highest water absorption was obtained at 950°C with 16.37%. The surface morphology analysis showed that, as the temperature increased, the pores became larger and some of them merged and bonded together. However, the optimum sintering temperature was achieved at temperature 900°C recording to its lower density (1.75 g/cm^3) and high MOR (19.64 MPa) that met the ISO 13006 standard's minimum requirement of higher than 15 MPa.

Keywords: *Glass ceramic, Porous, Silica sand, Soda lime glass, Sinter*

1. INTRODUCTION

Waste from production and industrial operations are currently a major concern that has evolved into serious issues as the world's population and living standards have increased. Furthermore, garbage generation has become a source of concern in recent years, as the increasing volume of waste produced has put a strain on the nation's landfills [1]. Because of the harmful hazards, the solid waste generated could harm the environment and persons. Recycling material is crucial in a country of roughly 200 million people with increasing urbanization, industrialization, and living standards, so that resources are continuously gathered and managed for sustainable development [2].

According to Hobson et. al. to minimise waste, it is crucial to concentrate on a six-pronged model that includes reducing, reusing, repairing, remanufacturing, recycling, and recovering resources efficiently. This will help minimise the extraction of resources from nature and extend their longevity [3]. Recycling is a method that can be utilised to reduce pollution in the air and water, as well as emissions of greenhouse gases that contribute to the acceleration of climate change. Recycling also contributes to the conservation of energy and natural resources [4].

Glass garbage is one of the most plentiful types of waste on the planet, and the best part is that it is easily recyclable. There are several methods for recycling glass waste. For instance, container glass can be recycled endlessly to make new containers while maintaining its quality or purity because it is 100% recyclable. A variety of applications can be found for waste glass, including its use as a fluxing agent in the production of ceramics and bricks, as an aggregate in construction concrete, as an abrasive substance, and as a medium for water filtering [5].

Numerous studies have been conducted over the last 20 years using industrial and glass waste to produce glass ceramics, among others, for construction purposes. Due to its simplicity and cost-effectiveness, many researchers believe that utilising the sintering crystallisation process for manufacturing glass ceramics is an efficient way to recycle and repurpose various types of industrial waste.

According to the sintering method, the densification and crystallization processes can be completed quickly and at relatively low temperatures by optimizing the processing parameters and reducing the size of the powder particles, which will reduce the cost of waste treatment. Apart from the advantages in manufacturing, the sintering process can result in the production of unique crystal phases that yield glass ceramics with exceptional mechanical properties.

Glass ceramic offers a wide range of properties, with crystalline and non-crystalline materials having similar qualities. Porous glass ceramics have been popular in recent years due to their porous structure's ability to make lightweight items. Solid material with void, pores, channels, and interstices can be called porous according to Mejia, 2015 [6]. Porous glass ceramics are heterophase porous materials with small, homogenous pores that are loaded up between them.

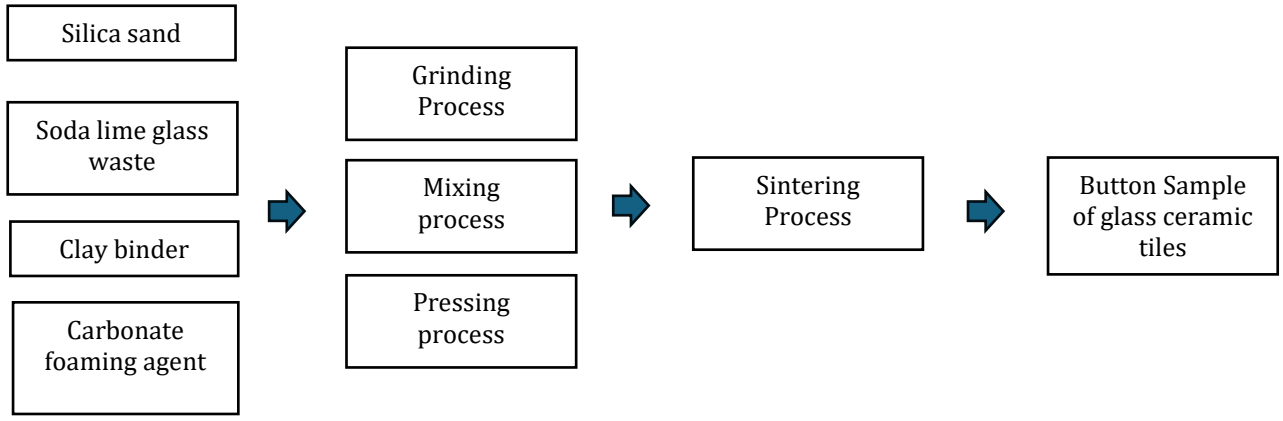

Figure 1. Procedural sequence for the preparation of glass ceramic tiles sample.

The glass ceramic body's pores were formed by gases released during the foaming agent's reaction at high temperature. Because of the foaming agent reaction, countless small gas bubbles formed within the glass ceramic, resulting in a highly porous structure. The existence of these pores improves the thermal insulation qualities of the material, making it appropriate for applications such as insulation panels or lightweight construction materials [7]. Trapped gas (gas phase) causes the formation of pores, while crystallized glass (solid phase) creates the thin wall of a single unit cell. Additionally, the benefits of porous glass ceramic are its lightweight and ability to keep the required mechanical strength and physical shape [8].

Porous glass ceramics were created by experimenting with various sintering temperatures in combination with industrial waste (soda lime glass) and natural silica sources (silica sand). This work's goal is to determine whether porous glass ceramics, or tiles, can be made using

industrial waste (soda lime glass) and natural silica (silica sand) using the traditional solid-state powder sintering process. The materials' formulation was created to decrease the quantity of silica sand while preserving the calibre of the final product. Because silica sand comprises multiple crystalline phases, such as quartz, it is thermally stable and needs to be destabilised at temperatures greater than 1500 °C.

As a result, more energy is required, which is unfavourable from an economic perspective. To reduce the processing temperature, silicate-rich soda lime glass was added to the formulation to generate a glassy phase that supported the entire structure, reducing energy consumption during the sintering process. As a foaming agent, carbonate foaming agent was applied to stimulate the porosity structure of the sintered body. Thus, the purposes of this research are to produce porous glass ceramics from silica sand and soda lime glass waste by the sintering crystallization method and investigate the effect of sintering temperature on the properties of physical, mechanical, and morphology behaviour of the glass ceramics.

2. MATERIALS AND METHODS

2.1. Raw Materials

Silica sand, soda lime glass, clay binder, and carbonate foaming agent are the raw materials used to make glass ceramics. Silica sand that was used as filler and soda lime glass as fluxing agent were supplied by the Terengganu Silica Consortium (TSC) at Setiu, Terengganu, Malaysia, and MCIS Safety Glass Sdn. Bhd., Senawang, Negeri Sembilan, Malaysia respectively. The purpose of the fluxing agent is to expedite the sintering process by generating a glassy phase [9]. Other elements used to make porous glass ceramics include clay binder, which acts as a binder or filler, and carbonate, which acts as a foaming agent to create a pore structure. Clay binder is a natural ingredient that aids in the binding of all materials in a mixture, giving the finished product strength and stability. Carbonate, on the other hand, works as a foaming agent, causing tiny air bubbles to form inside the mixture, reducing density and making it lighter.

2.2. Sample Preparation

The laboratory scale was employed to fabricate samples of porous glass ceramic in this study. Figure 1 illustrates the procedural sequence for the preparation of glass ceramic tiles. Initially, the primary constituents, silica sand and soda lime glass, underwent a thorough cleansing process to eliminate any contaminants. Subsequently, the two primary substances were pulverized and reduced to a particle size of -75 μm utilizing a mortar grinder. The primary components utilized, namely silica sand (40-50 wt%), soda lime glass waste (20-30 wt%), carbonate foaming agent (1.5 wt%), and clay binder (20 wt%), were mixed in a mechanical rotary mixer for a duration of five hours to produce LGCP30 samples.

The dry mixing process ensures that the materials are evenly distributed and thoroughly combined. This step is crucial in achieving a homogenous mixture, which is essential for the subsequent stages of the manufacturing process. Once the five-hour mixing period is complete, the resulting mixture is ready for further processing. A uniaxial hydraulic press with a pressure of 20 tons was used to

create porous glass ceramic (LGCP30) samples in the shape of buttons that were 35.5 mm in diameter and 10 mm thick.

In this laboratory scale experiment, the LGCP30 button samples mitially sintered at temperatures ranging from 30 to 500°C for 30 minutes soaking time at 10°C/min heating rate to remove residual water, moisture, and organic matter. The process was repeated at different sintering temperatures (800, 850, 900, and 950°C) with a heating rate of 5 °C/min and then 30 minutes soaking time achieved. The sintered samples were finally slowly cooled for 24 hours at room temperature.

2.3. Sample Characterization

The bulk density, water absorption, apparent porosity, linear shrinkage, and modulus of rupture (MOR) tests were applied to the LGCP30 sintered sample. In accordance with ASTM C373-88 [10], the bulk density and water absorption of the specimens were ascertained utilizing the Archimedes principle. The modulus of rupture (MOR) was ascertained through a three-point bending test conducted under compression on a universal testing machine (Instron 3367). The specimens utilized for this objective measured approximately 30 mm in length and 10 mm in width. A minimum of five distinct samples are utilized in order to verify the mean value of each sample.

3. RESULTS AND DISCUSSION

3.1. Chemical Composition of Raw Materials

The chemical composition of silica sand, soda lime glass, clay binder, and carbonate foaming agent was analyzed using X-ray fluorescence spectrometer (XRF). The results, displayed in Table 1, indicate that all elements had been measured in the form of oxides. From Table 1, the silica sands primarily consist of SiO_2, with a weight percentage of 97.30%. The main constituents of soda lime glass waste are SiO_2, CaO, and Na_2O, with weight percentages of 74.87, 8.70, and 4.84 wt%, respectively. The main constituents of the clay binder utilised in this investigation are SiO_2, Al_2O, and Fe_2O_3, with weight percentages of 40.31, 5.56, and 3.84 wt%, respectively. With a weight percentage of 70.13, calcium oxide (CaO) is the only major oxide element present in carbonate foaming agents.

Comparatively, the SiO_2 content in silica sand is higher than clay binder and soda lime glass waste. The high content of SiO_2 in silica sand indicated the purity of silica sand and the presence of the quartz phase, which is a characteristic of silicate systems [11]. However, the carbonate foaming agent has a lower SiO_2 (0.33 wt%) concentration and a higher CaO concentration when compared to other materials. As can be seen from the result, the loss on ignition (LOI) for clay binder and carbonate foaming agent is discovered to be higher than silica sand and soda lime glass waste, respectively. The LOI of clay binder is measured at 45.95 wt%, and that of carbonate foaming agent is measured at 28.64 wt%. The reason for the higher LOI is because clay binder loses about half of the combination of crystal water, organic carbon, and sulphur. While for the carbonate foaming agent, the carbonates already decompose and release CO_2 gas after firing at 850°C [12].

3.2. Effect of Sintering Temperature

The condition of the LGCP30 button samples after sintering at different temperatures (800, 850, 900, and 950°C) as depicted in Figure 2. It is plainly visible that the shape of the button specimens does not undergo a significant change as a result of the temperature being raised. The reason behind this is that the button shape was preserved while the driving force of densification was increased by utilising an appropriate sintering temperature that was lower than 950°C [13]. Overall, pore sizes were small and uniform across the board, regardless of sintering temperature. The majority of the pores were connected to one another in a connection. In addition, the pores were rounded in shape and distributed in a uniform manner throughout the material.

Figure 3 showed the MOR, bulk density, and water absorption result at different sintering temperatures. It was found that the highest value bulk density (1.91 g/cm^3) and MOR (21.97 MPa) was achieved at 800°C. However, the water absorption at 800°C is the most minimal (11.43%) compared to other temperatures. In this regard, the glassy phase's viscosity was relatively high, preventing pores from expanding due to gas pressure inside the body. Furthermore, because the glassy matrix was not soft enough to trap the released gas, the majority of it escaped through small and uniform pores [14]. As the temperature increased, the pores grew larger, and some of them coalesced and interconnected. The reduced viscosity of the glassy phase causes an increase in gas pressure within the softening body as carbonate oxidation expands [14].

More constricted pores result from the decrease in viscosity, which also encourages densification and increases the difficulty of gas escape. But when the pressure increases above a certain threshold, the gas leaks out through the pore wall and manifests itself as interconnected pores in the body of glass ceramic [15]. Consequently, as the sintering temperature rose, water absorption rose, and bulk density and MOR were considerably decreased. At 850°C, the MOR drops to 20.44 MPa and the bulk density to 1.83 g/cm^3. As the outer pores become larger, it will lead to an increase in water absorption, which is measured at 15.16%. This can be proven by looking at Figure 2, where large pores are clearly visible at the sintering temperature of 850°C compared to 800°C.

At a temperature of 900°C, the viscosity decreases even more and additional gases are released through carbonate oxidation. This leads to the merging and connection of more small and medium-sized pores, resulting in the formation of larger pores that become open. Consequently, the bulk density decreases to 1.75 g/cm^3 and the MOR also decreases to 19.64 MPa. Conversely, the water absorption value rises to 17.36%. Upon raising the sintering temperature to 950°C, the bulk density is increased to 1.77g/cm^3. The viscosity exhibits a similar pattern as previously observed, with a continued decrease resulting in the formation of additional pores.

Table 1 Chemical composition of the main materials of porous glass ceramics.

Oxide	Silica sand (wt. %)	Soda lime glass waste (wt. %)	Clay binder (wt. %)	Carbonate Foaming agent (wt. %)
SiO_2	97.30	74.87	40.31	0.33
Na_2O	0.04	4.84	0.13	0.04
CaO	0.42	8.70	2.43	70.13
MgO	0.28	2.24	1.03	0.60
Al_2O	0.34	0.81	5.56	0.04
K_2O	0.06	0.59	0.43	0.03
Fe_2O_3	1.39	1.04	3.84	0.12
TiO_2	0.08	0.21	0.28	0.02
LOI	0.04	6.66	45.95	28.64

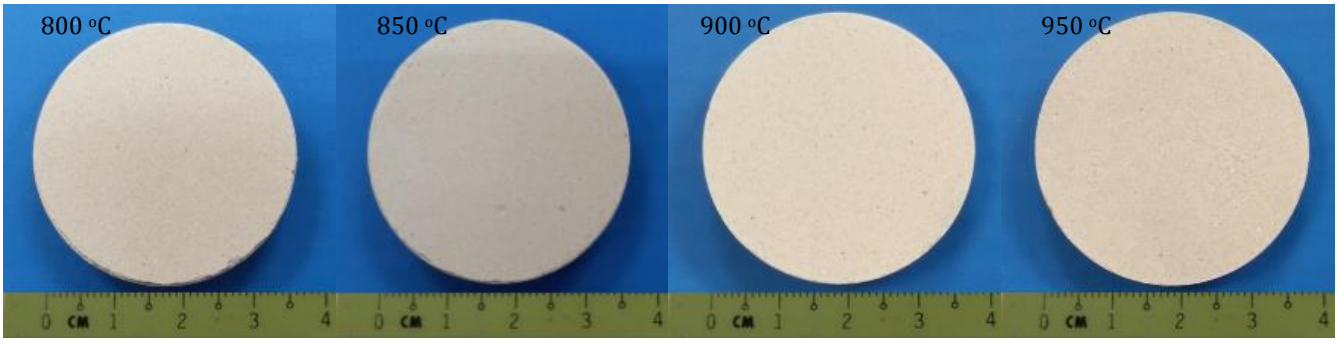

Figure 2. LGCP30 button sample after sintering at 800, 850, 900, and 950 °C.

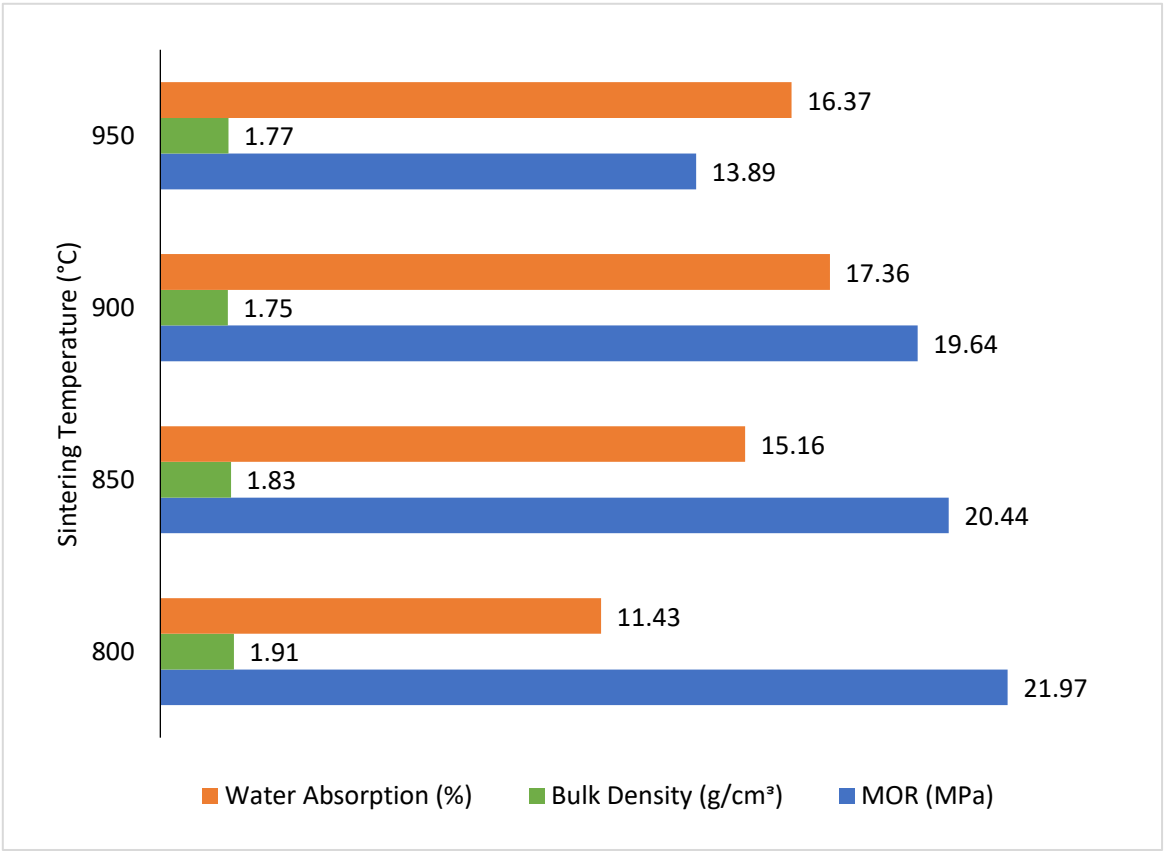

Figure 3. The result of MOR, bulk density, and water absorption of LGCP30 sintered samples at different sintering temperatures.

Consequently, this leads to a reduction in both the MOR (13.89 MPa) and water absorption (16.37%) respectively. Based on the findings, the optimal sintering temperature was determined to be 900°C. This temperature was chosen because it satisfies the requirements of ISO 13006, having a good MOR of 19.64 MPa and a lower density of 1.75 g/cm^3. These characteristics can result in a lightweight material with low sintering temperature.

3.3. Surface Morphology Analysis

Figures 4 illustrate the significant crystallisation that was identified by FESEM. The surface morphology of glass ceramic samples sintered at different temperatures from 800-950°C are shown in Figure 4. Sintering occurs at a temperature of 800°C, resulting in the formation of extremely minute pores (Figure 4a). This explains why water absorption was reported to be lowest (11.43%) at this temperature, given that water absorption is predominantly dependent on open pores. Furthermore, water absorption was diminished due to the production of minimally sized pores. Additionally, at this temperature, the glass-ceramic reaches its maximum densification (1.91 g/cm^3).

Numerous small pores were observed to form as the temperature rose to 850°C (Figure 4b), and the water absorption increased by 15.16 %. Nevertheless, water absorption was disrupted when the temperature was increased to 900°C (Figure 4c), resulting in the formation of numerous small pores and a few larger pores on the surface attributable to the lowest densification (1.75 g/cm^3). The largest water absorption (17.36%) occurs after increasing the temperature to 900°C. When the temperature rises to 950°C, it is found that there is no obvious distinction between 900°C and 950°C. However, the surface is flatter than the others, and the smaller and larger pores are getting lesser causing the water absorption to decrease (16.37 g/cm^3). As seen in Figure 4, the crystalline phase and pores are spread throughout the glassy matrix. Due to the local volume reduction involved with crystal formation, voids emerge in the structure when the glass is devitrified [16].

4. CONCLUSION

This investigation utilized silica sand and soda lime glass waste as the primary components for the production of porous glass-ceramic tiles through the utilization of the conventional solid-state powder sintering process. Clay binder served as the binder, and carbonate served as the foaming agent for the purpose of accelerating the development of pores. Increasing the sintering temperature from 800 to 950°C has had an overall impact on the physical and mechanical properties of the porous ceramic glass body that was produced.

The values of bulk density and MOR decreased as the sintering temperature increased, while the values of water absorption increased while the sintering temperature increased. The foaming agent that was utilized in this densification method results in the creation of pores and a lightweight body throughout the glass ceramic body samples. Based on the results, a sintering temperature of 900°C was chosen as the best among others since it has a lower density (1.75 g/cm^3) and good MOR (19.64 MPa) and meets the ISO 13006 standard requirement. According to ISO 13006 standard, the minimum acceptable modulus of rupture (MOR) for wall tiles must exceed 15 MPa [17].

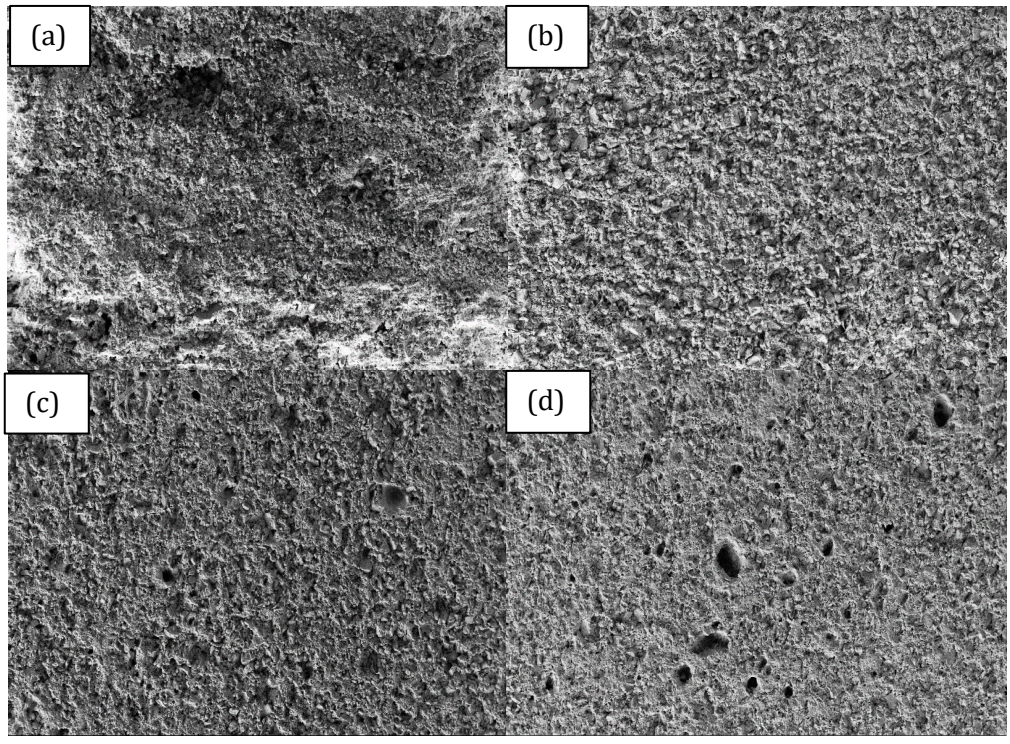

Figure 4. The surface morphology of LGCP30 samples sintered at (a) 800°C, (b) 850°C, (c) 900°C and (d)950°C.

ACKNOWLEDGMENTS

It is with great gratitude that the authors express their gratitude to the Senior Director of Mineral Research Centre, Department of Mineral and Geoscience Malaysia, for his unwavering cooperation in the production of this research and technical study. In addition, the authors would like to take this opportunity to express their appreciation to all of the support staff members working at the Mineral Research Centre and the Silica Based Technology Section for their consistent guidance, assistance, and support.

REFERENCES

[1] Lee J. C., Thedy W. H., Ng J. L., & Lee S. (2023). Recycled waste of construction industry in Malaysia: A review, IOP Conference Series: Earth and Environmental Science. Vol. 1205, Issue 1, 012030.

[2] Shado A. S., & Aliu E. T. (2021). Effects of Waste Glass on The Compressive Strength of a Sintered Ceramic Tile. I-Manager's Journal on Material Science. Vol. 19, Issue 2, 12-18.

[3] Hobson, K., Holmes, H., Welch, D., Wheeler, K., & Wieser, H. (2021). Consumption Work in the circular economy: A research agenda. Journal of Cleaner Production, 321, 128969.

[4] Villalba, G., Segarra, M., Fernandez, A. I., Chimenos, J. M., & Espiell, F. (2002). A proposal for quantifying the recyclability of materials. Resources, Conservation and Recycling, Vol. 37, Issue 1, 39-53.

[5] Mustaffar M. I., Mahmud M. H., & Hassan M. A., (2017). Development of dense glass-ceramic from recycled soda-lime-silicate glass and fly ash for tiling. In AIP Conference Proceedings. Vol. 1901, Issue 1, 030011, 1-8.

[6] Mejia E. E. (2015). Characterization and Preparation of Lightweight Silica Based Ceramics for Building Applications. Doctoral thesis, Luleå University of Technology. 1-45.

[7] Mustaffar M. I., Mahmud M. H. (2018). Processing of highly porous glass ceramic from glass and fly ash wastes, AIP Conference Proceedings. Vol. 2031, 020010, 1–6.

[8] Novais, R. M., Seabra, M. P., & Labrincha, J. A. (2015). Lightweight dense/porous bi-layered ceramic tiles prepared by double pressing. Journal of Materials Processing Technology, Vol. 216, 169-177.

[9] Sokolář, R., Keršnerová, L., & Šveda, M. (2017). The effect of different fluxing agents on the sintering of dry pressed porcelain bodies. Journal of Asian Ceramic Societies, Vol. 5, Issue 3, 290-294.

[10] American Society for Testing and Materials (ASTM), C 373 – 88 (2006). Standard Test Method for Water Absorption, Bulk Density, Apparent Porosity, and Apparent Specific Gravity of Fired Whiteware Products, 1-2.

[11] Mangutova, B. V., Fidancevska, E. M., Milosevski, M. I., & Bossert, J. H. (2004). Production of highly porous glass-ceramics from metallurgical slag, fly ash and waste glass. Acta Periodica Technologica, Vol. 35, 103-110.

[12] Rodinger, T., Ćorić, D., & Kováčik, J. (2024). The Effect of Foaming Agents on the Thermal Behavior of Aluminum Precursors. Materials, Vol. 17, Issue 3, 710.

[13] Mouiya, M., Bouazizi, A., Abourriche, A., El Khessaimi, Y., Benhammou, A., Taha, Y., & Hannache, H. (2019). Effect of sintering temperature on the microstructure and mechanical behavior of porous ceramics made from clay and banana peel powder. Results in Materials, 4, 100028.

[14] Francis, A. A., Rahman, M. A., & Daoud, A. (2013). Processing, structures, and compressive properties of porous glass-ceramic composites prepared from secondary by-product materials. Ceramics international, Vol. 39, Issue 6, 7089-7095.

[15] Zhang Q., He F., Shu H., Qiao Y., Mei S., Jin M., & Xie J. (2016). Preparation of high strength glass ceramic foams from waste cathode ray tube and germanium tailings. Journal on Construction and Building Materials. Vol. 111, Issue 15, 105-110.

[16] Cetin S. (2023). Production of sintered glass-ceramic composites from low-cost materials. Ceramics International. Vol. 49, Issue 13, 22386-22392.

[17] International Standard, ISO 13006 (2018). Ceramic tiles: Definitions, Classification, Characteristics and Marking, 1-60.