



MINING AND METALLURGY INSTITUTE BOR

and



TEHNIICAL FACULTY BOR, UNIVERSITY OF BELGRADE



**53rd International October Conference
on Mining and Metallurgy**

PROCEEDINGS

Editors:

**Ana Kostov
Milenko Ljubojev**

3 – 5 October 2022

Hotel “Albo” Bor, Serbia

53rd International October Conference on Mining and Metallurgy

Editors: Ana Kostov, Milenko Ljubojev

Publisher: Mining and Metallurgy Institute Bor

Printed in: "GRAFOMED-TRADE" Bor

Text printing preparation: Vesna Simić

Disclaimer: Authors are responsible for the content, translation and accuracy.

Circulation: 100 copies

CIP – Каталогизација у публикацији
Народна библиотека Србије, Београд

622(082)

669(082)

INTERNATIONAL October Conference on Mining and Metallurgy (53 ; 2022 ; Bor)
Proceedings / 53rd International October Conference on Mining and
Metallurgy - IOC 2022, 3 % 5 October 2022, Bor ; [organizer] Mining and
Metallurgy Bor and Technical Faculty in Bor, University of Belgrade ;
editors Ana Kostov, Milenko Ljubojev. - Bor : Mining and Metallurgy
Institute, 2022 (Bor : Grafomed-trade). - XV, 251 str. : ilustr. ; 25 cm

Tiraž 100. - Bibliografija uz svaki rad. - Registar.

ISBN 978-86-7827-052-9

a) Рударство - Зборници b) Металургија - Зборници

COBISS.SR-ID 74763529

Bor, October 2022

Conference is financially supported by the
Ministry of Education, Science and Technological
Development of the Republic of Serbia



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SINTER-CRYSTALLIZATION OF COAL FLY ASH BASED GLASS

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Abstract

The increasing production of coal fly ash waste from the thermal power plants has compounded environmental and economic problems worldwide. In this research, the coal fly ash was mixed with glass cullet and CaCO₃ and melted at T= 1500°C. The glass was milled, pressed and sintered at T= 900°C to obtain the glass-ceramic. The obtained glass-ceramic was well sintered with low crystallinity. This material, produced from the secondary raw materials, could find a potential application in the construction industry as a substitute for the commercial wall and floor tiles.

Keywords: glass, glass-ceramic, coal fly ash

1 INTRODUCTION

Coal fly ash is mainly produced by the burn of coal, petroleum products as well as the other related substances in the thermal power plants. In recent years, more and more coal fly ash has been generated and released into the environment due to the high energy demand, resulting in the pollution of environment. Specifically, the accumulation of coal fly ash would enhance the level of PM 2.5, which may lead to the respiratory disease. At the same time, discharges of a large amount of coal fly ash would also cause the pollution of soil, which may cause a heavy destruction to the ecological environment [1]. For example, the potentially toxic substances in the coal fly ash could be leached into the soil and groundwater and accumulate in the food chain [2]. The treatment of coal fly ash is imminent and has attracted more and more attention worldwide. At present, the most commonly used treatment method is dropping it into the landfills, which would not only cause waste of resources, but also lead to more serious environment pollution problems. Coal fly ash is composed of the metal oxides such as silica and alumina, which are the major components of coal fly ash alongside with oxides of iron, calcium and magnesium and other. The coal fly ash contains large amount of SiO₂ and Al₂O₃, which are the main glass network formers, therefore it is feasible to use coal fly ash as a raw material to develop glass and glass-ceramic materials [3]. During the high-temperature treatment of the coal fly ash in order to obtain glass, the destruction of organic pollutants occurs. Recycling of hazardous waste in the glass and glass-ceramic production results in many advantages; the use of a "free" raw material, conservation of natural resources, utilization of waste materials in order to reduce pollution, etc. The sinter-crystallization technique is used to obtain the glass-ceramic. This technique requires no nucleation agents and no nucleation step, which reduces the price and shortens the time of crystallization heat treatment [4]. In this research, the coal fly ash was collected from the open pit near the TPS "Nikola Tesla" – Republic of Serbia. Because there is a low concentration of glass modifier cations in the coal fly ash, the glass cullet from green

bottle glass and CaCO_3 were introduced in mixture to alter the chemical composition to a lower viscosity of melt for glass production. The potential application of glass and glass-ceramic materials, obtained from the coal fly ash can be realized in the construction industry as construction materials, building blocks, wall tiles, wear-resistant parts, anticorrosive container lining, insulation materials, etc. [5].

2 EXPERIMENTAL

The glass was prepared by a standard melt quenching procedure. The coal fly ash, glass cullet from the green bottle glass and CaCO_3 , were mixed and homogenized in an agate mortar in 37.5:37.5:25 mass ratio. The melting was performed in an electric furnace in zirconium silicate crucible at $T = 1500^\circ\text{C}$ for $t = 1$ h and the glass was obtained by quenching the melt on a steel plate. The obtained glass was first crushed in the agate mortar into cullet and then pulverized into fine particles with a TENCAN planetary mill with 400 rpm for 30 min. The obtained glass powder was sieved in order to obtain the glass powder particles size under $63 \mu\text{m}$. The glass powder was uniaxially pressed in a Manfredi C 95 laboratory hydraulic press at 20 MPa for $t = 60$ s with addition of 5% moisture as a binder. The obtained pellets weighted 1 g (15 mm in diameter and 3 mm tick) were placed in an electric furnace Carbolite CWF 13/13 and sintered for $t = 60$ min at $T = 900^\circ\text{C}$. The phase composition was defined by the XRD - Philips PW-1710 automated diffractometer with a Cu $K\alpha$ radiation tube operating at 40 kV and 32 mA. The microstructure of the glass-ceramic samples was examined using a scanning electron microscope MIRA3 XM TESCAN.

3 RESULTS AND DISCUSSION

The chemical composition of the coal fly ash and glass cullet is shown in Table 1.

Table 1 Chemical composition of the coal fly ash and glass cullet

Sample	Oxide	SiO_2	Al_2O_3	CaO	MgO	Na_2O	K_2O	Fe_2O_3	TiO_2	Cr_2O_3	L.o.i
Coal fly ash	mass%	55.33	25.11	6.26	1.42	0.32	1.23	7.08	0.50	/	2.52
Glass cullet	mass%	71.8	2.46	10.21	2.05	12.74	/	0.42	/	0.117	0.2

The coal fly ash used in this research is defined as the class F [6], and therefore chemical composition is not suitable for glass production. The glass cullet and CaCO_3 introduced necessary cations in mixture for glass production.

The chemical composition of the glass sample is shown in Table 2.

Table 2 Chemical composition of the obtained glass

Oxide	SiO_2	Al_2O_3	CaO	MgO	Na_2O	K_2O	TiO_2	Fe_2O_3	Cr_2O_3
mass %	49.54	10.58	32.88	1.58	1.44	0.72	0.34	2.68	0.08

The melt had an optimal viscosity and was poured on a steel plate. Black glass sample without visible gas bubbles was successfully obtained.

Diffractogram of sintered glass- ceramic sample is shown in Figure 1.

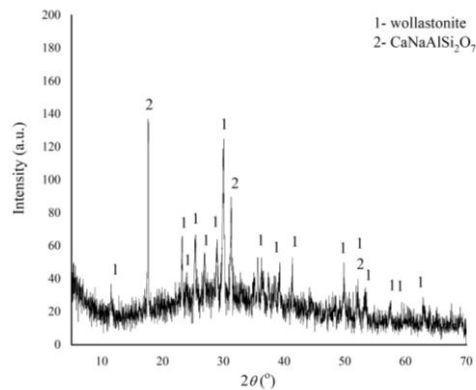


Figure 1 XRD of sintered glass- ceramic sample

The presence of the following phases was determined in the analyzed sample: wollastonite and $\text{CaNaAlSi}_2\text{O}_7$. Wollastonite is a more dominant phase compared to $\text{CaNaAlSi}_2\text{O}_7$. The degree of crystallinity of sintered sample is very low.

Figure 2 shows a micrograph of cross- section of sintered sample.

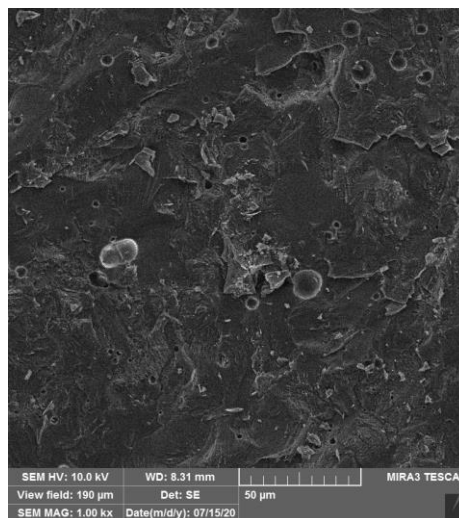


Figure 2 SEM micrographs of sintered sample at 1000x magnification

During heat treatment of sample, the sintering and crystallization processes can take place simultaneously or independent. If the mentioned processes take place simultaneously, the crystallization can inhibit sintering, so it is important to define the sintering temperature in order to obtain a high-density glass-ceramic material, because the poor sintered samples have bad mechanical properties [7]. When the glass powder is heated, in glass with surface nucleation, the processes of densification and crystallization take place at the same temperature interval, so this way of obtaining the glass-ceramic



materials is called the sinter-crystallization. As it can be seen from Figure 2, sample is well sintered with only a few internal pores left.

4 CONCLUSION

The object of this work was to produce a functional glass-ceramic material from the secondary raw materials, coal fly ash and glass cullet, with the addition of CaCO_3 . Usage of the secondary raw materials promotes the environmental protection, lowers the energy consumption and reduces the amount of waste landfilled. Glass-ceramic with high density and low amount of wollastonite and $\text{CaNaAlSi}_2\text{O}_7$ crystals was obtained. Potential application of the obtained glass-ceramic can be found in the construction industry, as wall or floor tiles.

ACKNOWLEDGEMENTS

This work was supported by the Ministry of Education, Science and Technological Development of the Republic of Serbia (Contracts No. 451-03-68/2022-14/200023 and 451-03-68/2022-14/200135).

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