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Scientific paper

PRODUCTION OF GLASS-CERAMICS FROM WASTE MATERIALS AND OPTIMIZATION OF THE MAIN PROCESS PARAMETERS

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Abstract

In this study the 3D surface model was successfully applied in investigating the influence of the process parameters on the physical and mechanical properties of the glass-ceramics. Glass-ceramics was produced from coal fly ash and waste glass through the sintering method. The raw material was taken from "REK Bitola", a thermal power plant in the Republic of Macedonia. Glass-ceramics was obtained through the process of consolidation. Compacts with different ratio of fly ash and waste glass were pressed at 45 MPa, sintered in the temperature interval from 1000 to 1100^{o}C, and isothermal time at the final temperature from 1h to 5h. The process of optimization was conducted on the process parameters such as quantity of glass, sintering temperature and isothermal time. According to the results of the process of optimization presented by the software package, a final model equations of the density and bending strength dependence were obtained.

Key words: *optimization, consolidation, process parameters, fly ash, glass-ceramics*

1. INTRODUCTION

Significant amounts of solid waste have been produced during the combustion of coal in thermal power plants. They are known as coal combustion products-CCPs or coal combustion by-products-CCBs. Worldwide production of coal combustion products was approximately 780 Mt in 2011. Effective utilization was 415Mt tones or 53% of the whole production and it varies widely within the countries [1]. The largest amount, approx. 65 wt.% of total production of CCBs belongs to the fly ash (FA) [2]. The disposal of fly ash presents the increased economical and environmental problem for the society. In Europe, more than 55wt. % of the entire fly ash production is disposed off in landfills or ash ponds. The biggest quantity of the total generated fly ash has been utilized in the construction industry either as addition to concrete [3, 4] or cement raw material [5, 6], road construction [7], ceramics [8, 9], mineral fillers [10], agriculture [11] etc. However the various reports indicate a growing interest for recycling and new applications. Numerous research and developments have been conducted to utilized fly ash for glass and glass-ceramics. Generally, there are two approaches for glass-ceramics production. The first is controlled crystallization, meaning that melted fly ash is exposed to the one or two steps of the heat treatment for crystallization followed by nucleation and crystal growth [12-15]. The second technique developed for glass-ceramics production involves sintering of the

fly ash with or without any additives [16-18]. Researchers [18] have investigated the crystallization behavior of the mixture of fly ash and waste glass in order to produce glass-ceramics and by optimizing the annealing temperature they manage to avoid the two step thermal process. The obtained glass-ceramics can be potentially used for industrial buildings because of the sufficient mechanical strength. The aim of this study was to optimize the main parameters of the process for production of glass-ceramics from fly ash and waste glass by using response surface method [19].

2. MATERIALS AND METHODS

The fly ash, coded FA, used in this study was collected from the thermal power plant "REK Bitola" in the Republic of Macedonia. Waste glass was taken as laboratory waste glass cullet mark Pyrex, coded WG. The raw materials were ground in planetary mill (Fritsch pulverisette 5) during 2h and screened through a 63 □m screen. The consolidation of the fly ash and waste glass was performed on Weber Pressen KIP 100 at 45MPa using PVA as binder.Sintering of the compacted samples was realized in the chamber furnace at temperatures from 1000^{o}C to 1050^{o}C using heating rate of 10^{0}C/min and isothermal treatment at the final temperature from 1 to 5 h. Bulk density of the glass-ceramics was determined by the water displacement method according to EN-993. The value of theoretical density of the compacts was calculated based on the composition of the initial mixture of fly ash and waste glass. The bending strength of the glass-ceramics was determined with a 3-point bending strength tester (Netzsch 401/3) with a 30 mm span and a 0.5 mm/min loading rate on the sintered samples (with the dimensions of 50 mm x 5 mm x 5 mm).The optimization of the process for production of glass-ceramics (GC) was performed with software package "Statgraphics Centurion" [20]. The influence of the main process parameters: sintering temperature, quantity of glass and isothermal period and their interactions on the physical and mechanical properties of obtained dense glass-ceramic was determinate. The optimization was realized through the application of the 3D surface model and the results are presented in a graphical and an analytical form.

3. RESULTS AND DISCUSION

Morphology, chemical composition and phase composition of the fly ash and waste glass were previously reported [21]. It can be concluded that the morphology of fly ash particles consists of particles with diameter from 5 to 60 μm with irregular shape, spherical and partially spherical while the particles of WG have irregular geometry and dimension between 10 and 60 μm.

3.1. Optimization process 1: optimization response of density as a function of sintering temperature, glass content and isothermal period

The optimization process was based on the influence of three main process parameters: sintering temperature, glass content and isothermal period. The obtained data are presented in Figure 1-4. According to the Pareto chart (Fig.1) the isothermal period at the final sintering temperature is highly influential to the density of the glass-ceramics (GC) followed by glass content and then sintering temperature. The interaction between two parameters, temperature and isothermal period (AC), has the insignificant influence to the density of the GC. The influence of the all investigated parameters on the density refers to their maximal values.

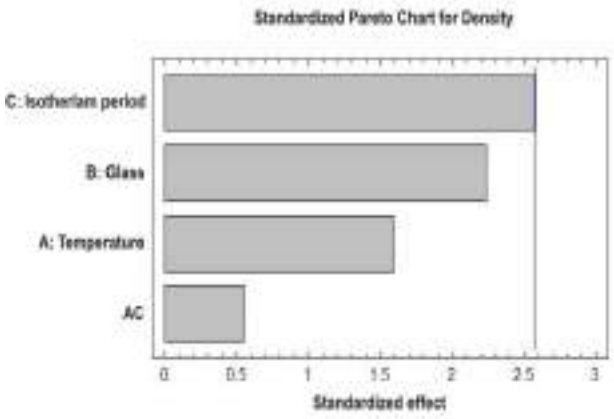

Figure 1. Statistic influence of the main process parameters and their interactions on the density of the GC compacts

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The 3D optimization diagram of the main effects obtained at constant isothermal period of 3h and variable sintering temperature and glass content (Fig. 3) shows that the maximal value of the density can be achived with a 30wt.% content of glass and sintering temperature at 1050°C (right top corner). It's possible to achive the same values of the density (broad red area, Fig. 4) by reducing the sintering temperature and increasing the glass content up to 40 wt.% which is more acceptable due to economical considerations.

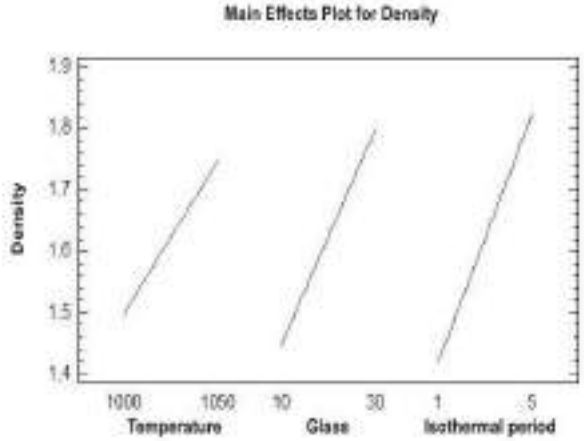

Figure 2. Diagram of the main effects of the process parameters on the density of GC compacts

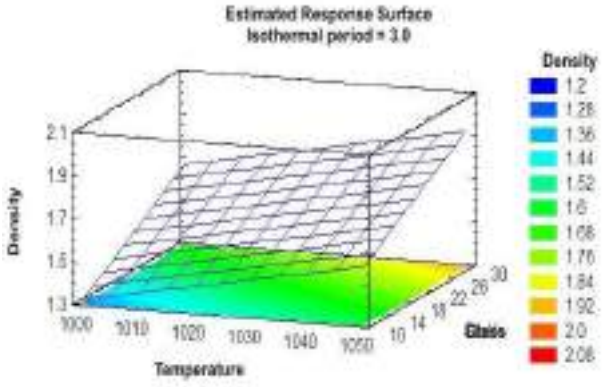

Figure 3. 3D optimization diagram of the main effects at constant isothermal period (3h) and variable sintering temperature (°C) and glass content (wt.%)

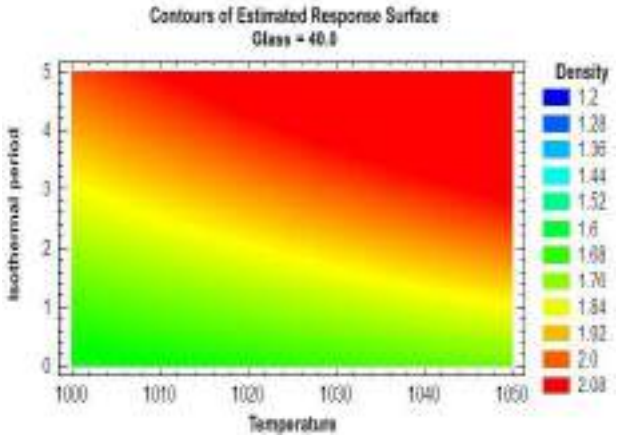

Figure 4. Optimization diagram of the main effects at constant glass content (wt.%) and variable sintering temperature (°C) and isothermal period (h)

According to the results of the process of optimization presented by the software package, a final model equation for the density (D) dependence on the process variables is:

$$D=-1.49515+0.0024025*T+0.00175875*G-0.787875*IP+0.0006875*T*IP \quad (1)$$

where T is the sintering temperature, G is glass content and IP is isothermal period at the final temperature.

3.2. Optimization process 2: optimization response of bending strength as a function of sintering temperature, glass content and isothermal time

The second optimization process was conducted by investigating the influence of the glass content, sintering temperature and isothermal time as process variables on the bending strength as response function. The results of this optimization are presented in the Fig. 5-8.

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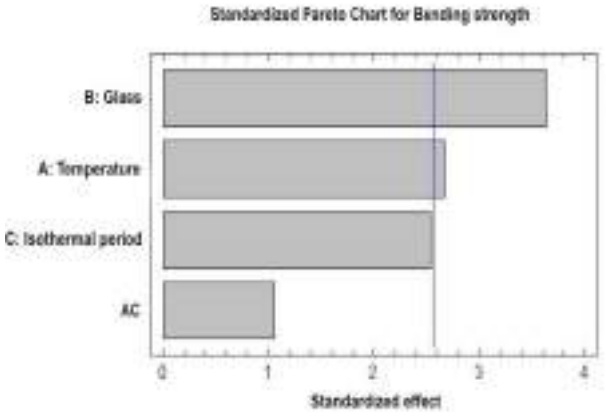

Figure 5. Statistic influence of the main process parameters and their interactions on the bending strength of GC compacts

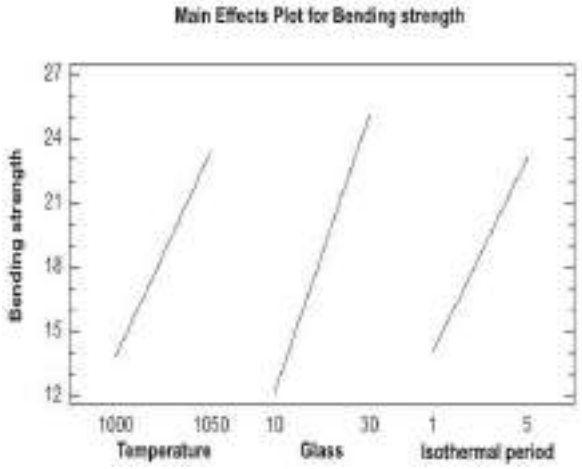

Figure 6. Diagram of the main effects of the process parameters on the bending strength of GC compacts

By analyzing the Pareto chart (Fig. 5), it is evident that bending strength of the GC compacts is directly dependent on the glass content, but also sintering temperature and isothermal period have significant influence as variables. As the previous optimization, the interaction between two parameters temperature and isothermal period (AC) has the insignificant influence on the bending strength of the GC. The conclusion from the 3D optimization diagram (Fig. 7) is that the maximal value for the bending strength of 33MPa is achived at sintering temperature of 1050°C, isothermal period of 3h and glass content of 30 wt. % (top right corner). For the constant glass content, the maximum value of bending strength can be obtained by broad combination of the parameters presented with red colour on the optimization diagram of the main effects (Fig. 8). A final model equation fof the bending strength (σ) dependence on pthe process parameters is:

$$\sigma = -80.709 + 0.07755*T + 0.65075*G - 36.625*IP + 0.03795*T*IP \quad (2)$$

where T is the sintering temperature, G is glass content and IP is isothermal period at the final temperature.

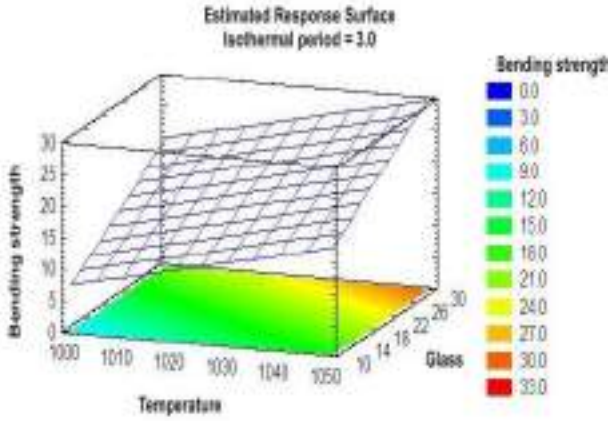

Figure 7. 3D optimization diagram of the main effects at constant isothermal period (3h) and variable sintering temperature (°C) and glass content (wt.%)

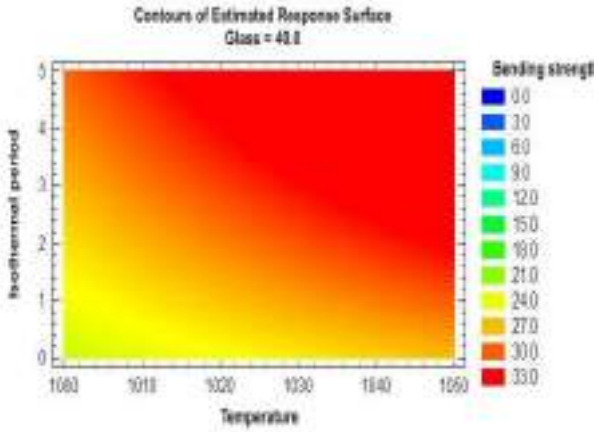

Figure 8. Optimization diagram of the main effects at constant glass content (wt.%) and variable sintering temperature (°C) and isothermal period (h)

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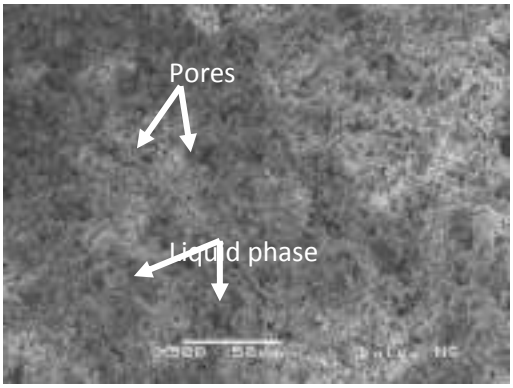
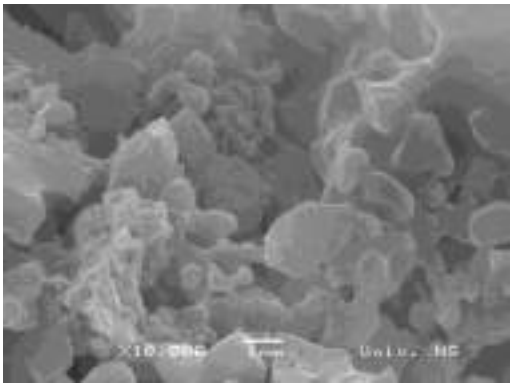
3.3. Microstructure of the glass-ceramics

The investigation of the microstructure of the glass-ceramics performed by SEM analysis presented in Fig. 9 and Fig. 10 shows that the fractured surface of the sample with 10 wt. % glass content is granular with evident presence of grains from the particles of raw materials. In some areas the liquid phase is recognizable, as well as the presence of some open pores. (Fig. 9a)

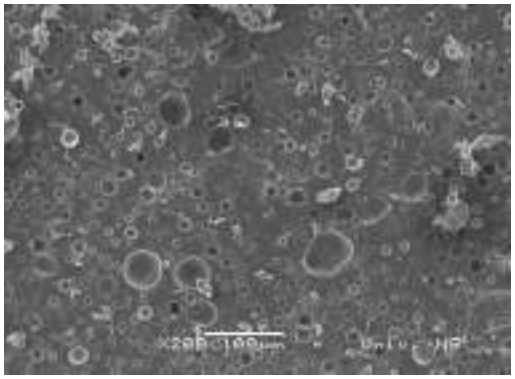


(a) x500, x500, bar 50μm

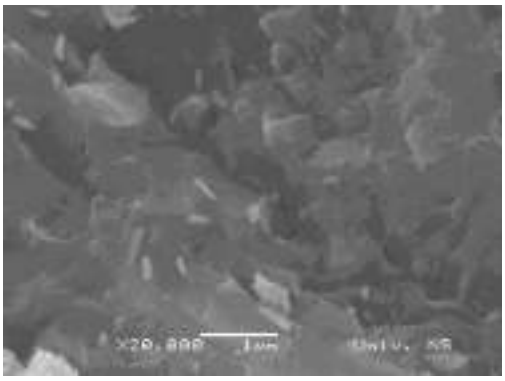


(b) x10000, bar 1μm

Figure 9. SEM micrograph of GC with 10wt% glass content, sintered at, T=1050°C and 1h isothermal period



(a) x200, bar 100μm



(b) x20000, bar 1μm

Figure 10. SEM micrograph of GC with 40wt% glass content, sintered at, T=1050°C and 1h isothermal period

The microstructure of the fractured surface of glass-ceramics obtained with glass content of 40 wt. % is much denser and smoother compared to the previous one, sintered at the same temperature. The grains of fly ash are well embedded in the glassy matrix. Unconnected spherical pores with dimensions 5-50 m are present due to softening of the glassy phase or decomposition of the alkaline metal salts [22].

4. CONCLUSION

The process of optimization was conducted based on the influence of the main process parameters and their interactions on the properties of obtained glass-ceramics.

The final model equation for the density dependence on the process parameters for glass-ceramics is:

D = -1.49515 + 0.0024025*T + 0.00175875*G - 0.787875 *IP+ 0.0006875*T*IP

The final model equation of the bending strength dependence on the process parameters for GC is:

σ = -80.709 + 0.07755*T + 0.65075*G-36.625*IP +0.03795*T*IP

The obtained model equations for the density and bending strength depending on the main process parameters present solid basic data for modeling the process of glass-ceramic production.

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