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Original scientific paper

Optimization of the Process of Production of Ceramics From Waste Coal Ash Case Study: The Influence of the Mechanical Activation

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Abstract: Fly ash, a waste by-product obtained in a thermal power plant has been a generated problem of the disposal all over the world. Morphological characteristics, physicochemical properties and pozzolanic activity make this waste potential material for production of ceramics. In this study high density ceramics compacts were produced by using fly ash from the power plant REK Bitola, Republic of Macedonia. In order to increase geometrical factor of activity, the mechanical activation of the fly ash was applied. The process of optimization of the main process parameters is conducted, such as time of mechanical activation, sintering temperature and heating rate and their interactions on the properties of obtained dense ceramic porosity and bending strength as a response function. The optimization was performed through application of 3D surface method and the obtained results are presented in the graphical and analytical form using "Statgraphics Centurion" software package.

Key words: fly ash, optimization, consolidation, mechanical activation, ceramics.

Introduction

Great quantities of coal ash are produced in thermal power plants causing economical and environmental problem to the society. According to [European Coal Combustion Products Association, 1990] the total quantity, in Western Europe in 2009, there was 34 million tons with tendency of growing up to 66 million tons and more. **Fly ash** is obtained by electrostatic precipitation of dust-like particles from the gases of furnaces fired with coal at 1100 to 1400°C. Fly ash is a fine powder, which is mainly composed of spherical glassy particles. These particles present a significant source of oxides such Fe_2O_3, Al_2O_3, SiO_2, MgO, Na_2O and others.

Fly ash is utilized in a wide range of applications in the construction industry [Wang, Lin, & Huang, 1994] [Monzo, Paya, & Peris-Mora, 1994] [Caires, & Peters, 2011] [Feng, & Clark, 2011], replacement material in road construction [Lav, Lav, & Goktepe, 2005] [Mistra, Upadhyaya, & Biswas, 2003], as mineral fillers [Çokça, 2001] [Pimraksa, & Thongchai, 2006] and as fertilizers [Kumar, Zacharia, & Goswami, 2011].

There has been considerable research on the production of ceramics from coal fly ash with the addition of the natural raw materials [Fukumoto, & Kanda, 2009] [Maitra, Das, Das, & Basumajumdar, 2005] or waste materials [Angjusheva, 2011] [Bossert, et al, 2004] [Mangutova, & et al, 2004] [Yoon, & Yun, 2005].

There are few works that deal with the topic of production of ceramics from pure fly ash [Pimraksa, et al] [Angjusheva, Fidancevska, & Jovanov, 2012]. The effect of particle size distribution, as a key process variable that controls sintering and the properties of the ceramics, was reported [Ilic, et al, 2003]. They studied the influence of treatments such as sieving and grounding on the properties of the sintered ceramics.

The aim of this paper is to optimize the process of production of dense ceramics by applying mechanical activation of the fly ash.

The process of optimization is performed by the process parameters, such as time of mechanical activation, sintering temperature and heating rate and their interactions on the physical and mechanical properties of ceramics.

Materials and Methods

The fly ash from thermal power plant "REK Bitola" from Republic of Macedonia was used as a raw material. Investigation was conducted on the fly ash fraction lower than 63μm, coded as FA/63.

The raw materials were ground in the ball mill during 5 and 10 h coded as FA/63/5 and FA/63/10, respectively.

Pressing of the samples was performed by uniaxial press (Weber Pressen KIP 100) at P= 45 MPa using PVA as a plastificator.

Sintering of the compacted samples was realized in the chamber furnace in the air atmosphere at temperatures 950, 1000, 1050, 1100, 1150^{0}C using heating rate of 3 and 10^{0}C/min and isothermal treatment at the final temperature of 60 min.

Bulk density of the sintered samples was determined by water displacement method according to EN-993. Porosity of the samples was calculated from the relative density.

The bending strength was measured on the sintered samples (with the dimensions of 50 mm x 5 mm x 5 mm), which were subjected to a 3-point bending strength tester (Netzsch 401/3) with a 30 mm span and a 0.5 mm/min loading rate. For the mechanical tests at least five samples were used and the results were averaged.

The optimization was conducted based on the influence of the main process parameters: sintering temperature, time of activation and heating rate and their interactions on the properties of obtained dense ceramic such as porosity and bending strength. The optimization was performed through application of 3D surface model and results are presented in the graphical and analytical form. Software package used was "Statgraphics Centurion".

Results and Discussion

The chemical composition of the raw fly ash (a fraction less than 63μm) is given in the previous work [19]. Investigated fly ash contains high amount of SiO_2, Al_2O_3 and Fe_2O_3 and CaO. The content of CaO is higher than 10% and can be characterized as Class C fly ashes [Annual Book of ASTM Standards].

Investigation of the phase composition shows the presence of SiO_2 - quartz, $CaAl_2Si_2O_8$ - anorthite, Fe_2O_3 - hematite, $NaAlSi_3O_8$ - albite, $CaSO_4$ - anhydrite and an amorphous phase [Angjusheva, Fidancevska, & Jovanov, 2012].

Optimization process 1: optimization response of porosity as a function of time of mechanical activation, sintering temperature and heating rate.

The process of optimization was conducted based on the influence of the main process parameters and their interactions on the property of obtained ceramics. The response function is presented in the following figures:

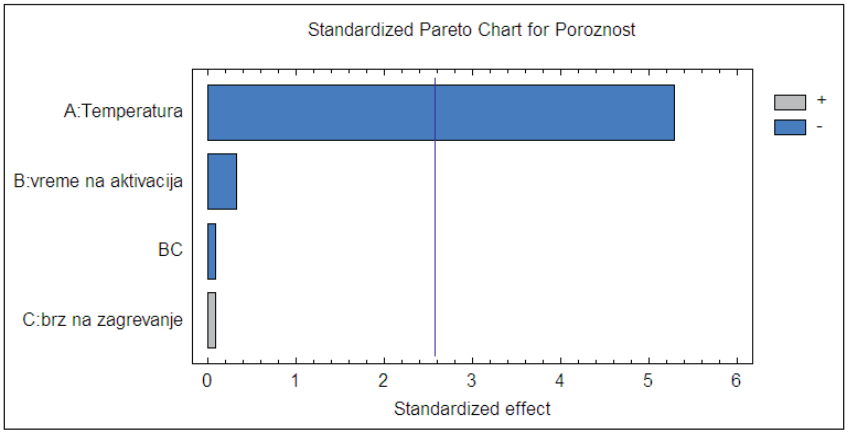

Figura 1. Statistic influence of the main process parameters and their interactions on the porosity of FA compacts

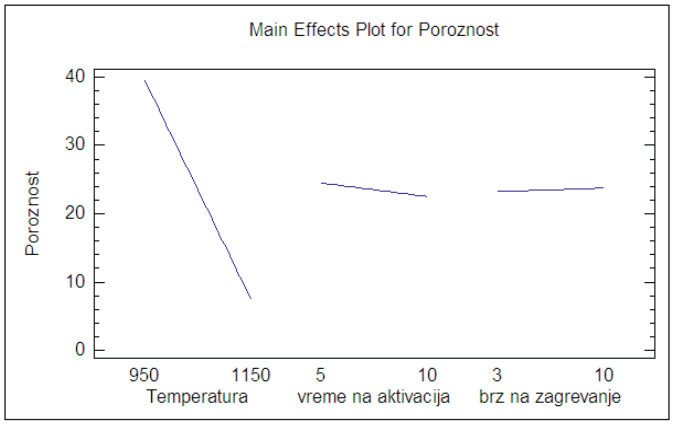

Figura 2 Diagram of main effects of process parameters on the porosity of FA compacts

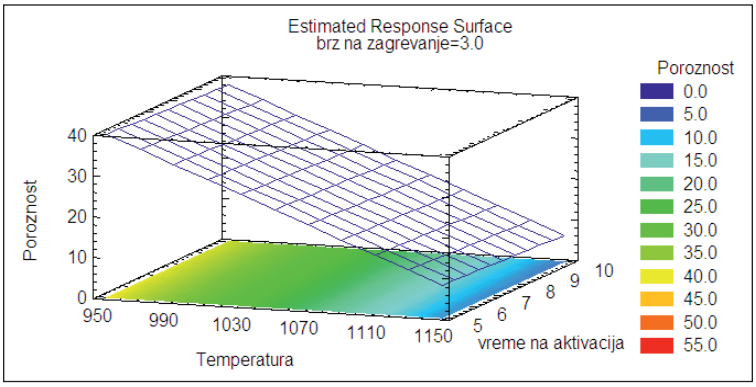

Figura 3. 3D optimization diagram of the main effects at constantvalue of heating rate (°/min) and variable temperature (°C) and activation time (h)

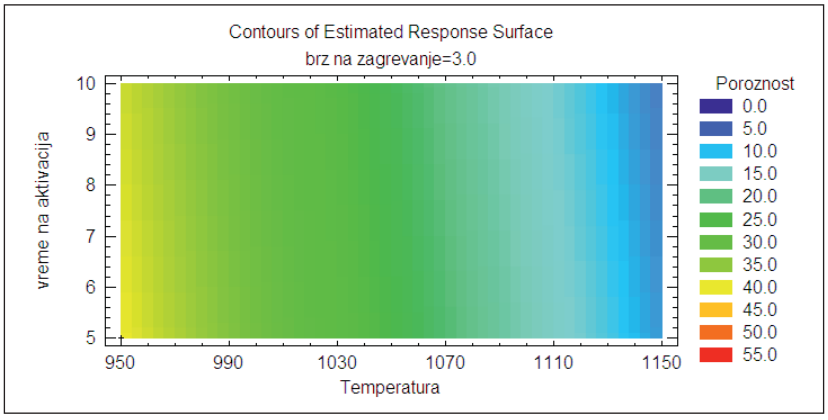

Figura 4. Optimization diagram of the main effects at constant value of heating rate (°/min) and variable temperature (°C) and activation time (h)

According to the results of the process of optimization presented by the software package, a final model equation of the porosity is:

Porosity= 192.463-0.16*Temperature-0.21428*Activation time+0.285714*Heating rate-0.0285714*Activation time*Heating rate

In this case of optimization, the response value of porosity of the FA compacts was examined as a function of the process parameters – sintering temperature, activation time and heating rate. It is evident that the porosity of the FA compacts, as the main process parameter that defines the densification of the ceramic body, is directly dependent on the sintering temperature. The influence of the activation time and heating rate on the porosity of the FA compacts is smaller compared to the sintering temperature. From the

surface area (3D) optimization diagram (Fig.3) and optimization diagram of the main effects (Fig.4) it is evidant that optimal parameters are sintering temperature 1100°C and activation time of 5 h at constatnt heating rate of 3 °C/min.

Optimization process 2: optimization response of the bending strength as a function of the sintering temperature, activation time and heating rate.

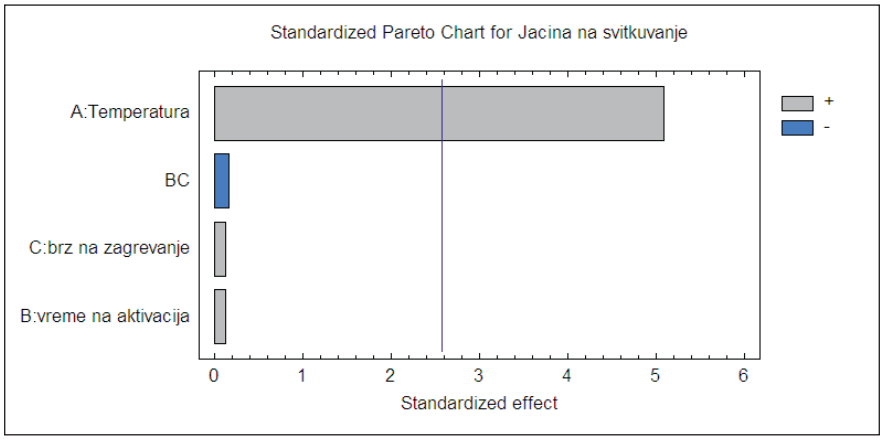

Figura 5. Statistic influence of the main process parameters and their interactions on the bending strength of FA compacts

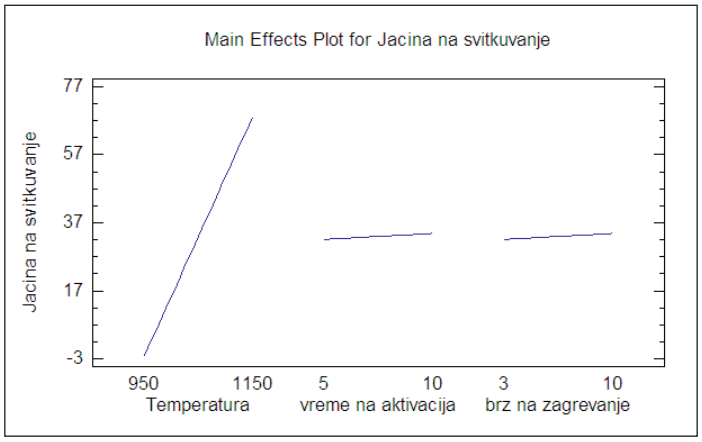

Figura 6. Diagram of main effects of process parameters on the bending strength of FA compacts

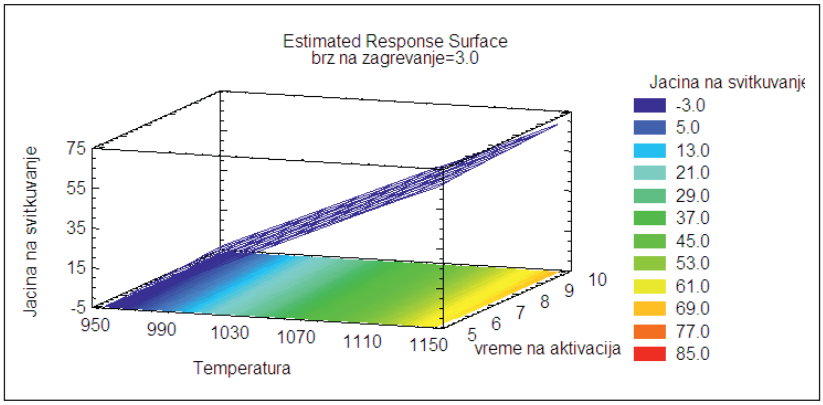

Figura 7. 3D optimization diagram of the main effects at constant heating rate (°/min) and variable temperature (°C) and activation time (h)

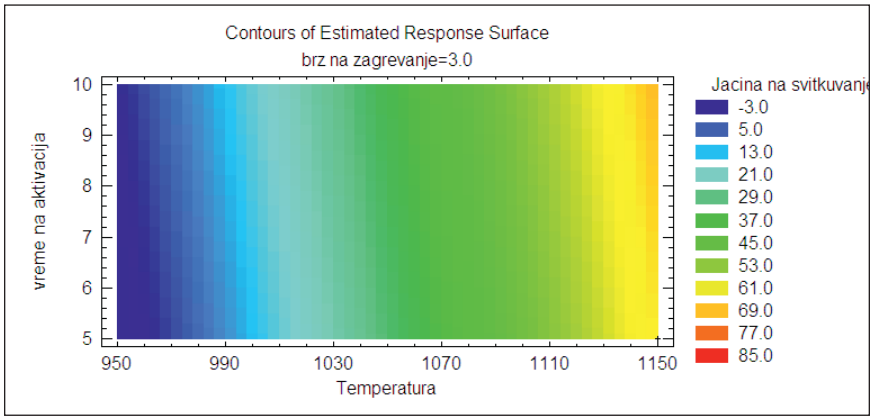

Fig. 8 Optimization diagram of the main effects at constant heating rate (°/min) and variable temperature (°C) and activation time (h)

According to the results of the process of optimization presented by the software package, a final model equation of the bending strength dependence is:

Bending strength = -346.63+0.35125*Temperature+1.18571*Activation time+1.21429* Heating rate-0.128571*Activation time*Heating rate

In this case of optimization, the response value of the bending strength of the FA compacts was examined as a function of the process parameters – activation time, sintering temperature and heating rate. By analyzing the Pareto chart (Fig.5), it is evident that the bending strength of the compacts is directly dependent on the sintering temperature. The influence of the heating rate on the mechanical properties is smaller compared to the other parameters. From the 3D optimization diagram (Fig.7) and optimization diagram of the main effects (Fig.8) it is evidant that optimal maximum for the bending strength (60-75 MPa) was obtained at sintering temperature 1100°C and activation time of 5 h.

Conclusion

- In these investigations, the fly ash, by-product from the thermal power plant REK Bitola, Republic of Macedonia, was used for production of dense ceramics;
- 3D optimization method has been successfully applied in order to determine the optimal operating areas;
- The final model equation for porosity as a response function is:

  Porosity= 192.463-0.16*Temperature-0.21428*Activation time+0.285714*Heating rate-0.0285714*Activation time*Heating rate
- The final model equation of the bending strength dependence is:

  Bending strength = -346.63+0.35125*Temperature+1.18571*Activation time+1.21429* Heating rate-0.128571*Activation time*Heating rate
- The presented investigations in this article show the possibility of performing the optimization of the process for the production of dense ceramics from coal fly ash with the accent on the mechanical activation of the fly ash.
- However, for further investigation it is possible to apply the optimization method including other process parameters affecting physical and mechanical properties of the ceramics.

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