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Original scientific paper

Production and Characterization of Porous Ceramics From Coal Fly Ash and Clay

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Abstract: The disposal of coal fly ash obtained in thermal power plants presents the general problem all over the world. Significant research on the utilization of fly ash has been carried out in the area of construction materials. The aim of this study was to develop porous ceramics based on coal fly ash and clay (60wt.%clay and 40wt.% fly ash). Three types of pore creators: two types of wood cutting (Quercus and Facus sylvatica) and C-powder were used for creating of the porous ceramics. The mixtures based on fly ash and clay and different content of pore creators (2, 5, 10 and 20wt.%) were consolidated (P=45 MPa, T = 900, 1000, 1050 and 1100^{o}C/1h) to obtain porous ceramic (PC). The results indicate that the properties of the porous ceramics depend on the type and content of the pore creators. Furthermore, the sintering temperature was found to be main factor affecting the properties of the sintered products. The maximal bending strength (26 MPa) was achieved by using 2wt% P3 (C-powder) and the porous ceramics has the density and porosity of 1.90g/cm^3 and 22%, respectively. By using the highest content (20wt.%) of each pore creator (P1, P2 and P3) the lowest bending strength cca 5 MPa was achieved and the variation of the density and porosity was in the range from 1.22 to 1.32 g/cm^3 and 44 to 48%, respectively. Water absorption, durability and the microstructure of the obtained porous ceramics are also reported in this paper.

Keywords: fly ash, clay, porous ceramics, bending strength, E-modulus.

Introduction

Rapid growth of necessity for electricity in the world contributes for the gradually increasing of amount of coal ash generated in the power plants. In Europe over 62 million tons of fly ash is generated annually (ECOBA, 2010), approximately 43 wt.% is used mostly in the construction industry while the rest is stored or sent to disposal which present a major economical and environmental problem. The highest utilization of coal fly ash is in the cement industry for production of cement or as concrete addition (Manz, 1997). However the increasing generation of coal ash made it necessary to develop new application. Clay based ceramics is the mostly used in the construction industry. The similarity in the chemical composition of the fly ash and clay make the fly ash a possible substitute for clay consumption. By using different materials as pore creators there are many procedures that are usually employed to make porous ceramics products (Wu, Boccaccini, Lee, Kershaw and Rawlings, 2006; Bossert et al., 2004; Mangutova, Fidancevska, Milosevski and Bossert, 2004). Wang et al. (2008) reported for developing lightweight aggregate from fly ash and dry sewage sludge. They concluded that sintering temperature and the proportion of the coal ash are the primary factors affecting the properties of the final product. Attractive sintered foam materials with excellent mechanical properties and durability are obtained by Zhao et al. (2010) by alkali activated coal ash and 13 wt.% foaming agent. The foam materials with compressive strength of 6.76 MPa have homogenous microstructure with interconnected pores which made them good engineering material. Ramamurthy and Harikrishnan (2006), investigated the influence of the three binders on properties of sintered fly ash aggregate. They reported a significant improvement of the strength and reduction in water absorption of sintered fly ash aggregate when 20% sodium bentonite was added to the fly ash. The insulating bricks from fly ash and clay were developed by Gonzalez Otero, Bianco Garcia and Ayala (2004) by adding sodium silicate as a binder and hydrogen peroxide as foaming agent.

Manufactured bricks composed of fly ash, clay, sodium silicate and hydrogen peroxide were obtained at optimal sintering temperature of 1000°C.

The aim of this paper was to investigate the possibility for production porous ceramics (starting from the mixture 60wt.% clay and 40wt.% fly ash) by using three types of pore creators: two types of wood cutting (Quercus and Facus sylvatica) and C-powder. The type and the content of the pore creators as well as the sintering temperature were discussed in relation to the density, porosity, water absorption and mechanical properties (bending strength and E-modulus) of the porous ceramics.

Materials and Methods

Fly ash (REK, Bitola, Republic of Macedonia) and clay (near the region of Bitola) were used as raw materials for creating porous structure. The composite composition employed in this investigation was 60wt.% clay and 40wt.% fly ash. Three types of pore creators were used for creating porous structure of the clay – fly ash composite. Morphological characterization of the pore creators was realized with stereo optical microscopy (Leica). Two types of wood cutting P1 (Quercus); P2 (Facus sylvatica) and P3 (C-powder) all with dimensions smaller than 0.5 mm were incorporated in the composite mixture as pore creators in quantity of 2, 5, 10, 20 wt.%. The ash content of the pore creators was determined after 1 hours of thermal treatment at 800°C. The moistened samples were uniaxial pressed (Weber Pressen KIP 100) at P=45 MPa. The green bars were dried 24 h at room temperature and 24 h at 110°C. Green density was calculated from the mass/volume ratio of the pressed samples.

Sintering was realized in the chamber furnace in the air atmosphere at temperatures 900, 1000, 1050 and 1100°C, using the following heating regime: RT-600°C with heating rate of 2°C/min and 600°C -FT with heating rate of 5°C/min and isothermal treatment at the final temperature (FT) of 1 hour. Bulk density of the sintered samples was determined by water displacement method according to EN-993. The obtained porous ceramics (example: PC2%P1) are assigned as: PC – porous ceramics; 2, 5, 10 and 20 wt.% - content of the pore creator, P – type of the pore creator (mentioned above).

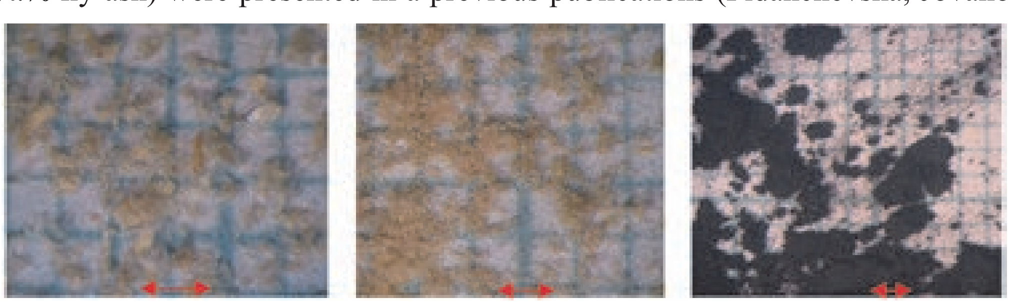
Three point bending tester (Netzsch 401/3) with 30 mm span and 0,5 mm/min crosshead speed was used to determined mechanical properties (E- modulus and bending strength). Water absorption was determined according ISO 10545-3 using boiling method.

The microstructure of the porous ceramics was followed by using scanning electron microscopy (SEM Leica S 440i).

Chemical durability of porous samples (sintered at 1100°C/1h) was tested using the standard methods for ceramics. It was determined as mass loss after treatment in neutral (H_2O) and aggressive mediums (0.1M HCl and 0.1M Na_2CO_3) during the period of 24, 168 and 720 hours.

Results and Discussion

The characterization of fly ash and clay and the optimization of the composite composition (60wt.% clay and 40 wt.% fly ash) were presented in a previous publications (Fidanchevska, Jovanov, Angusheva



P1, (bar 1 mm) P2, (bar 1 mm) P3, (bar 1 mm)

Figure 1. Photographs of the pore creators P1, P2 and P3

and Srebrenkoska, 2014; Angjusheva, Jovanov, Fidancevska and Lisickov, 2013). The morphology of the three pore creators employed in this investigation are presented in Fig.1. It is evident that the particles are with irregular geometry, while the pore creator P1 has more elongated particles.

The ash content of pore creators P1, P2 and P3 was 0.23, 1.85 and 7.29 wt.%, respectively, obtained after firing at 800°C/1h. The P3 pore creator (C-powder) showed the biggest ash content (7.29 wt%) due to the presence of the mineral phases, while the both wood pore creators P1 and P2 showed lower content of the ash which is due to the presence of the water and organic compounds (cca 99 and 98 wt.%).

The broad spectrum of porous ceramics was obtained after the consolidation (pressing at 45 MPa and sintering upon the above described temperature regime) procedure. The density and porosity of the porous ceramics (PC) obtained at above temperatures and different types and content of the pore creators are presented in the Fig. 2 and 3. The density and porosity in relation to the content of the three types of pore creators (P1, P2 and P3) for the porous ceramics (PC) sintered at 1100°C/1h are shown in Fig.4 and 5. Fig. 6 shows the water absorption of the porous ceramics (PC) in relation to the sintering temperatures and different types and content of the pore creators.

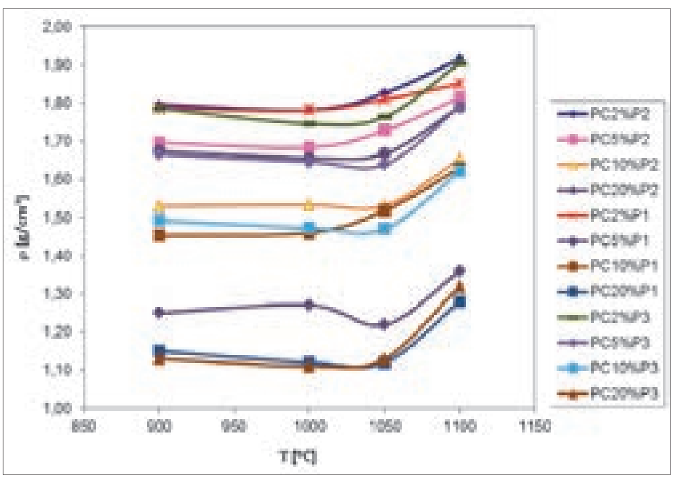


Figure 2. Density of the porous ceramics in relation to sintering temperature

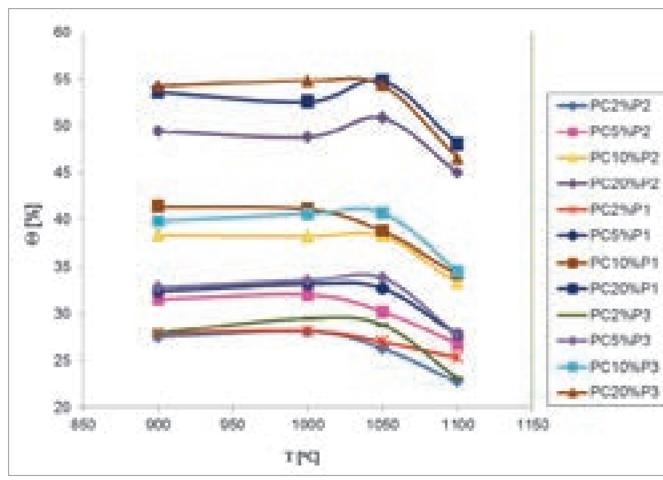


Figure 3. Porosity of the porous ceramics in relation to sintering temperature

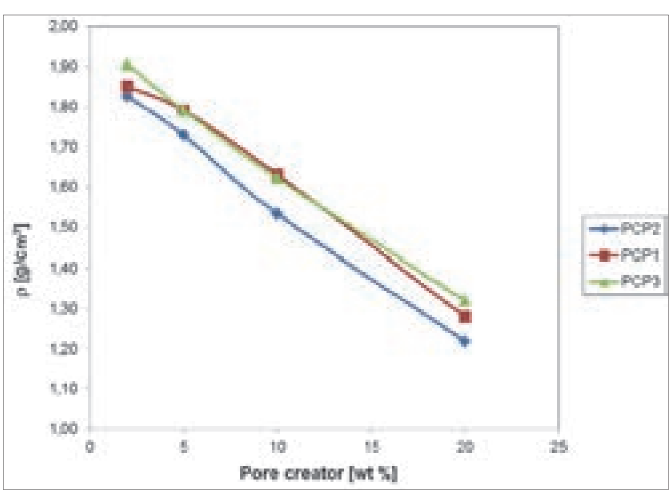


Figure 4. Density of the porous ceramics in relation to content of pore creator for PC samples sintered at 1100°C/1h

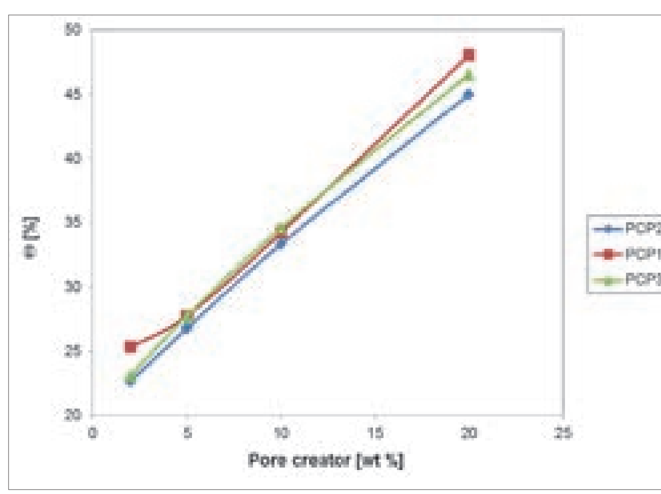


Figure 5. Porosity of the PC in relation to content of pore creator for samples sintered at 1100°C/1h

Generally, it is evident from the Fig. 2 and 3 that significant increase of the density and decrease of porosity for the sintered porous ceramics occurred at temperature range 1050 -1100°C for the three applied pore creators. As the content of each pore creator increased (2, 5, 10 and 20 wt.%) the density decreased (in the range from 1.83-1.91g/cm^2 for 2 wt.% up to 1.22-1.32 g/cm^2 for 20 wt.% of the applied pore creators) and porosity increased (in the range from 23 – 25% for 2 wt.% up to 45-48 % for 20 wt.% of the pore creators P1-P3). The temperature of 1100^{o}C/1h is considered as optimal for the investigated temperature region where the porous ceramics achieved the maximal densification. Namely, the density, Fig.4, varied from 1.22 to 1.91 g/cm^3 and the range of porosity, Fig.5, from 23 to 48 % was achieved depending on the type and content of the pore creator. The water absorption of the porous ceramics sintered at 1100^{o}C/1h varied from 26 % when 20 wt.%P3 was used as pore creator to 8.4 % when 2 wt.% of P1 and P3 were used. When the addition of pore creators is 2 wt.% the water absorption is lowest as the result of smaller content of interconnected pores. By increasing the volume of pore creators more interconnected pores appeared in the porous ceramics resulting in higher water absorption.

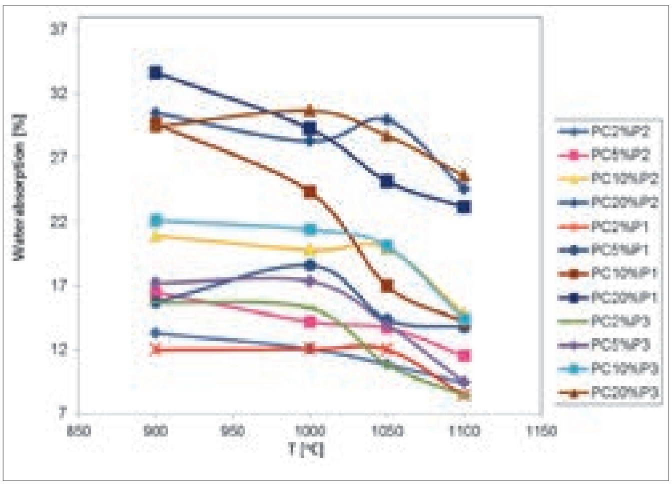


Figure 6. Water absorption of the PC in relation to sintering temperature

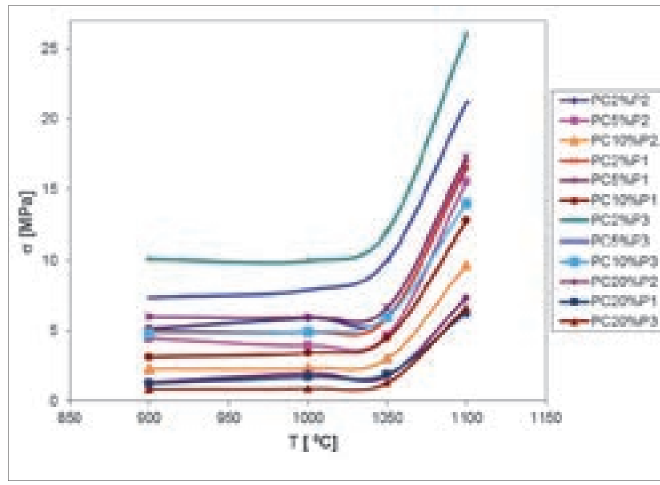


Figure 7. Bending strength of the porous ceramics in relation to sintering temperature

Fig.7 and Fig. 8 reveal the variation of mechanical properties (bending strength and E-modulus) of the sintered PC specimens in relation to the sintering temperature, while Fig.9 presents the bending strength of the PC depending on the type and content of the pore creators for samples sintered at 1100^{o}C/1h.

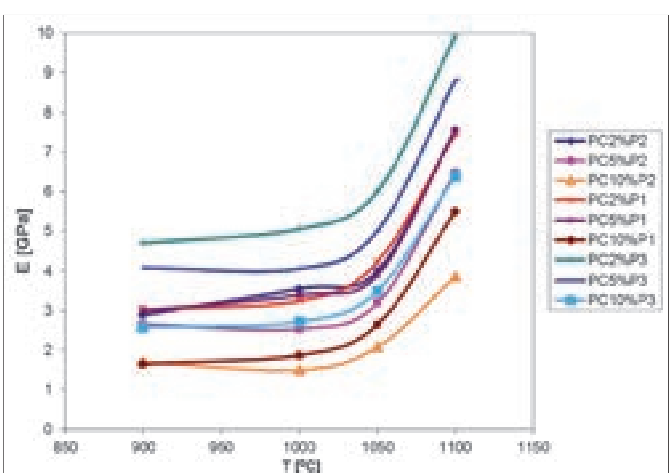


Figure 8. E-modulus of the porous ceramics in relation to sintering temperature

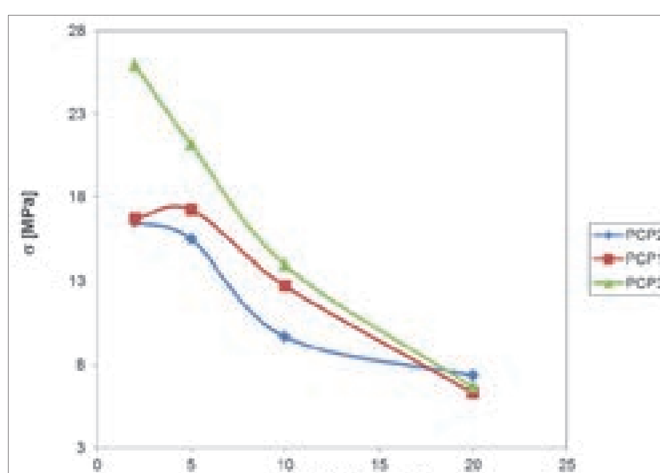


Figure 9. Bending strength of the PC in relation to content of pore creator for samples sintered at 1100^{o}C/1h

Up to 1000°C/1h the mechanical properties of the PC samples are lower than 10MPa for the bending strength and 5 GPa for the E-modulus (Figs. 7 and 8). The highest values of bending strength for each composition of PC were achieved at 1100°C/1h. Bending strength is inversely proportional to the content of the pore creator. Furthermore it can be noted that for the content of the pore creator of 2, 5 and 10 wt% the bending strength of the PCP3 is higher than the PCP1 and PCP2. The highest value of the bending strength of 26 MPa was achieved on the porous ceramics created with 2 wt.% C sintered at 1100°C/1h (Fig.9). The difference of bending strength for the porous ceramics composed by using 20 wt.% of each pore creator is insignificant.

The durability of the investigated porous systems sintered at 1100°C/1h presented as mass loss after 24h, 168h and 720h in acid, base and neutral medium (Figs. 10, 11 and 12) shows that it is correlated with the time of exposure and the porosity of the samples. More obvious mass loss is in the acid media for all investigated periods, while mass loss in base and neutral media were 0.52 % and 0.25 %, respectively.

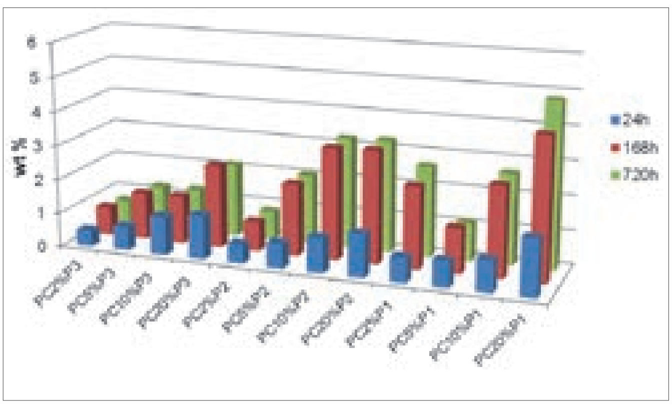


Figure 10. Leaching of the PC in acid medium (0.1MHCl) for samples sintered at 1100°C/1h

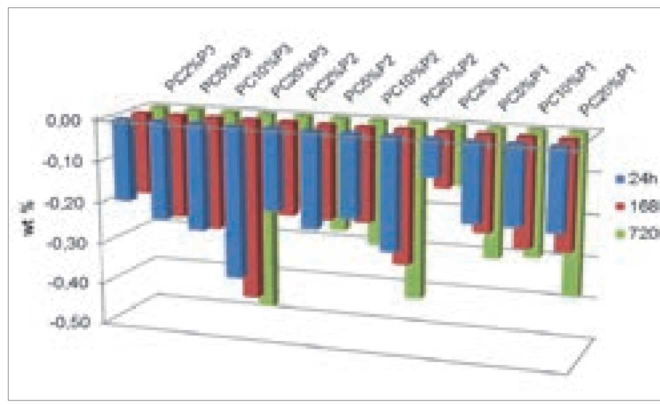


Figure 11. Leaching of the PC in base medium (0.1MNa_2CO_3) for samples sintered at 1100°C/1h

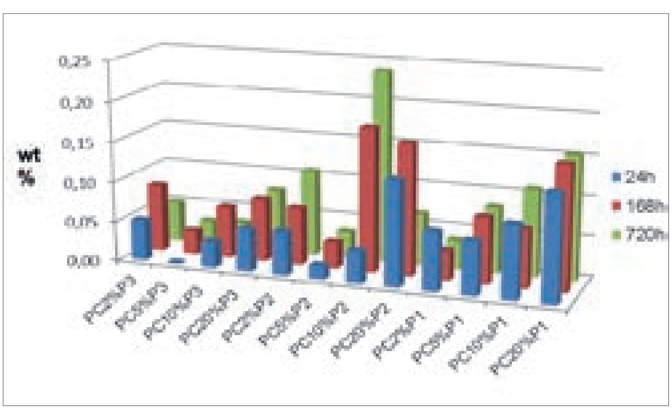
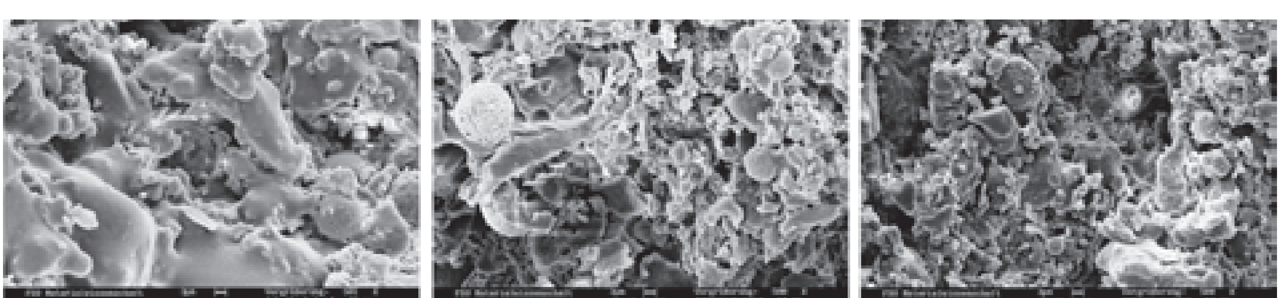


Figure 12. Leaching of the PC in neutral medium for samples sintered at 1100°C/1h

SEM micrographs of the fractured surfaces of PC20%P1, PC20%P2 and PC20%P3 sintered at 1100°C/1h are presented in Fig.13. Morphology of fly ash particles, especially spherical particles, is partially recognizable. The presence of the liquid phase is evident in the three microstructures, but at least present in the sample PC20%P3 which microstructure is less homogenous. According to Fucumoto and Kanda (2009) the sintering process of fly ash is performed by solid state sintering in the frame of the particles, while the sintering process of compos-



a) PC20%P1 b) PC20%P2 c) PC20%P3

Figure 13. SEM Micrograph of the fracture surface of PC samples sintered at 1100°C, x500, bar 3μm

ite material including clay is carried out by liquid and solid phase sintering due to lower melting point of clay. Therefore the surfaces around fly ash particles are surrounded by the melted clay which is solidified as binder around fly ash particles.

Conclusions

A wide range of porous composite (60 wt.% clay and 40 wt.% fly ash) ceramics have been produced by applying three pore creators i.e. two wood cutting (P1-Quercus and P2-Facus sylvatica) and P3 (C-powder).

The type and content of the pore creator as well as the sintering temperature influenced on the density, porosity, water absorption and mechanical properties of the obtained porous ceramics.

Relating to the sintering temperature, the optimal values i.e. the highest value for density and mechanical properties of the obtained porous ceramics were achieved at 1100^{o}C/1h. The type of the pore creator do not influenced significantly on the degree of porosity of the final porous ceramics, but the content of pore creator plays the main role on the properties of porous ceramics. Namely, the maximal bending strength (26 MPa) was achieved by using 2 wt% P3 (C-powder) and the porous ceramics has the density and porosity of 1.91g/cm^3 and 23%, respectively. The water absorption of the same porous ceramics was 8.4 %. Porous ceramics produced by using 20 wt.% of the above mentioned pore creators showed the lowest bending strength cca 5 MPa and the variation of the density and porosity were in the range from 1.22 to 1.32 g/cm^3 and 45 to 48 %, respectively. Generally, the presented investigation showed that by using the above mentioned pore creators and sintering at 1100^{o}C/1h a wide range of porous ceramics (porosity: 23-48 % and water absorption: 8.4- 25.5 %) can be produced based on utilization of clay and fly ash as raw materials. The obtained porous ceramics can find the potential application in construction industry.

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