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PARTIAL REPLACEMENT OF CLAY "ČAVKA" WITH FLY ASH "STANARI" IN BRICK PRODUCTION

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ABSTRACT

Fly ash class F from Thermal Power Plant "Stanari" was used in combination with highly plastic brick clay from Busovača to develop building bricks. The brick clay and fly ash were characterized for chemical and mineralogical composition, grain size distribution, and specific gravity. The fly ash has a higher proportion of larger particles and lower specific gravity than clay. The brick clay was mixed with 20, 40, and 60 wt. % of fly ash, formed in a prismatic shape, dried and then fired at 800°, 900°, and 1000 °C. The fired prisms were characterized for water absorption, apparent porosity, apparent density, and strength and compared with conventional clay prisms. The addition of fly ash of 20, 40, and 60% reduced the mass of the samples by 14, 23, and 31%, increased apparent porosity by 60, 100, and 160%, and reduced the compressive strength by 50, 80 and 90%, respectively. Conducted tests and comparative studies have shown that the fly ash "Stanari" can be used as an additive to the clay "Čavka" in the maximum amount of 20 % and the optimum firing temperature is 900 °C.

Keywords: clay; fly ash; brick; firing temperature; environment

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1. INTRODUCTION

Brick products belong to the oldest building materials. Clays, as the basic raw material for the production of brick products, are polymineral raw materials of sedimentary origin. During the production of brick products, huge amounts of natural raw materials are consumed, and this certainly has a negative impact on the environment. The technology for the production of brick products is constantly evolving, so in recent times, waste materials are used as a substitute for clay. These include fly ash, sawdust, paper residues, slag, rice husks, coffee grounds, cigarette butts, etc. [1-9].

Fly ash is produced as a by-product in thermal power plants during coal combustion and flue gas purification in electrostatic and bag filters. Disposal of fly ash in large fields pollutes the environment, and its use in the production of brick products, cement, concrete, geopolymers, and other building materials is very justified [10-12]. In the manufacture of bricks, fly ash can be used with clay as part replacement. The use of fly ash in the production of brick products is twofold regarding environmental protection. Firstly, the costs and consequences of fly ash disposal are reduced, and on the other hand, the consumption of raw materials for the production of bricks is reduced. Fly ashes can vary considerably in terms of chemical and mineral composition, color, as well as physical and morphological properties which depend upon many parameters such as coal quality, type of coal pulverization and combustion process, manner of ash collection, and disposal technique, etc. Concerning requirements of fly ash for brick production, the chemical composition should be similar to ordinary brick clays – the silica content should be over 40%, aluminum oxide not less than 15%, iron oxide not less than 5%, and sulfide and soluble sulfite content should be insignificant. However, not all the clays and fly ashes are suitable for brick making.

Many previously published studies [13-24] concluded that fly ash can be used as a partial replacement for clay in ceramic materials. Some of them [13,16,18,20-22,24] replaced clay with fly ash in a high ratio (up to 80 mas%). All research found that fly ash addition leads to a reduction in bulk density and an increase in porosity and water absorption. The positive effect of fly ash substitution was the reduction in mass [16,17,21,22]. However, when it comes to strength, the results differ significantly. Some of the researchers reported an increase in strength [13-17] while most noted a decrease in strength [18-23].

This paper contributes to solving the problem of large amounts of fly ash "Stanari" which has not found application in the cement or other industry. The purpose of this study is to evaluate the maximum quantity of incorporating this fly ash in clay bricks made of high plastic clay "Čavka" to produce a masonry acceptable brick. Despite the multitude of papers related to this topic, the utilization of fly ash in the ceramic industry still has not reached its full potential.

2. EXPERIMENTAL SETUP

2.1. Materials and sample preparation

The raw materials used in this investigation were clay from deposit "Čavka" situated near Busovača and fly ash from Thermal Power Plant "Stanari" near Doboj in Central Bosnia and Herzegovina. The clay was crushed, dried at 100 ± 5 °C, and sieved through the 1-mm sieve. Only for standard consistency and plasticity determination, clay was sieved through the 425-µm sieve. The fly ash was also dried at 100 ± 5 °C. The mixtures of clay with 0, 20, 40, and 60% fly ash were made. The mixtures were manually kneaded with tap water and the prisms with dimensions of 160x40x40 mm (Figure 1) were prepared by filling the metal mold. Marks and composition of samples, firing temperatures, and quantities of starting components are given in Table 1.

Figure 1. Metal mold for prisms (left) and making and marking prisms (right)

Table 1. Markings and composition of samples, firing temperatures, and quantities of starting components

Prism mark	Firing temperature (°C)	Fly ash content (%)	Water content (%)	Quantity		
				Clay (g)	Fly ash (g)	Water (ml)
I	800	0	19.78	1500	0	370
I_{20}		20	26.83	1200	300	550
I_{40}		40	30.69	840	560	620
I_{60}		60	35.64	520	780	720
II	900	0	19.78	1500	0	370
II_{20}		20	26.83	1200	300	550
II_{40}		40	30.69	840	560	620
II_{60}		60	35.64	520	780	720
III	1000	0	19.78	1500	0	370
III_{20}		20	26.83	1200	300	550
III_{40}		40	30.69	840	560	620
III_{60}		60	35.64	520	780	720

The samples were air-dried for 3 days, followed by drying in an oven for 24 hours at 100 ± 5 °C. The dried samples were then fired at 800°, 900°, and 1000 °C in an electric furnace according to the designed heating program shown in Figure 2. Firing temperatures and the heating program are similar to those of commercial clay bricks.

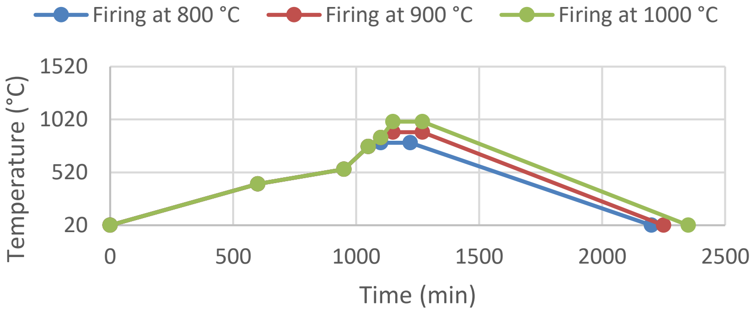

Figure 2. Heating program for the firing of samples

2.2. Methods of characterization

The chemical composition of clay was determined by the following procedures: loss of ignition (LOI) was determined by the gravimetric analysis after annealing at 900°C; SiO_2 content was also determined by the gravimetric method, and the contents of Al_2O_3, Fe_2O_3, TiO_2, CaO, MgO, K_2O, Na_2O, and MnO were determined after the acidic dissolution at the Atomic Absorption Spectrophotometer (Perkin Elmer 3100), while P_2O_5 was determined by photometry. Specific gravities of clay and fly ash were determined according to the standard EN 993-2.

Particle size distribution was determined by laser device Malvern Mastersizer 2000. The phase composition of the clay was investigated by X-ray diffraction analysis on a Shimadzu diffractometer XRD-6000 with Cu Kα radiation, with accelerating voltages of 40 kV and current 30 mA, in the range of angles 2-80° 2θ with a step 0.02° 2θ and a dwell time of 0.6 seconds. Differential thermal analysis (DTA) and thermogravimetric (TG) analysis were carried out on NETZSCH thermal analysis instrument STA 409 CD in a nitrogen atmosphere up to 1200 °C for clay and up to 1000 °C for fly ash, with a heating rate of 20 K/min. Microstructure property or fly ash was analyzed by using a scanning electron microscope MIRA 3 TESCAN.

Standard consistency was determined using the Vicat apparatus. By adding different amounts of water, the pastes of different consistencies were prepared and tested to determine the penetration depth of the Vicat needle. A clay paste has a standard consistency if the Vicat needle penetrates 5 minutes through a paste of 4 cm height [25]. Standard consistency was calculated by equation (1) and water content by equation (2).

$$\text{Standard consistency} = \frac{m_1 - m_2}{m_2} \cdot 100\ (\%) \quad (1)$$

$$\text{Water content} = \frac{m_1 - m_2}{m_1} \cdot 100 \quad (\%) \quad (2)$$

where is: m_1 - mass of wet clay (g), m_2 - mass of dry clay (g).

Plasticity was determined by the Pfefferkorn plasticity tester. The cylinders were made in metal mold (Ø = 30 mm, h = 40 mm) with different amounts of water and impact on the Pfefferkorn device, where the heights after impaction were read off. The diagram of the dependence of the Pfefferkorn height and water content of the clay pastes was drawn. The coefficient of plasticity is the water content (calculated according to equation (2)) required to achieve a 30% to the initial height of a test body under the action of a standard mass, i.e. height of 12.1 mm. The following types of clay are distinguished by the coefficient of plasticity:

13.8% - 16.7% → poorly plastic,
16.7% - 20.0% → moderately plastic,
20.0% – 23.1% → good plastic and
23.1% - 28.6% → highly plastic [26].

The behavior of the mixtures during drying and firing was monitored through mass loss and shrinkage. The equations for mass loss and shrinkage determinations are:

$$\text{Mass loss} = \frac{G_0 - G_1}{G_0} \cdot 100\ (\%) \quad (3)$$

$$\text{Shrinkage} = \frac{S_0 - S_1}{S_0} \cdot 100\ (\%) \quad (4)$$

where is: G_0 – mass of prism before thermal treatment (g), G_1 - mass of prism after thermal treatment (g), S_0 – dimension before thermal treatment (mm), S_1 - dimension after thermal treatment (mm).

To saturate the open pore space the prisms were soaked in water to half of their height for 24 hours. After that, water is added to completely cover the samples and thus left for another 24 hours. The following equations were used to determine water absorption (WA), apparent porosity (P_a), and apparent density (γ):

$$WA = \frac{m_3 - m_1}{m_1} \cdot 100 \quad (\%) \quad (5)$$

$$P_a = \frac{m_3 - m_1}{m_3 - m_2} \cdot 100 \quad (\%) \quad (6)$$

$$\gamma = \frac{m_1}{m_3 - m_2} \cdot \rho_w \quad \left(\frac{\text{g}}{\text{cm}^3}\right) \quad (7)$$

where: m_1 – mass of dry prism (g), m_2 – mass of saturated prism in water – hydrostatic weighing (g), m_3 – mass of saturated prism in the air (g), ρ_w – water density (g/cm^3).

Compressive and flexural strength were determined according to the standard EN 196-1. The images of crushed surfaces after testing flexural and compressive strength were taken by binocular light microscope OLYMPUS BX60M at a magnification of 200 times.

3. RESULTS AND DISCUSSION

3.1. Chemical composition

Table 2 presents the chemical composition and specific gravity of the clay "Čavka" and fly ash "Stanari" [27].

Table 2. Chemical composition and specific gravity of clay and fly ash

Component	Chemical composition (%)	
	Clay	Fly ash
SiO_2	54.1	54.81
Al_2O_3	19.1	20.50
Fe_2O_3	10.2	7.92
TiO_2	1.5	1.41
CaO	0.36	7.65
MgO	2.82	2.20
K_2O	3.51	0.825
Na_2O	1.15	0.51
MnO	0.14	-
P_2O_5	0.183	0.05
SO_3	-	2.11
LOI	6.57	3.97
Specific gravity (g/cm^3)	2.748	2.5163

It can be observed that the examined clay contained SiO_2 and Al_2O_3 in major quantities and is typical brick clay with higher content of Fe_2O_3 and a lower content of CaO. In accordance with the ASTM C618 standard fly ash "Stanari" belongs to class F because the total content of SiO_2, Al_2O_3, and Fe_2O_3 is over 70%, SO_3 is less than 5% and LOI is less than 6%. Its chemical composition is similar to clay and the silica content is over 40%, aluminum oxide not less than 15% and iron oxide not less than 5%. The clay has higher LOI and specific gravity than the fly ash.

3.2. Particle size distribution

The clay particle size distribution in Figure 3 shows that this is a bimodal distribution, with the largest number of particles being between 1 and 100 µm. The poor presence of the particles smaller than 2 µm and the biggest number of the clay particles between 5 and 50 µm indicates that the investigated clay contains small quantities of clay minerals. The fly ash particle size distribution in Figure 4 shows that the ash particles are also found mainly between 1 and 100 µm, but most of the particles are between 10 and 90 µm indicating that it has a higher proportion of larger particles than clay.

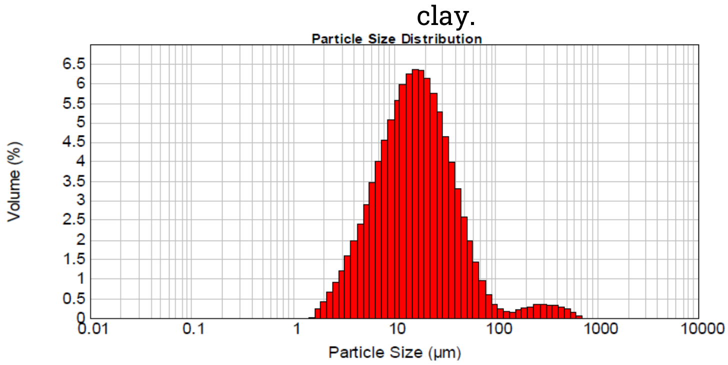

Figure 3. Particle size distribution of clay

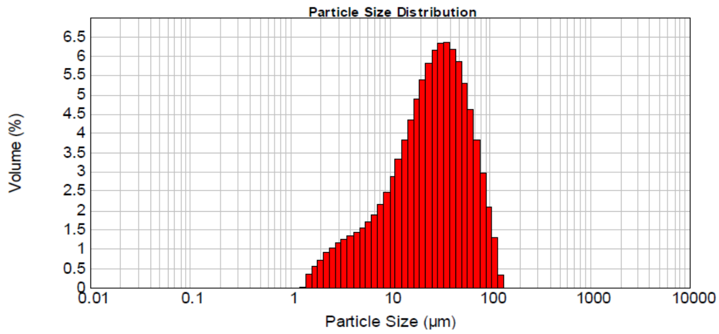

Figure 4. Particle size distribution of fly ash

3.3. Mineralogical composition

Figure 5 shows that clay "Čavka" consists of the following minerals: quartz, illite, kaolinite, chlorite, and anorthite. The mineral composition of fly ash "Stanari" is as follows: quartz, olivine, garnet, corundum, and apatite. The most common mineral in the analyzed sample is quartz [27].

Figure 6 shows the changes that occur by heating the clay sample. The first two peaks on the differential thermogravimetric curve correspond to mass loss due to evaporation of interlayer water in clay minerals and decomposition of iron hydroxide. The presence of illite, chlorite, and kaolinite is confirmed by mild endothermic change on the DTA curve and the main mass loss between 500° and 700 °C on the TG curve. A very weak exothermic peak at about 900 °C which belongs to the recrystallisation of spinel phase or mullite indicates that there is a low kaolinite content in clay. The total weight loss from 100° to 1000 °C of 6.77% agrees well with the LOI in the chemical analysis (6.57%).

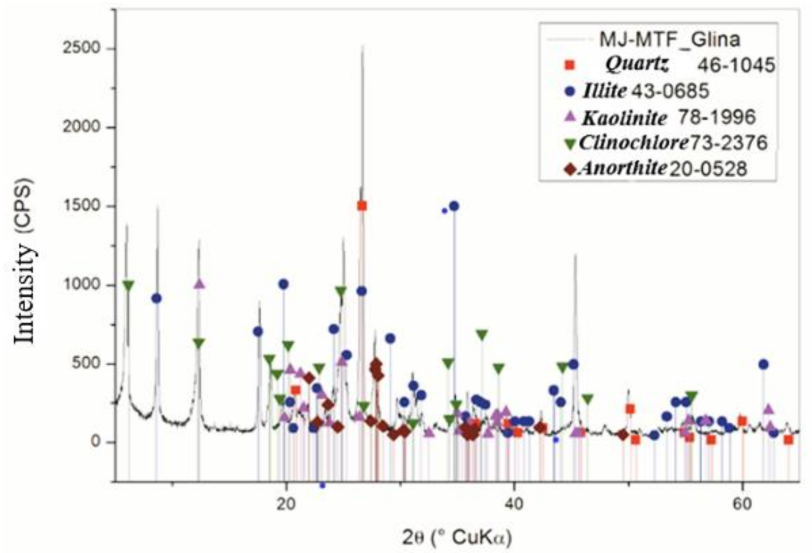

Figure 5. The phase composition of clay "Čavka"

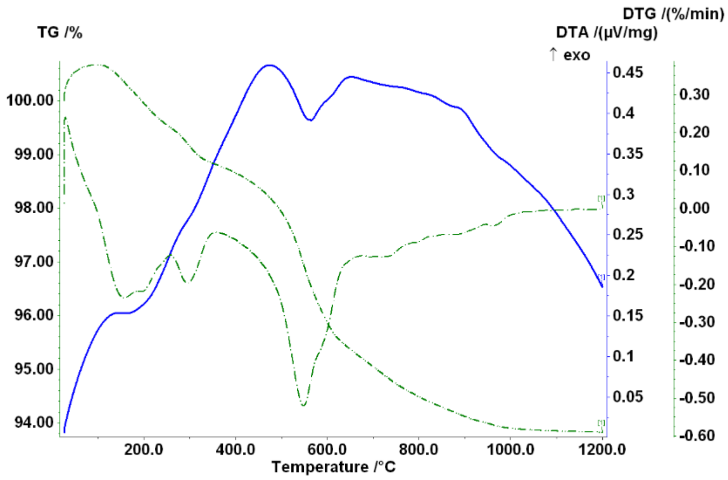

Figure 6. DTA/TG/DTG of clay "Čavka"

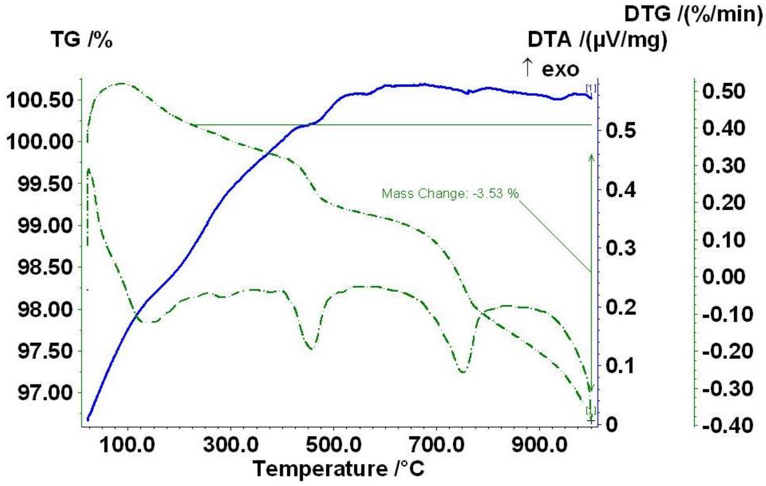

Figure 7. DTA/TG/DTG of fly ash "Stanari"

In Figure 7 it can be seen that during the heat treatment of the fly ash there are three steps of mass loss. The first is the loss of water adsorbed on the surface of the particles. The second is the decomposition of the residual fuel at temperatures between 400° and 500 °C and the third is the apatite [28] or garnet [29] decomposition. The mass loss from 200° to 1000 °C is 3.53% which also agrees well with the LOI in the chemical analysis (3.97%). It should be borne in mind that the LOI is determined by heating to 900 °C and dwelling for 2 hours, while loss of mass in TG analysis is without dwelling at the final temperature.

3.4. SEM analysis of fly ash

The SEM images in Figure 8 show the availability of irregularly shaped particles and aggregates in fly ash. Some of these grains are very porous. This shape of the particles, with a much larger specific surface area compared to most ashes that have a lot of spherical particles, with additional porosity of some particles, explains the phenomenon of increasing the amount of water in the preparation of mixtures with fly ash (Table 1). Several other papers have reported an increase in the amount of water required to form mixtures of clay with fly ash [16,23].

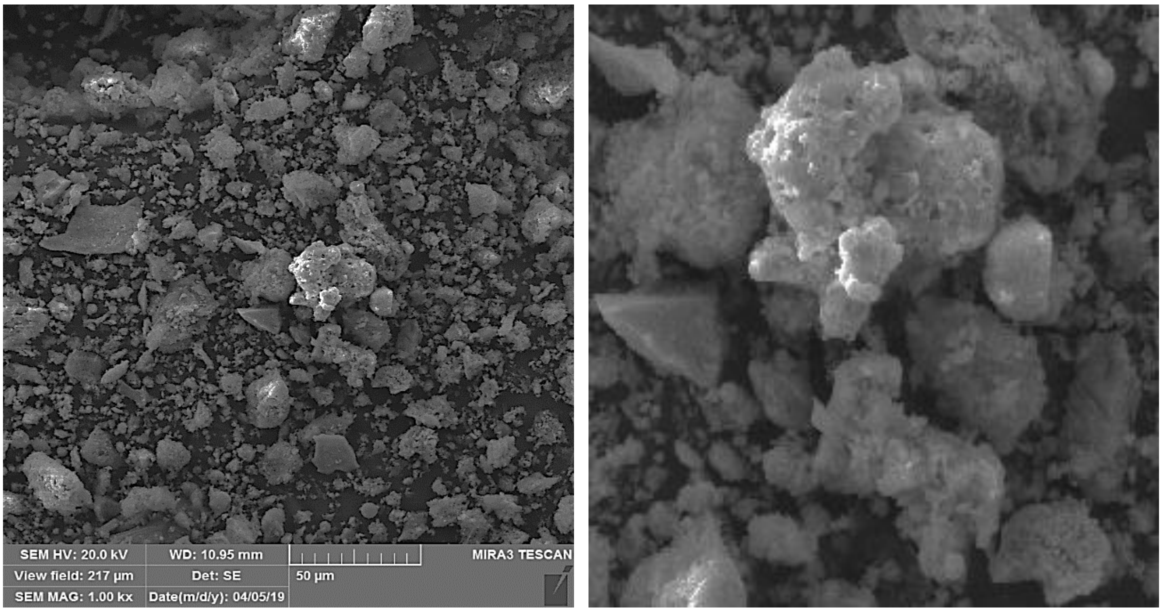

Figure 8. SEM images of fly ash (right image is enlarged part of the left image with porous gains)

3.5. Standard consistency and plasticity

The standard consistency for brick clays is usually 15 to 35%. The standard consistency of the tested clay is 24.18% which is within the limits for brick clays. The coefficient of plasticity of the clay "Čavka" is 26.95%, which means that the tested clay belongs to the highly plastic clays.

3.6. Behavior during drying and firing

Table 4 shows the results of the behavior of mixtures during drying. As the ash content increases, the mass loss on drying increases due to the evaporation of water added during molding (Table 1). The porous and absorbent nature of fly ash stabilizes the drying behavior of the clay by reducing the shrinkage on drying, despite the increase in water demand during molding. In clay without additives, the distance between the particles is reduced as water molecules leave the clay during the drying process so that the clay body undergoes significant shrinkage. On the other hand, clay mixtures containing fly ash lose water during drying, but a significant portion of that evaporating water is contained in the pores of the fly ash particles themselves. The addition of ash in the amount of 20, 40, and 60% reduces the shrinkage by 7, 40, and 45%, respectively. Reduction of shrinkage is a very favorable effect because it reduces the sensitivity of the mixture to drying and the possibility of cracking.

Table 4. Mass loss and shrinkage on drying

Fly ash content (%)	Mass loss (%)	Shrinkage (%)
0	20.56	4.09
20	27.37	3.82
40	31.35	2.40
60	35.69	2.24

At the same time, it allows to increase the drying rate, reduce the drying time and save energy.

Table 5 shows the results of mass loss and shrinkage during firing. The effect of fly ash addition to the clay indicates that the mass loss of all samples mixed with different concentrations of fly ash is decreasing with the increasing amount of fly ash because the ash has a smaller mass loss during firing than clay (LOI in Table 2). As the firing temperature of the samples increases, a gradual increase in mass loss is noticeable. With increasing ash content, the shrinkage is less, which is the same case as with drying. As the firing temperature increases, the shrinkage due to the sintering process increases. The total shrinkage during drying and firing is also significantly less in samples with fly ash compared to clay without the addition of ash.

Table 5. Mass loss and shrinkage on firing

Prism mark	Firing temperature (°C)	Mass loss (%)	Shrinkage (%)	Total shrinkage (%)
I	800	5.61	3.50	7.59
I_{20}		5.12	2.01	5.83
I_{40}		4.82	1.42	3.82
I_{60}		4.44	1.23	3.47
II	900	5.92	4.50	8.59
II_{20}		5.43	2.81	6.63
II_{40}		5.00	2.11	4.51
II_{60}		4.60	1.88	4.12
III	1000	6.00	5.50	9.59
III_{20}		5.8	3.02	6.84
III_{40}		5.84	2.76	5.16
III_{60}		5.59	2.22	4.46

Figure 9. The appearance of prisms after firing (left – firing at 800 °C, middle – firing at 900 °C, right – firing at 1000 °C)

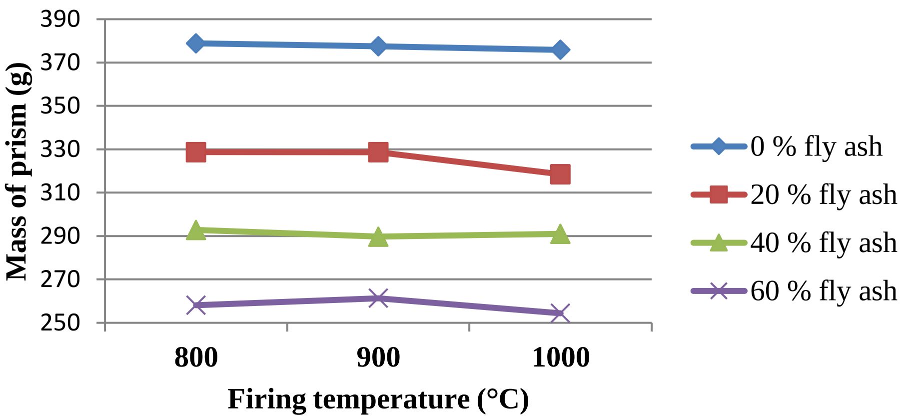

Figure 10. The masses of prisms after firing

Figure 9 shows the appearance of prisms after firing, where even with the bare eye can see that the prisms with a higher addition of ash are larger, or that they have a smaller shrinkage. A reduced reddishness in color is noticed in prisms containing fly ash as compared to prisms without fly ash. The addition of ash in the amount of 20, 40, and 60% reduces the mass of prisms by 14, 23, and 31%, respectively (Figure 10) because the specific gravity of the ash is less than the clay specific gravity. Further, fly ash usually contains unburned coal particles, but they are burned during the firing process and therefore, the product is lighter due to the formation of new pores. Some other authors also reported a reduction in the mass of the samples with the addition of ash [16,18,22,23]. The reduction in the mass of bricks results in a great deal of savings to the consumer that results from the increased number of units during transportation and load reduction on structural elements.

Table 6. Properties of fired samples

Prism mark	Firing temperature (°C)	Water absorption (%)	Apparent porosity (%)	Apparent density (g/cm^3)
I	800	18.0	31.2	1.73
I_{20}		29.4	42.4	1.44
I_{40}		37.4	48.4	1.26
I_{60}		47.0	53.6	1.13
II	900	15.24	27.4	1.79
II_{20}		26.7	39.9	1.48
II_{40}		37.5	48.0	1.28
II_{60}		46.7	53.3	1.14
III	1000	8.34	16.8	2.00
III_{20}		23.7	36.9	1.56
III_{40}		35.7	46.7	1.30
III_{60}		45.5	52.5	1.15

3.7. Properties of fired products

Table 6 shows the results for water absorption, apparent porosity, and apparent density of the mixtures. There is a close relationship between porosity and water absorption and apparent density of bricks. Apparent density decreases and apparent porosity and water absorption increase with increasing ash content for these reasons:

- lower specific density of fly ash compared to clay,
- a higher volume of prisms with fly ash due to less shrinkage during drying and firing,
- a smaller mass of prisms with flying ash,
- the porosity of fly ash itself, and

- additional porosity due to combustion of unburned coal.

As the firing temperature increases, the density of prisms increases, and porosity and water absorption decrease due to sintering processes which include vitrification. The vitreous phase penetrates pores and closes them. A lower density is generally reflected in better thermal insulation properties of the building ceramics. Fly ash addition increases significantly the porosity and water absorption of prisms, much more than in other studied cases. The addition of ash in the amount of 20, 40, and 60% to clay fired at 800 °C increases the water absorption by about 60, 100, and 160%, respectively. This influence is even more pronounced in prisms fired at 900° and 1000 °C. Such a notable effect of ash on porosity and water absorption has been reported only in [17,30]. The peculiarity of this influence of "Stanari" fly ash is in the morphology of its particles (Figure 8). The maximum porosity reported in other papers is 40% and water absorption is 31% [15]. Such characteristics in this study show only prisms with 20% fly ash.

The test results of flexural and compressive strength of fired samples are given in Figure 11 and Figure 12, respectively. The strength of prisms decreases significantly as the amount of fly ash increases. This inverse relationship between compressive and flexural strength and fly ash content may be attributed to the rising volume of pores. It can also be observed that a significant increase in strength with increasing firing temperature is only in clay with 0 and 20% ash, while in clay with 40 and 60% ash firing temperature has a small effect. The reduction in compressive strength for the addition of ash in the amount of 20, 40 and 60% is about 50, 80, and 90%, respectively. Significant reductions in strength with the addition of fly ash to clays were also recorded in the papers [18,20-23].

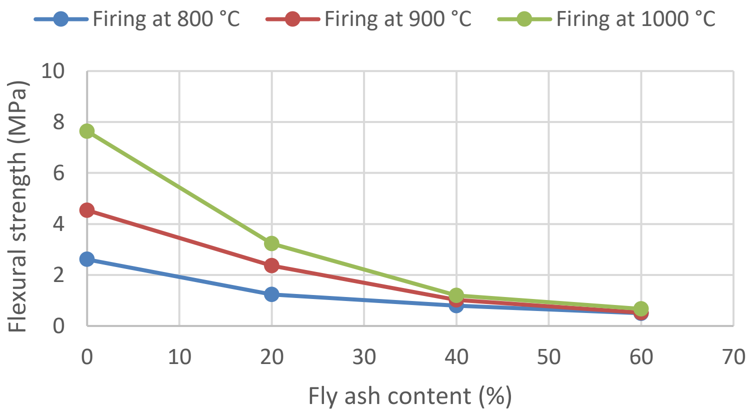

Figure 11. Flexural strength of prisms

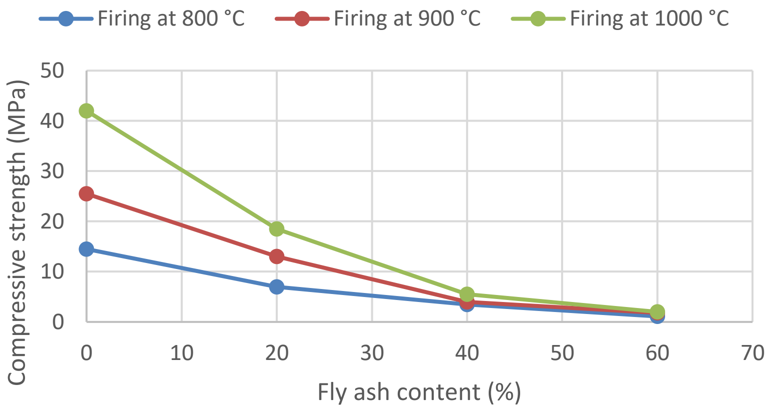

Figure 12. Compressive strength of prisms

Domestic regulations allow the use of bricks with a minimum compressive strength of 1.5 MPa for lightweight bricks, while for solid bricks this limit is 7.5 MPa. Prisms with 20% fly ash fired at 800°, 900°, and 1000°C have compressive strengths of 7, 13, and 18.5 MPa, respectively. However, prisms with 40% fly ash also have higher compressive strengths than those shown in [13,14], but they do not have satisfactory water absorption. Prisms with 20% fly ash fired at 900 °C have satisfactory strength and water absorption. From the microscopic image in Figure 13, it is clear that the structure of the sample with 60% fly ash is more homogeneous, but with significantly more pores compared to the clay sample without the addition of fly ash.

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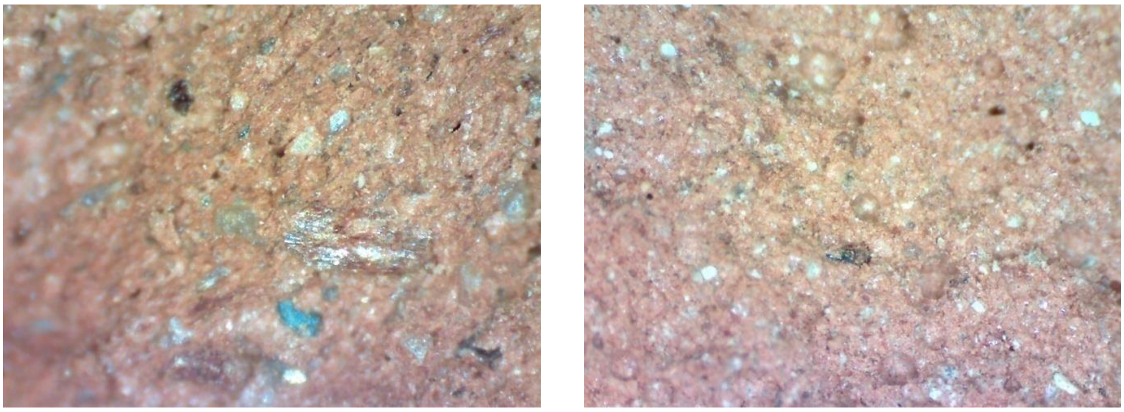

Figure 13. Microphotograph of clay (left) and clay with 60% of fly ash (right) fired at 900 °C

4. CONCLUSION

Fly ash has a very significant effect on the properties of the tested clay. It has a similar chemical composition to the clay but lower LOI and specific gravity. Its particles are slightly coarser than particles of clay. The particles of fly ash are of irregular shape with a larger surface area and a lot of porosity. For that reason, the mixes with fly ash demand higher quantities of water for forming. This further results in higher mass loss during drying. The favorable effect is a reduction in shrinkage on drying because particles of fly ash lose water without reducing distances between particles. Reduction of shrinkage reduces the sensitivity of the mixture to drying and the possibility of cracking, increases the drying rate, reduces the drying time, and saves energy.

After the firing, the mass of samples with fly ash in the amount of 20, 40, and 60% are reduced by 14, 23, and 31%, respectively. This is also a favorable effect that results in savings concerning an increased number of units during transportation and load reduction on structural elements.

Low specific density, high porosity, and low shrinkage of fly ash are the main reasons for apparent density decrease and apparent porosity and water absorption increase with increasing fly ash content in specimens. Samples fired at 800 °C have increased water absorption by about 60, 100, and 160% with the addition of fly ash in the amount of 20, 40, and 60%, respectively. This influence is even more pronounced in samples fired at 900° and 1000 °C. The increase in the firing temperature results in apparent density increase and porosity and water absorption decrease due to vitrification in sintering processes. However, only prisms with 20% fly ash have porosity and water absorption that do not significantly exceed the results obtained in other studies.

It was observed that an increase in fly ash content leads to a reduction both in flexural and compressive strength. The reduction in compressive strength for the addition of ash in the amount of 20, 40 and 60% is about 50, 80, and 90%, respectively. The effect of fly ash content on strength is more significant than the effect of firing temperature. Samples with 20 and 40% fly ash have satisfactory strengths, but only samples with 20% fly ash also satisfy water absorption.

Based on the test results, it can be recommended that the tested fly ash can be used in the maximum amount of 20% and the optimum firing temperature is 900 °C.
The use of fly ash as an additive to brick clays improves certain properties of the samples like thermal insulation, reduces shrinkage and mass, etc. It is worth mentioning that fly ash belongs to the group of waste materials, so its utilization in larger quantities would be beneficial to resolving the environmental problem of its disposal and preserving the natural clay resources.

Conflicts of Interest
The authors declare no conflict of interest.

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