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Effects of cigarette butts addition on thermal conductivity and ceramic properties of brick clay

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Abstract
The influence of the addition of cigarette butts, as dangerous waste material, to the properties of the brick clay "Čavka" near Busovača was studied. The cigarette butts were added to the clay in amounts of 5 mass% and 10 mass%. Testing of the samples included testing of consistency, plasticity, Atterberg plastic and liquid limits, drying and firing behavior and refractoriness. Also, apparent density, apparent porosity, water absorption, flexural and compressive strength were investigated on the samples fired at different temperatures. Thermal conductivity was tested on sample fired at 1173 K. Addition of cigarette butts improved insulation characteristics and drying sensitivity, while other properties remain within the required limits for brick industry.

Keywords: Clay; Firing; Thermal conductivity; Cigarette butts.

1. Introduction

Clay bricks are one of the oldest building materials and nowadays still most demanding masonry units because of their durability, fire resistance, strength, beauty, insulating and many other properties. The main raw material for their production is clay. However, reserves of the clay are limited, and it is necessary to find the additives that could partially replace the main raw material. Part of the raw material can be replaced by waste material which would lead to savings natural raw materials and solving problems related to disposal of certain types of the waste. Many attempts and different materials were used to utilize some wastes in the fired clay bricks like saw dust, paper pulp, coal dust, rice husks, seed-shell, waste glass, fly ash, sludge [1-4]. Some of these additives increase thermal insulation capacity by forming additional porosity beyond original porosity of the fired clay material.

One of the most common wastes that can be seen today are cigarette butts. It is estimated that every year about $4.5 \cdot 10^{12}$ cigarettes are littered in the world [5]. As the cigarette butts contain variety of chemicals, many of which are known to be carcinogenic, they present a serious environmental problem. One of the solutions resolving this problem is to incorporate them in the clay bricks.

Due to the environmental regulations, the bricks with high insulation capacity are more demanded than the traditional bricks. One way to increase the insulation capacity of the bricks is to generate and increase the porosity [6]. One of the most conventional ways to increase the porosity and reduce the thermal conductivity in ceramic materials is to modify their microstructure by incorporating lightening, pore-forming, organic combustible additives into the clay matrix. The combustible material burns out during the firing and leaves a large fraction of the pores within the fired body. The presence of the pores in the materials helps to reduce the thermal conductivity and increases therefore its insulation and produced bricks are lighter than traditional [7]. The thermal conductivity not only depends on total porosity, but also on pore size and shape, chemical and mineralogical composition [8]. The porous character of light bricks will increase the quality of structures in terms of heat insulation, thereby reducing heating costs in turn affecting environment positively [9].

Organic waste as energetically enriched material easily combusts during the firing process. With their combustion they contribute to the increasement of the temperature in the furnace. This allows economical use of the energy needed for the firing [7] and shortens the firing time [10].

Properties of the bricks are affected as a result of physical, chemical and mineralogical alteration. In this paper, the effect of cigarette butts addition to the clay was investigated.

2. Experimental

2.1 Materials

The raw materials used in this investigation were clay from deposit "Čavka" situated in the Central Bosnia and Herzegovina near Busovača and cigarette butts from the household. About 100 kg of the clay, which has undergone all stages of the preparation, was sampled at the Factory "Termo Ziegel Nigma Busovača". The clay was crushed in the Laboratory, sieved through the sieve 1 mm and dried at 373 ± 5 K. The cigarette butts were dried at 283 ± 5 K and grinded in electric kitchen chopper.

2.2 Methods of characterization

Standard methods (JUS B.B8.070, JUS B.H8.346, JUS B.H8.320) and atomic absorption spectroscopy (Perkin Elmer 3100) were used for determination of the chemical composition of the clay. DTA and TG analyses were carried out to investigate the clay, cigarette butts and mixtures behavior during the thermal treatment. It was performed on Netzsch STA 409 CD in nitrogen atmosphere up to 1473 K with heating rate 20 K/min. Particle size distribution was determined by laser device Malvern Mastersizer 2000. Phase composition of the clay was carried out with 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.

Three types of mixtures were prepared:

- Clay without additives,
- Mixture of clay and 5 mass% of cigarette butts,
- Mixture of clay and 10 mass% of cigarette butts.

The effect of cigarette butts addition on the standard consistency, plasticity and Atterberg plastic and liquid limits was investigated. Standard consistency was determined using Vicat apparatus. The clay is milled so that it passes through sieve 425 μm. 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 Vicat needle penetrates 5 minutes through a paste of 4 cm height [11].

Plasticity was determined by the Pfefferkorn plasticity tester. Cylinders were made in metal mould (Ø = 30 mm, h = 40 mm) with different amounts of water and impacted on the Pfefferkorn device, where the heights after impaction was 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 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 [12].

Standard consistency was calculated by the equation (1) and water content by the equation (2).

$$Standard\ consistency = \frac{m_1 - m_2}{m_2} * 100 \quad [\%] \tag{1}$$

$$Water\ content = \frac{m_1 - m_2}{m_1} * 100 \quad [\%] \tag{2}$$

where: m_1 - mass of wet clay [g], m_2 - mass of dry clay [g].
The Atterberg plastic limit (PL) is defined as minimum water content (equation 1) at which the clay paste can be rolled into the cylinder of thickness 3.2 mm and length 70 mm on the flat non-porous surface. Cylinder was made from plastic paste, after which it was being reformed until the gapes show up due to evaporation of water. For Atterberg liquid limit (LL) determination fall cone method was used with a standard 30° - 80 g cone penetrating 20 mm. The plasticity index (I_P) is the measure of the plasticity, i.e. water quantity needed for the paste to change from semisolid to liquid state. This index represents the difference between the liquid limit and plastic limit [13]:

$$I_p = LL - PL \quad [\%] \tag{3}$$

where: LL – liquid limit [%], PL – plastic limit [%].
For testing the clay behavior during drying and firing and the properties of fired samples, handmade tiles, prisms and plates were prepared by using appropriate mould. The dimensions of the tiles were 80x40x14 mm. Diagonals were drawn on the prepared tiles and then a circle with diameter 50 mm with a center at the intersection of the diagonals was drawn. The cross-section of the circle with the diagonals gave the benchmark points which were used to determine drying and firing shrinkage. The tiles were also used to test the mass loss and shrinkage during drying and firing and the water absorption, apparent porosity and apparent density. The prisms with dimensions 160x40x40 mm were used for the testing of compressive and flexural strength at the compression and flexural testing machine. Thermal conductivity was tested on the samples with dimensions 50x50x20 mm (cut from plates 300x300x30 mm) by device Hot Disk TPS 2200 (the product of the Hot Disc AB Company from Gothenburg in Sweden). The samples were air-dried for 3 days, then one day at 303 K and one day at 373 K in a drying oven. After drying, all the samples (tiles, prisms and plates) were fired according to the regime shown in Figure 1.

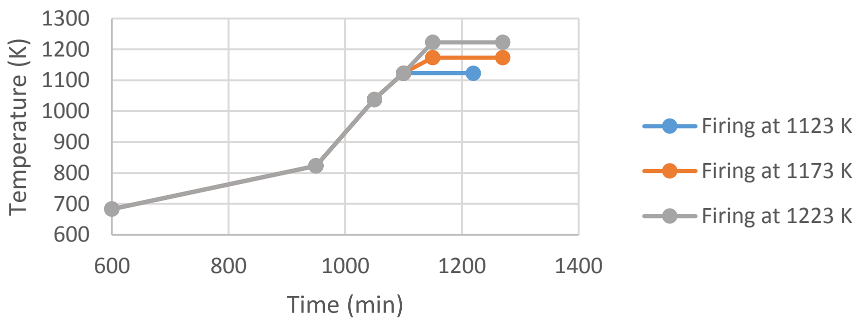


Figure 1. Regime for the firing of samples.

The equations for mass loss and shrinkage determinations are:

$$Mass\ loss = \frac{G_0 - G_1}{G_0} * 100\ [\%] \tag{4}$$

$$Shrinkage = \frac{S_0 - S_1}{S_0} * 100\ [\%] \tag{5}$$

where: G_0 - mass of tile before thermal treatment [g], G_1 - mass of tile after thermal treatment [g], S_0 - distance between benchmark points before thermal treatment [mm], S_1 - distance between benchmark points after thermal treatment [mm].
To saturate the pore space the tiles were soaked in water to the 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):

$$WA = \frac{m_3 - m_1}{m_1} * 100 \quad [\%] \tag{6}$$

$$P_a = \frac{m_3 - m_1}{m_3 - m_2} * 100 \quad [\%] \tag{7}$$

$$\gamma = \frac{m_1}{m_3 - m_2} * \rho_w \quad [g/cm^3] \tag{8}$$

where: m_1 – mass of dry tile [g], m_2 – mass of saturated tile in water – hydrostatic weighing [g], m_3 – mass of saturated tile on air [g], ρ_w – water density [g/cm^3].
Refractoriness of the fired samples was investigated in Criptol furnace according to the BAS EN 993-2:200. The images of crushed surfaces after testing of compressive and flexural strength were taken by the binocular optical microscope OLYMPUS BX60M at magnification of 12.5 times. Microstructures were examined by scanning electron microscopy (SEM) JEOL, JSM – 5800.

3. Results and discusion

Table 1 presents the chemical composition of the clay "Čavka". According to these results it can be observed that investigated clay is typical brick clay with higher content of Fe_2O_3. The presence of the quartz, illite, kaolinite, clinochlore and anorthite was noticed on X-ray diffraction analysis in Figure 2. This composition is usual for the clays in the area of central Bosnia [14]. The particle size distribution of the clay sieved through the sieve 0.5 mm is shown in Figure 3 and it indicates that the investigated clay contains small quantities of clay minerals. This could be due to the poor presence of the particles smaller than 2 µm, but the biggest number of the clay particles is between 5 and 50 µm.

Table 1. Chemical composition of clay.

Component	Chemical composition [mass%]
SiO_2	54.1
Al_2O_3	19.1
Fe_2O_3	10.2
TiO_2	1.5
CaO	0.36
MgO	2.82
K_2O	3.51
Na_2O	1.15
MnO	0.14
P_2O_5	0.183
LOI	6.57

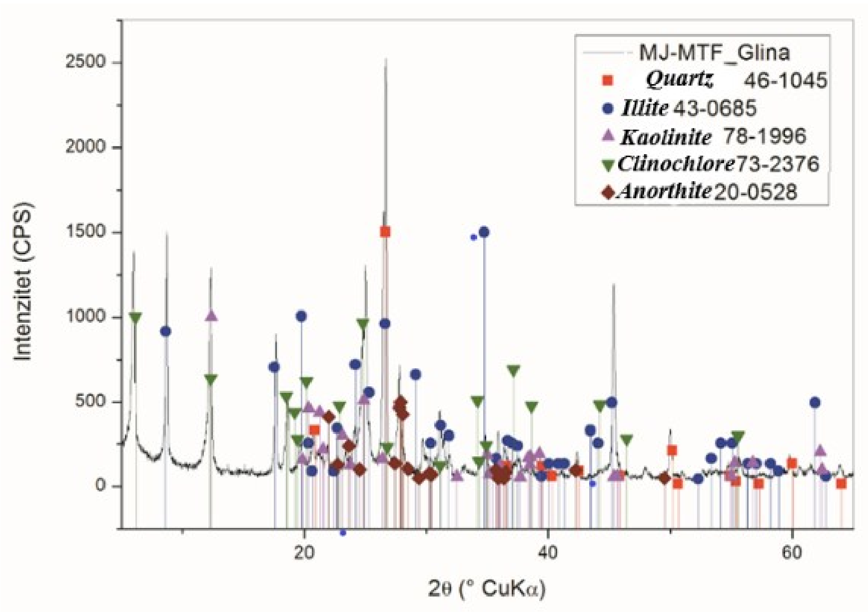


Figure 2. Phase composition of the clay "Čavka".

d(0.1): 5.165 um d(0.5): 16.126 um d(0.9): 48.558 um

Mesh No	Aperture µm	Volume In %	Vol Below %
8	2000	0.00	100.00
10	1700	0.00	100.00
12	1400	0.00	100.00
14	1180	0.00	100.00
16	1000	0.00	100.00
18	850	0.00	100.00
22	710	0.10	100.00
25	600	0.25	99.90
30	500		99.65

Mesh No	Aperture µm	Volume In %	Vol Below %
30	500	0.32	99.65
36	425	0.40	99.33
44	355	0.41	98.93
52	300	0.46	98.52
60	250	0.37	98.06
72	212	0.29	97.69
85	180	0.23	97.40
100	150	0.19	97.16
120	125		96.97

Mesh No	Aperture µm	Volume In %	Vol Below %
120	125	0.24	96.97
150	106	0.44	96.72
170	90	0.91	96.29
200	75	1.51	95.38
240	63	2.32	93.87
300	53	3.10	91.54
350	45	4.21	88.44
400	38		84.24

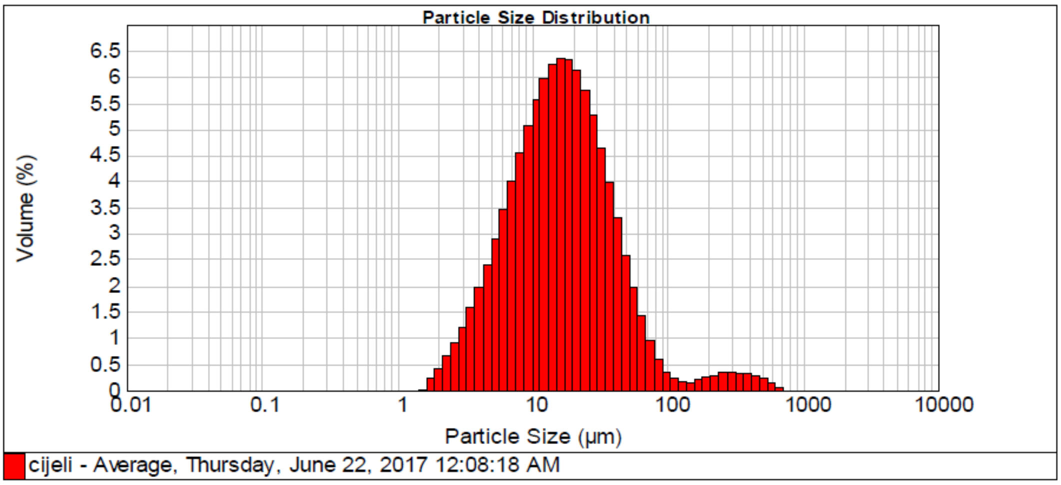


Figure 3. Particle size distribution of the clay "Čavka".

Figure 4 shown the DTA/TG curves of clay, cigarette butts, clay with 5 mass% cigarette butts and clay with 10 mass% cigarette butts. The DTA curve of cigarette butts show board exothermic changes corresponding to decomposition of organic components. The decomposition occurs mostly below 673 K and total mass loss was over 80 mass%. The presence of kaolinite and illite in the clay, identified by XRD analysis, is confirmed by mild endothermic change on DTA curve of clay and mass loss between 773 and 873 K on TG curve. At this temperature interval, the transition from low-temperature quartz to high-temperature modification occurs. The total mass loss of clay is below 5 mass%, while loss of ignition is over 6 mass%. Thermogravimetry was made in a nitrogen atmosphere, so there was no complete combustion of the substance as in the chemical analysis. The TG curves of the mixtures confirm that even in the mixtures, the cigarette butts are almost completely decomposed.

The results for the standard consistency, Pfefferkorn plasticity and Atterberg limits are given in Table 2. Standard consistency, Pfefferkorn plasticity and Atterberg limits increase with increasing of the cigarette butts content. This means that the addition of the cigarette butts requires a larger amount of water to obtain the desired clay workability. Water content, density of the particles and mass loss on drying effect on the porosity of the clay brick. The higher water absorption of the additives, the larger amount of water to obtain the desired clay workability is needed. Behavior of the clay particles and additives on drying is different, and as water leaves the system during the drying, the distance between the particles is reduced. On the other hand, cigarette butts don't show significant volume changes during the drying [15]. This interesting phenomenon of decreasing the shrinkage by increasing the percentage of cigarette butts is very favorable as it reduces clay sensitivity on the drying process and possibility of cracking. On this way it is possible to reduce drying time and save energy.

In Table 3 are presented the results of mass loss and shrinkage on drying and in Table 4 are shown the results of mass loss and shrinkage on firing. With increasing the cigarette butts content mass loss during the drying process increases which is expected, because the greater is the amount of water added to the mixes, so it evaporates when drying. A slight increase in mass loss was observed in all clay mixtures with the increase of the firing temperature. Increasing the amount of cigarette butts also increases the mass loss, which is expected, because additives mainly transform to the gaseous component in the firing process. With the increase of the firing temperature, an increase in shrinkage occurs in all samples, especially at 1223 K, indicating that significant sintering of the samples occurs at this temperature (third phase of firing when silicate formation and solid-state reaction occur).

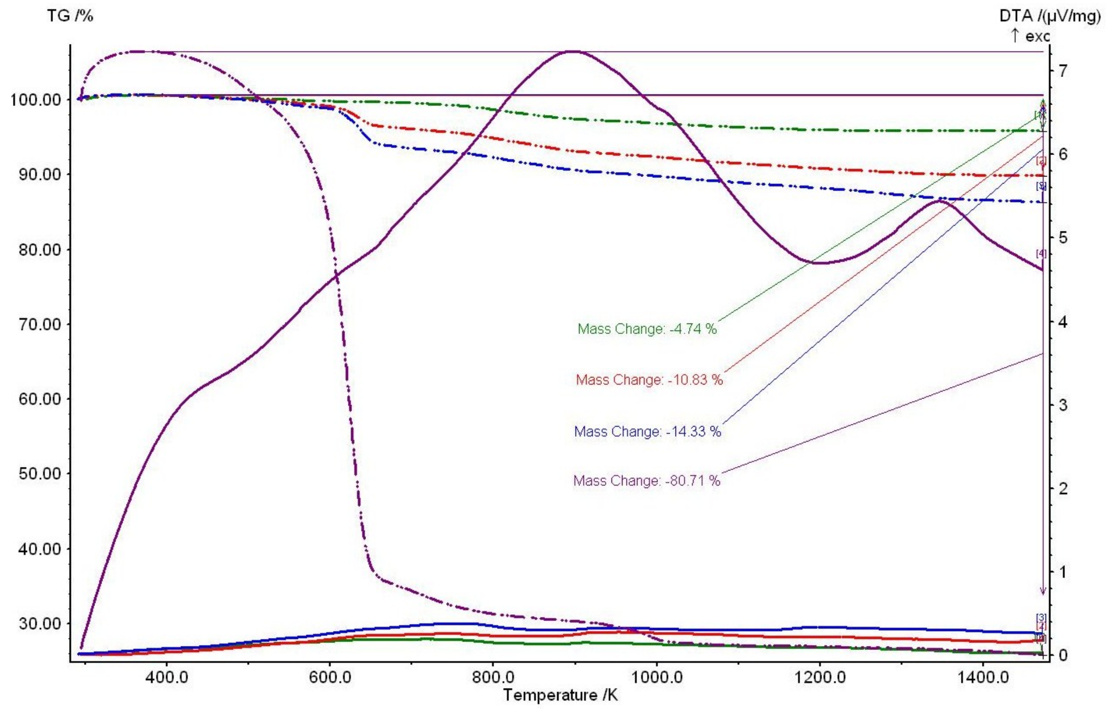


Figure 4. DTA/TG curves for clay (green), cigarette butts (purple), clay with 5 mass% cigarette butts (red) and clay with 10 mass% cigarette butts (blue).

Table 2. Standard consistency, plasticity and Atterberg limits.

Cigarette butts content [%]	Standard consistency [%]	Pfefferkorn plasticity	LL [%]	PL [%]	I_P [%]
0	25.7	25.51	32.45	20.3	12.13
5	30.4	33.0	39.4	24.0	15.4
10	34.0	40.78	45.4	29.0	16.4

Table 3. Mass loss and shrinkage on drying.

Cigarette butts content [%]	Mass loss [%]	Shrinkage [%]
0	20.60	4.73
5	23.73	3.82
10	26.34	3.32

Table 4. Mass loss and shrinkage on firing.

Cigarette butts content [%]	Temperature [K]	Mass loss [%]	Shrinkage [%]
0	1123	6.28	0.42
	1173	6.33	1.62
	1223	7.08	4.04
5	1123	10.85	0.21
	1173	10.87	0.86
	1223	10.94	2.57
10	1123	14.51	0.15
	1173	15.01	0.62
	1223	15.05	2.18

Addition of cigarette butts to the clay has positive effect on insulation properties. From Figure 5 it can be seen that the thermal conductivity decreases with increasing of cigarette butts content. That can be explained by the porosity increasing with increasing the cigarette butts content. Figure 6 shows that the addition of the cigarette butts slightly decrease refractoriness, in fact these type of additives does not have significant effect to the fire resistance.

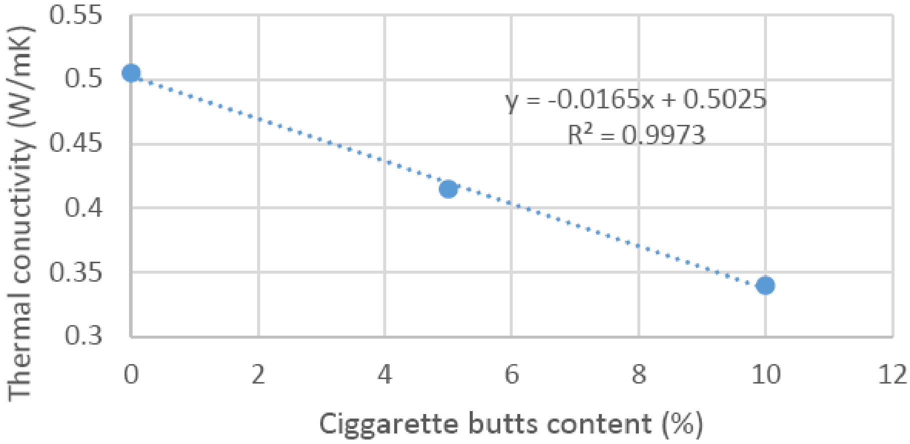


Figure 5. The dependence of the thermal conductivity on butts content.

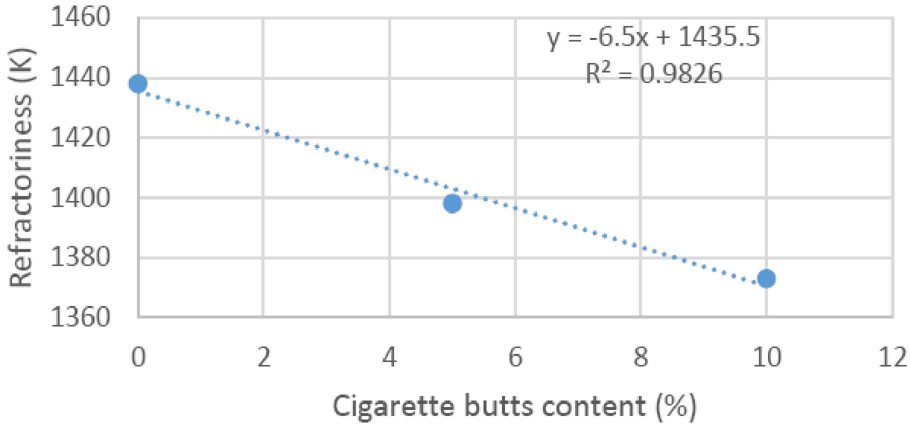


Figure 6. The dependence of the refractoriness on butts content.

Table 5 presents effect of change of temperature and cigarette butts content on fired clay samples properties. Apparent porosity and water absorption also increase with increasing of cigarette butts content. Cigarette butts combustion leaves pores which results in apparent density decreasing. Specimens with lower apparent density are lighter that can be consider as positive effect for the transport aspect, handling and installation of bricks.

Due to increasing of porosity, flexural and compressive strength decrease, although compressive strength is still acceptable according the standard JUS B.D1.015. Compressive strength for brick is in range 2 to 20 MPa and flexural strength is not standardized [9].

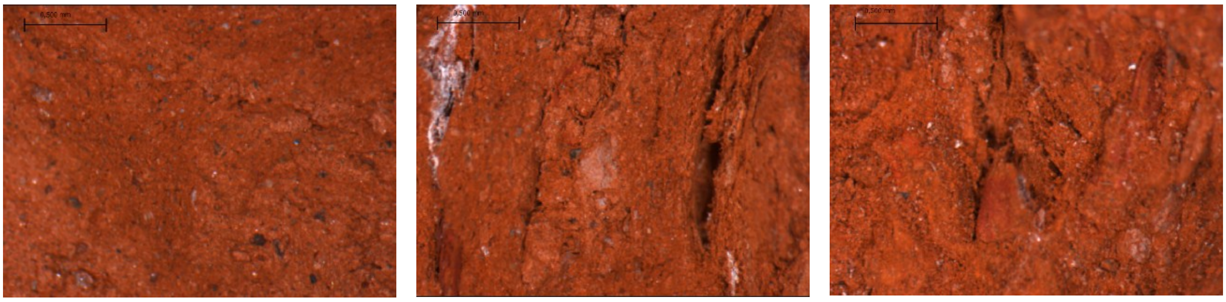
Photographs from optical microscope of samples fired at 1173 K are shown in Figure 7. It can be seen from the micrographs that the addition of cigarette butts changes structure considerably. In the samples with cigarette butts larger, elongated or eyelash pores and significant inhomogeneity of the structure are observed, which is why the strength of these samples is smaller in comparison to the samples without the addition.

SEM micrographs of samples fired at 1173 K are shown in Figure 8. In the samples with the addition of the cigarette butts there is considerable inhomogeneity of the structure and in certain places where larger pieces of cigarette butts are observed, the fibrous structure is seen as a result of the well-preserved original morphology of the butts.

Analyzing the samples fired at 1123 K were obtained favorable results in terms of apparent density and apparent porosity, and since the insulation material is porous, it should be as high as possible. In examining the samples fired at 1223 K there was a large sample shrinkage, porosity reduction and density increase. From presented results it can be concluded that the optimal temperature of firing is between 1123 K and 1173 K.

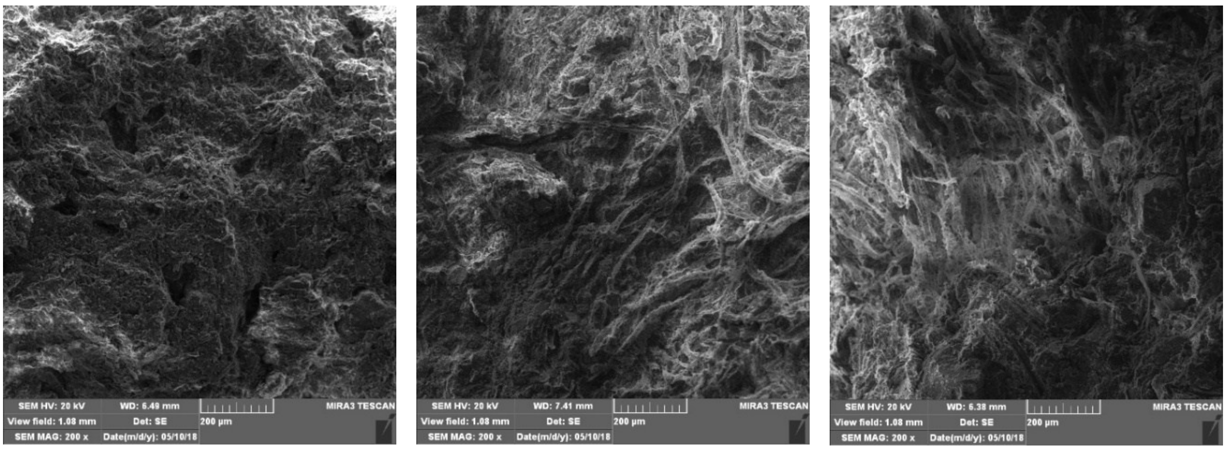
Table 5. Properties with the change of temperature and cigarette butts content.

Cigarette butts content [%]	Temp. [K]	Apparent porosity [%]	Water absorption [%]	Apparent density [g/cm^3]	Flexural strength [MPa]	Compressive strength [MPa]
	1123	29.19	17.01	1.716	3.5	18.3
0	1173	27.21	15.64	1.74	4.2	21.1
	1223	22.91	12.56	1.82	6.5	26.4
	1123	37.2	25.33	1.45	2.3	8.8
5	1173	36.57	25.07	1.47	2.6	10.7
	1223	32.31	20.61	1.56	3.2	14.6
	1123	43.32	34.98	1.23	1.0	3.2
10	1173	42.84	33.88	1.26	1.5	4.7
	1223	38.89	28.78	1.35	1.7	5.0



Clay Clay+ 5% of the butts Clay+ 10% of the butts

Figure 7. Recordings of clay and clay with cigarette butts on the optical microscope.



Clay Clay+ 5% of the butts Clay+ 10% of the butts

Figure 8. SEM of the clay sample and samples of the clay with butts addition.

4. Conclusions

By examining the influence of the addition of cigarette butts in the amount of 5 mass% and 10 mass% to the properties of brick clay from "Čavka" deposit (Busovača), the following conclusions were reached:

- The addition of the cigarette butts requires a larger amount of water to obtain the desired clay workability.
- Addition of the cigarette butts has positive effect on the drying of the clay. Although the mass loss during the drying is higher, the drying shrinkage is lower which minimize possibilities of cracks forming. All these factors enable using higher drying rate thus saving the energy and time.
- During the firing cigarette butts mostly burn out leaving pores behind. The result of this process is increase of mass loss, but decrease of shrinkage.
- Addition of the cigarette butts decreases apparent density, and increases porosity and water absorption.
- As the porosity increases, thermal conductivity decreases and final products have better insulating properties. At the same time, due to higher porosity, the products are lighter than the traditional bricks that is more favorable for manipulating and transporting.
- Higher porosity of the mixtures is related with decreasing of compressive and flexural strength which are still higher than minimal requirements for the wall bricks.

Using the waste material as the cigarette butts, one part of the waste is disposed, and production process enable saving of energy and time. Apart from the production process, energy is saved in buildings made of bricks due to their low thermal conductivity.

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