RECYCLING MUNICIPAL SOLID WASTE INCINERATION (MSWI) FLY ASH AS ADDITION FOR CLAY BRICK

V Voišnienė¹, O Kizinievič² and V Kizinievič²
¹ PhD student, VGTU, Department of Building Materials and Fire Safety, LT
² Senior Research Fellow, VGTU, Institute of Building Materials, LT
E-mail: olga.kizinievic@vgtu.lt

Abstract. The authors analyses the possibilities of recycling municipal solid waste incineration (MSWI) fly ash in clay brick manufacture. The paper analyses the effect of MSWI fly ash (FA) on physical-mechanical characteristics, environmental toxicity of clay bricks. The clay bricks were prepared by adding 2.5-7.5 % of FA and firing them at 1000 °C temperature. Physical and mechanical characteristics of clay brick change depending on the content of FA added: shrinkage, density and compressive strength reduce, water absorption and total porosity increase. Clay bricks containing 2.5-5 % FA do not exceed the limit values of inert substances according to the Waste Directive 2003/33/EC. According to the test results and taking into consideration clay brick physical - mechanical characteristics and environmental toxicity, the recommended content of the addition to the moulding compound is 2.5-5 % FA.

1. Introduction
Nowadays, there are growing pressures to use waste materials within the construction sector. This use of wastes in civil engineering requires an evaluation of both the environmental and technical suitability of the waste.

Thousands of millions of tons of municipal solid waste (MSW) are produced every year. Waste management and utilization strategies are major concern in many countries. Incineration is a common technique for treating waste, as it can reduce waste mass by 65-80 % and volume by up to 85-90 % [1], as well as providing recovery of energy from waste to generate electricity. Generally, municipal solid waste incineration (MSWI) produces two main types of ash, which can be grouped as bottom ashes (BA) and fly ashes (FA). MSWI Fly ash has classified as a hazardous waste (Code in European waste catalogue, code 1 01 12) [2].

Literature analysis revealed that the main components of fly ash are CaO, SiO₂ and Al₂O₃ [3-6]. There is also a significant content of chlorides. Fly ash also contains high levels of heavy metals, such as Zn, Pb, Cu, Cd, Cr and other [7-9].

The ceramic industry is highly promising for the final disposal of MSWI, is known about reuse of fly ash waste in clay brick. The reason for such a lack of data in this area relates to the fact that MSWI fly ash chemical composition is very different [3-9].

MSWI fly ash was used successfully during the preparation of clay brick [10, 11]. Jordan M.M et al. [12] propose to add 1 %–10 % of fly ash and 15 %–35 % of marble waste to ceramic brick moulding compound and fire the bricks at 975 °C–1050 °C temperature. It was found that with higher content of fly ash the flexural strength decreases and water absorption increases.
It was proven that clay bricks containing water treated fly ash have better physical and mechanical characteristics compared to unmodified clay bricks. The proposed FAW content is 5%. Such products have density of 1.63 g/cm³, porosity of 41%, water absorption of 25.5%, and compressive strength of 8 MPa [13].

The main objective of the present study is to investigate the feasibility of Municipal solid waste incineration fly ash to be used for the production of clay bricks. The impact of the amount of MSWI fly ash in the clay mixture is discussed in terms of physical–mechanical properties, porosity and environmental toxicity.

2. Materials and methods

Mixture of the clay, sand and MSWI fly ash is used in the research. Raw materials and additive are dried at 105±5 °C temperature, ground and sieved out through 1.25 mm sieve. Initially, dry mixture of the components is mixed, then mixture is watered till the humidity reached the level suitable for the formation. The amount of water required for formation masses varies from 20% to 23%. The higher amount of MSWI FA is introduced into formation masses, the higher amount of water is required. The mixing of forming masses is performed manually. This formation mass is kept for three days at (95-5)% humidity, in order to evenly distribute humidity in the formation mass. After three days of hardening, 50×50×50 mm samples are formed from formation masses. Specimens which have been exposed at normal conditions, are dried in the drying oven: 60±5 °C temperature during the first day, next day at 105±5 °C temperature. Burning of the samples is carried out at 1000 °C temperature, overall burning period is 34 h, by keeping at the highest burning temperature for 1 hours. Composition of the formation masses is presented in Table 1.

| Raw materials | Composition of formation masses (% of the mass) |
|---------------|-----------------------------------------------|
| Clay + Sand   | 100 2.5 97.5 95 92.5                           |
| MSWI fly ash  | – 2.5 5 7.5                                    |

Chemical analysis of brick raw materials is made by using X-Ray Fluorescence Spectrometer (XRF). Physical and mechanical characteristics of the ceramic products are determined through the implementation of the standard methodologies: density - according to LST EN 772-13:2003, water absorption - according to LST EN 771-21:2011, compressive strength - according to LST EN 772-1:2011 [14-16]. Linear shrinkage is measured using a calliper after drying and burning steps. Total porosity is determined in accordance with the methodology [17]. Shrinkage is calculated by employing 1 equation.

\[ L = \frac{L_0 - L_1}{L_0} \times 100 \% \]

where: \( L_0 \) – distance between indentations on the formed sample, mm; \( L_1 \) – distance between indentations on the dry or burnt sample, mm.

The amount of heavy metals in the eluate from clay brick was determined by atomic absorption spectral analysis method using Buck Scientific 2010 VGP spectrometer with air-acetylene flame. The eluate was prepared according to standard LST EN 12457-2:2003 [18]. The solution for pH and specific conductivity tests was prepared from 2 g of test material and 100 g of distilled water. The prepared solution was agitated for 45 minutes in a magnetic stirrer, filtered afterwards and tested. The specific conductivity was measured by electric conductivity meter Cond 315i equipped with electrode Tetra Con 325. The alkalinity of the solution was measured by pH meter HANA HI 9321.
3. Results and discussion
The chemical analysis shows that the major components in the clay were SiO$_2$ (51.43 %), Al$_2$O$_3$ (20.54 %) and Fe$_2$O$_3$ (7.44 %). The next most abundant components were CaO (4.26 %), MgO (3.07 %), K$_2$O (3.3 %) and Na$_2$O (0.42 %), L.o.i – 9.54 %. Further X-ray diffraction analysis revealed that the clay used in this research mainly consisted of mica clay, quartz, kaolinite, feldspar. The mixture of clay and sand consists of 90 % of clay and 10 % of sand. According to particle-size distribution (Table 2), the clay is a dispersible material because the content of 0.01 mm-sized particles is about 70 %. The quartz sand with the following chemical composition was used for the tests: SiO$_2$ > 98.5 %, Fe$_2$O$_3$ < 0.05 %, Al$_2$O$_3$ < 0.60 %; the distribution particle size in sand: 0 % of 2500 μm particles, 0.20 % of 1250 μm particles, 0.30 % of 1000 μm particles, 1.50 % of 800 μm particles, 630 μm particles 2.70 %; 7.50 % of 400 μm particles, 9.40 % of 315 μm particles, 34.40 % of 200 μm particles 17.20 % of 160 μm particles, 17.80 % of 100 μm particles, 8.30 % of 63 μm particles, 0.65 % of 50 μm particles, and 0.05 % of bottom particles.

| Raw materials | 0.25 mm. | 0.25 – 0.05 | 0.05 – 0.005 | 0.05 – 0.01 | < 0.001 mm. |
|---------------|---------|-------------|-------------|-------------|-------------|
| Clay          | 0.10    | 0.52        | 14.85       | 25.67       | 58.86       |

The major components observed in the MSWI fly ash were CaO (45.17 %) and chloride (KCl, MgCl$_2$, NaCl) (19.01 %). The next most components were Na$_2$O (2.65 %), K$_2$O, MgO (1.15 %), Al$_2$O$_3$ (1.08 %), Fe$_2$O$_3$ (6938 mg/kg), TiO$_2$ (1543 mg/kg). High concentrations of heavy metals were observed in the fly ash samples (Table 3). When compared to data disclosed by the literature [4, 5], the MSWI fly ash employed in this work uses to have less silica. This result indicates that the fly ash must be considered as a hazardous solid waste material.

| Title and Test results and units of measurement (* mg/kg) |
|---------------------------------------------------------|
| P$_2$O$_5$ | Na$_2$O | K$_2$O | MgO | CaO | SiO$_2$ | Al$_2$O$_3$ | TiO$_2$ | Fe$_2$O$_3$ | MnO$_2$ |
| 0.55%       | 2.65%   | 3.52%  | 1.15% | 45.17% | 0.23% | 1.08% | 1543* | 6938* | 334* |
| As          | Pb      | Cd     | Cr   | Cu   | Ni     | Hg     | Zn     | Ba    | Mo   |
| 13.8*       | 1987*   | 83.4   | 50.5 | 625* | 12.2*  | 10.4*  | 10209* | 237*  | < 1* |
| Sn          | Be      | Co     | V$_2$O$_5$ | Sr | 367* | 5.99* | 9.55* | 239* |

The quality of clay brick obtained with up to 7.5 % MSWI fly ash after firing at 1000 °C was determined on the basis of their technological properties (linear shrinkage, water absorption, density, total porosity and compressive strength). Figure 1 shows the amount of shrinkage after drying and firing. The shrinkage of a control clay brick (0 % FA) after drying is 9.1 %. When the MSWI FA content in the mixture varied from 2.5 % to 7.5 %, the brick shrinkage after drying changed from – 7.5 % to 8.4 %. The shrinkage of a control clay brick (0 % FA) after firing is 11.2 %. When the MSWI FA content in the mixture varied from 2.5 % to 7.5 %, the brick shrinkage changed from – 8.5 % to 9.3 %.
During sintering, open and closed pores are usually formed. The maximum density and water absorption corresponds to the maximum volume of open pores in the clay brick. The measurements of the water absorption and density of clay brick with different proportions of MSWI fly ash are shown in Figure 2. Clay bricks (0 % FA) have a density of 1.92 g/cm$^3$, water absorption – 9.5 %. The results indicate that the water absorption of the bricks increased as the MSWI fly ash content increased. The less water that infiltrates the brick, the greater its durability and resistance to the natural environment are expected. The density (2.5–7.5 % FA) was from 1.6 g/m$^3$ to 1.82 g/m$^3$. The results indicate that when the MSWI fly ash content was decreased the density of the bricks increased.

**Figure 1.** Shrinkage after drying and firing of clay brick.

**Figure 2.** Water absorption and density of clay brick.
The compressive strength is the most important engineering quality index for building materials. The results of compressive strength testing of the clay bricks and MSWI fly ash mixtures are shown in Figure 3. The results indicate that the compressive strength of the clay bricks decreased as the MSWI fly ash content increased. The higher is the water absorption, the higher is the total porosity of clay brick. The lowest total porosity was found in control clay brick (0 % FA). The total porosity of this clay brick is 27 %. Total porosity parameter indicates the changes of macro- and microstructure during the firing.

The firing colour is another important parameter to qualify clay bricks. In this study, all clay brick pieces fired at 1000 °C presented a light red colour, regardless of the waste amount that had been added to them.

Leaching results of clay bricks were shown in Figure 4-5. All leaching results from sintered bricks were far lower than the Non-hazardous waste limit value (2003/33/EC directive), and thus heavy metals were stabilized in the body of clay brick through the sintering process. Hence, utilization of MSWI fly ash in making ceramic brick was an effective way to stabilize heavy metals. Clay bricks with fly ash addition also have higher pH and electrical conductivity values compared to clay bricks without FA addition (Table 4). Due to the existence of CaO, sulphate and chloride, MSWI fly ash is highly alkaline, which is detrimental to its reuse. The original pH of Clay + sand mixture is around 7.53, electrical conductivity (Sel) is 355 µS/cm. Clay with 7.5 % FA pH is 8.9, Sel 1887 µS/cm.

Although the limit values of non-hazardous waste set forth in Directive 2003/33/EC have not been reached, we assume that the limit values set for the leaching of inert materials should be observed when hazardous waste, such as MSWI fly ash, is utilised in the manufacture of building materials.

The use of 2.5 % MSWI fly ash as raw material for the production of clay bricks seems to be an important recycling way for final disposal of this abundant waste.
Figure 4. Leaching result of heavy metals Cr, Cd, Pb from clay bricks (* - inert waste limit values according to 2003/33/EC).

Figure 5. Leaching result of heavy metals Cu, Zn, Ni from clay bricks (* - inert waste limit values according to 2003/33/EC).

Table 4. Results of testing pH, Sel from clay bricks.

| pH, Sel, values | Clay brick (with FA addition) |
|-----------------|-----------------------------|
|                 | 0  | 2.5 | 5.0 | 7.5 |
| pH              | 7.53 | 8.17 | 8.53 | 8.9 |
| Sel, µS/cm      | 355  | 1160 | 1488 | 1887 |
Conclusions
In this study, the properties of clay bricks with MSWI fly ash are investigated. It is found that the additive of MSWI fly ash is additive which influences physical – mechanical properties and environmental toxicity:
- after the addition of 2.5% of MSWI additive into clay mixture and after burning samples at 1000 °C temperature, the clay brick with the following parameters is obtained: density – 1800 kg/m³, compressive strength – 27 MPa, linear shrinkage – 9.4% (after burning), water absorption – 15.3%, total porosity – 31.0%;
- tests of heavy metals leached from clay bricks with MSWI FA additions revealed that in specimens containing 2.5% of FA the leaching did not exceed the limit value for inert waste set in Directive 2003/33/EC.

References
[1] Bertolini L et al 2004 Cement and Concrete Research 34(10) pp 1899–1906
[2] Europe Waste Catalog. Guidance on using the European Waste Catalogue (EWC) to code waste November 2015
[3] Wunsch P et al 1996 Chemosphere 32 pp 2211–2218
[4] Pan J R et al 2008 Waste Management 28 pp 1113–1118
[5] Li X et al 2014 Waste Management 34 (12) pp 2494–2504
[6] Shi S H and Kan L L 2009 Journal of Hazardous Materials 164(2-3) pp 750–754
[7] Aubert J E et al 2006 Journal of Hazardous Materials 136(3) pp 624–631
[8] Tang J et al 2017 Journal of Cleaner Production 148 pp 595–605
[9] Belmonde L J et al 2016 Environmental Science and Pollution Research pp 1–13
[10] Haiying Z and Jingyu Q 2011 Waste Management 31(2) pp 331–341
[11] Jordán M M et al 2015 Fresenius Environmental Bulletin 24(2) pp 533–538
[12] Pan J R et al 2008 Waste Management 28 pp 1113–1118
[13] LST EN 772–13:2003 Methods of test for masonry units – Part 13: Determination of net and gross dry density of masonry units (except for natural stone)
[14] EN 772–21:2011 Methods of test for masonry units. Determination of water absorption of clay and calcium silicate masonry units by cold water absorption
[15] LST EN 772–1:2003 Methods of test for masonry units – Part 1: Determination of compressive strength
[16] Kizinievič O et al 2015 Ceram Inter 41 (9) pp 11234–11241
[17] LST EN 12457-2:2003 Characterisation of waste - Leaching - Compliance test for leaching of granular waste materials and sludges - Part 2: One stage batch test at a liquid to solid ratio of 10 l/kg for materials with particle size below 4 mm (without or with size reduction)