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Leaching of copper, lead and zinc from municipal solid waste incineration bottom ash

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Abstract

Municipal solid wastes incineration (MSWI) in modern waste-to-energy plants reduces the volume of MSW by up to 90 %. Incineration process also produces two types of by-products – bottom ash (BA) and fly ash (FA). Municipal solid wastes incineration BA is rich in heavy metals and salts. The disposal of MSWI BA may cause serious environmental problems. The objective of this study is to assess fresh bottom ash chemical composition and heavy metals such as copper (Cu), lead (Pb) and zinc (Zn) leaching values. The chemical composition determination indicates that Cu (400–800 mg kg^{-1}), Pb (680–1760 mg kg^{-1}) and Zn (1330–2010 mg kg^{-1}) are major heavy elements in MSWI bottom ash. The results showed that the concentrations of Cu were 0.1–2 mg kg^{-1}, Pb – 0.2–12.2 mg kg^{-1}, Zn – 0.4–5 mg kg^{-1}. In order to reduce the leaching of heavy metals from bottom ash, they should be pre-treated. The most commonly used technologies are weathering, washing and metals separation.

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Keywords: municipal solid waste incineration; bottom ash; copper; lead; zinc; leaching; chemical composition

1. Introduction

With increasing consumption of goods and rapid urbanization, the generation of municipal solid waste (MSW) has increased drastically [1]. Municipal solid waste incineration is one of many waste management technologies, which

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is important for sustainable economic and environmental development [2] Incineration reduces waste mass by 70 % and volume by up to 90 %. Moreover, it produces energy in the form of heat and electricity [3, 4].

According to the International Solid Waste Association (ISWA) there were 459 waste incineration plants in 28 European Union (EU) countries in 2013 as well as 86 waste incineration plants in the United States (excluding hazardous waste incineration plants) [5, 6]. Compared to with 2001, number of waste incineration plants in EU increased by 14.18 %. Waste incineration technology is widely applied in other regions of the world: 74 waste incineration plants operate in China, 19 in Taiwan, 7 in Australia, 5 in Canada and Singapore.

Municipal solid waste incineration (MSWI) produces two main types of combustion ash: bottom ash (BA) and fly ash (FA) [7, 8]. Accounting for nearly 80–85 % (16–35 % of the input waste mass) MSWI bottom ash is the most significant by-product [9–14]. In 2014, according to Eurostat data, it was burnt 64.377 mln. T. municipal waste and generated about 55 mln. T. bottom ash in the European Union [15].

Bottom ash (BA) is a highly heterogeneous mixture of slag, non-ferrous and ferrous metals, glass and ceramics, other non-combustible and residual organic matters [16]. Unlike FA, bottom ash is classified as non-hazardous waste by the European Waste Catalogue. MSWI bottom ash is mainly composed of silica (31.93–59.59 %), alumina (5.80–18.61 %), calcium (7.58–35.00 %) and iron (5.50–17.05 %) oxide (Table 1), which are natural aggregate compounds [10]. Table 1 gives chemical compositions of bottom ash, which were generated in MSWI plants in various countries.

Table 1. Amount of main chemical elements in MSWI bottom ash.

Oxide	Amount, % wt							
	Spain [8]	Italy [13]	Germany [17]	Netherlands [4]	Japan [11]	China [10]	Taiwan [18]	USA [19]
SiO_2	43.3	33.70	55.70	54.23	31.93	59.59	50.30	23.64
CaO	16.9	35.00	11.9	13.45	33.40	7.58	15.27	23.82
Fe_2O_3	14.1	5.37	8.80	13.83	5.97	5.50	7.72	17.05
Na_2O	7.58	2.27	1.40	2.81	2.53	1.32	1.30	1.70
Al_2O_3	5.80	13.31	14.1	7.86	16.65	18.61	16.43	14.25
MgO	2.22	4.62	2.70	1.81	3.33	1.32	n. d.	1.85
K_2O	1.11	1.66	1.2	0.88	2.22	2.29	2.14	0.42

Note: n. d. – no data.

However, chlorides, sulphates and heavy metals, such as copper (Cu), zinc (Zn), lead (Pb) often present in high concentrations in BA [16]. Table 2 gives amount of heavy metals in bottom ash.

The research conducted in different countries showed (Table 2) that bottom ash contains of large quantities of zinc (903–7732 mg kg^{-1}), copper (1041–7743 mg kg^{-1}), lead (687–4552 mg kg^{-1}) and barium (1126–3920 mg kg^{-1}). Also, there were detected chromium, nickel, arsenic. Concentrations of cadmium and cobalt in the bottom ash were small (1–92 mg kg^{-1}).

Table 2. Amount of heavy metals in MSWI bottom ash.

Metal	Amount, mg kg^{-1}				
	Japan [11]	Italy [13]	Sweden [7]	Germany [20]	Denmark [21]
Zinc (Zn)	3193	903	3800	7732	2600
Copper (Cu)	2321	1041	2700	7743	2060
Lead (Pb)	687	4552	1400	1022	1100
Chrome (Cr)	393	119	490	1158	449
Nickel (Ni)	105	45	240	356	356
Barium (Ba)	1126	n. d.	1300	3920	1600
Arsenic (As)	< 1	16	240	21	27
Cadmium (Cd)	1	92	4	14	3
Cobalt (Co)	5	14	33	67	20

Note: n. d. – no data

There are a lot of bottom ash management technologies in the world; the main one is landfilling used for the road and building construction elements [8, 16, 22–24]. The lowest investment-consuming but the least desirable one is the bottom ash removal in non-hazardous waste landfills. Environmental precipitation and various chemical reactions influence bottom ash, therefore leachate is formed, which contains high amounts of various salts and heavy metals. A leachate spill into the environment can cause harmful effects in soil and surface, and underground water [25]. In order to avoid the harmful effects of MSWI bottom ash removal of inert and non-hazardous waste landfills, the compliance requirements of EU directive 2003/33/EC [26] or country-specific legislation must be fulfilled.

The aim of this research is to determine MSWI bottom ash chemical composition and maximum leaching of heavy metals (copper, lead and zinc). Also, this work evaluates landfilling and re-use capabilities of bottom ash.

Nomenclature

w_w	water content, %
m_a	mass of the empty dish, g
m_b	mass of a dish containing the sample, g
m_c	mass of a dish containing the dried sample, g
f	conversation factor, f = 100 for expression of results in %

2. Materials and methods

2.1. Materials

BA samples were collected from a single MSW incinerator located in Klaipeda (Lithuania). The feed stream is commonly composed of household waste with smaller proportions of commercial waste and solid biofuel. The Grate furnace incinerator operates at temperatures between 850 and 1100 °C with a capacity of 698 tons/d. The representative six bottom ash samples (30 kg) were collected from the different spots of the large bottom ash piles (Fig. 1) one time in a week and then sealed in plastic buckets before further testing.

Fig. 1. Piles of MSWI bottom ash in the incineration plant in Klaipeda.

A test portion (2 kg) after homogenization and weighting was dried in the oven at 105 °C until a constant mass was obtained and the water content was determined according to the following formula [27]:

$$w_w = \left[1 - \frac{(m_c - m_a)}{(m_b - m_a)}\right] \times f \tag{1}$$

where

w_w	the water content, %;
m_a	the mass of the empty dish, g;
m_b	the mass of a dish containing the sample, g;
m_c	the mass of a dish containing the dried sample, g;
f	the conversation factor f = 100 for expression of results in per cent.

2.2. Chemical composition

The chemical composition of the major and minor elements in MSWI bottom ash was determined in duplicate by X-ray Fluorescence Spectroscopy (XRF) using a Axios mAX X-ray spectrophotometer (Fig. 2).

Fig. 2. X-ray fluorescence spectroscope with wave dispersion (WD-XRF) Axios MAX.

The chemical composition determination results are shown in Fig. 4.

2.3. Leaching test

The compliance leaching test was performed according to the Standard EN 12457-2 [28]. The samples, which originally and after a pre-treatment had a size below 4 mm, were put in contact with distilled water in capped bottles. Liquid to solid ratio was 10/1 (900 mL distilled water and 90 g dry BA) and the suspension was agitated for 24 h at room temperature, 20 ± 5 °C. Solid residue was separated by filtration, then each eluate pH (Metrel Toledo) and conductivity (WTW Terminal 740) were determined. Furthermore, after acidification (pH 2 using HNO3) metals (copper, lead and zinc) content was analysed by AAS according to ISO 15586:2003 [29]. The results (Table 4) were compared with the limit leaching values according to the 2003/33/EC.

3. Results and discussion

3.1. Water content in BA

After six samples drying to constant weight the amount of water was prescribed by Eq. (1). The results are shown in Fig. 3.

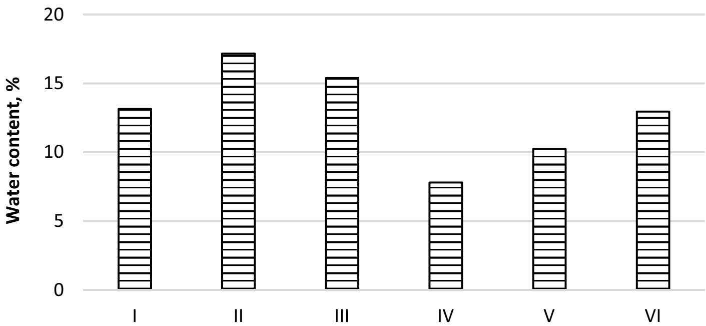

Fig. 3. X-ray fluorescence spectroscope with wave dispersion (WD-XRF) Axios MAX.

The results (Fig. 3) reveal that untreated bottom ash water content varies from 7.80 % to 17.16 % wt. Water content values are not stable, because bottom ash in the waste incineration plant is cooled by water. One part of the cooling water evaporates immediately, the other part is absorbed by BA.

3.2. Chemical composition

The six samples untreated bottom ash, taken from the incineration plant in Klaipeda bottom ash storage facilities in February – April, chemical composition determination research showed that the main components (> 10 %) were oxygen, silicon and calcium (Fig. 4). Also, it was found (1–8 %) aluminium, magnesium, sodium, potassium and iron. The above-mentioned elements were found in the form of oxides. Silicon, a part of a silicon dioxide (SO_2) is the main element of the composition of bottom ash. The research shows that this oxide constitutes more than half (57 ± 2 %) of the total weight of BA (Fig. 4). In BA also is 16 ± 2.5 % CaO, 8 ± 3.2 % Fe_2O_3, 5 ± 1 % Na_2O and 5 ± 0.5 % Al_2O_3.

Heavy metals (Cr, Mn, Ni, Cu, Zn, Sr, Pb) concentration in BA was relatively low (< 1 %). The highest amounts were of copper 400–800 mg kg^{-1}, lead 670–1760 mg kg^{-1} and zinc 1330–2010 mg kg^{-1}.

Bottom ash chemical composition depends mainly on the initial composition of the incinerated waste. After sorting remaining waste consists of inert waste, therefore, BA contains a high amount of silica and calcium oxide.

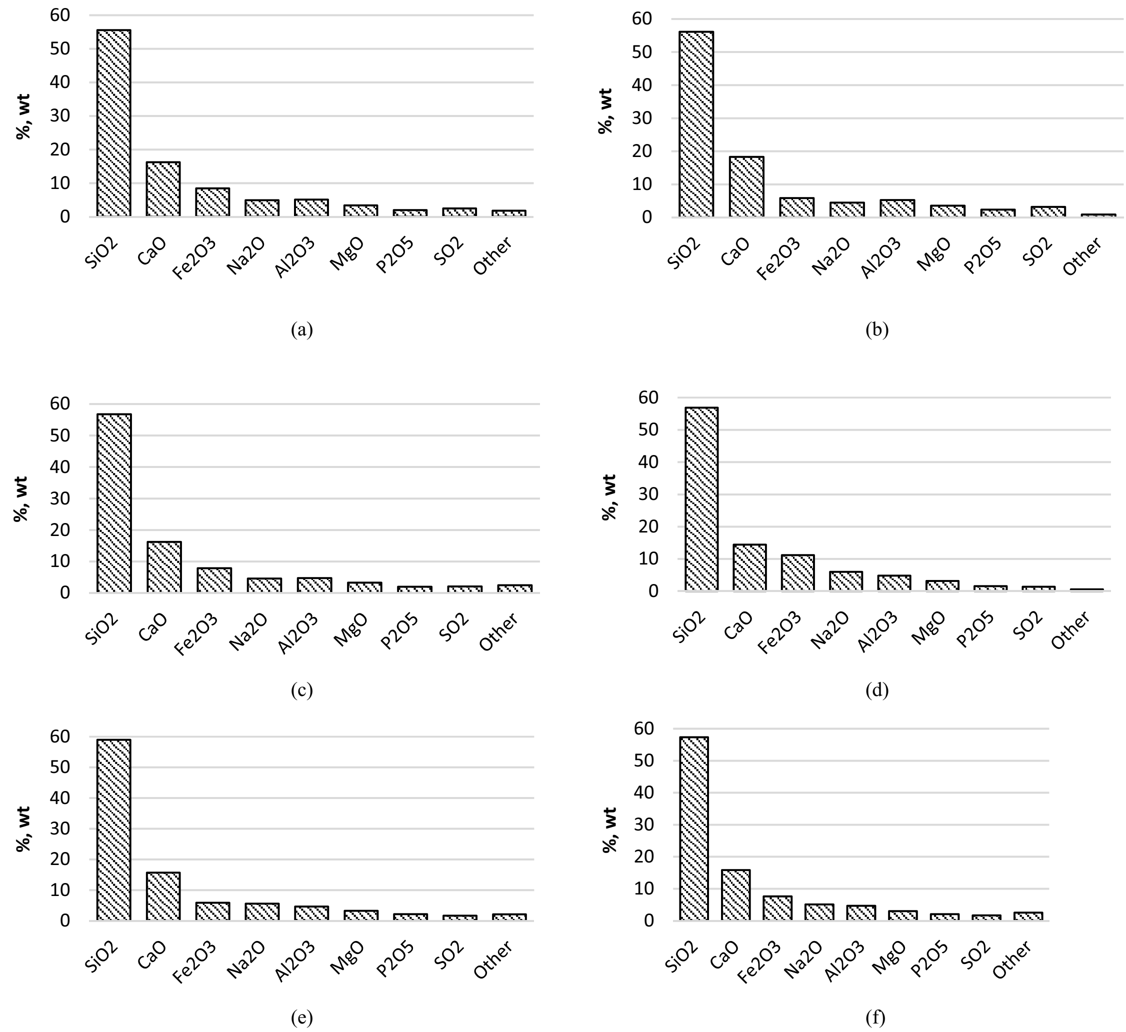

Fig. 4. Chemical composition of MSWI bottom ash, sample number: a) I; b) II; c) III; d) IV; e) V; f) VI.

The waste can also include different metals such as copper, iron, aluminium and another waste, on which directly depends the content of metals in BA. If we want to reduce the amount of metals from bottom ash, they should be removed from the municipal waste stream. However, this process is complicated and requires a large investment. Much simpler, although it is not economically beneficial due to the small content of recoverable metals, method is the separation of metals from municipal waste bottom ash. The separation of metals from bottom ash can be partial or complete.

3.3. Leaching results

The results of the natural pH batch leaching tests on the collected materials are shown in Table 3. The leaching data are compared to the European Commission decision 2003/33/EC regulatory limit values for waste removal in inert and non-hazardous waste landfills.

Two metals – zinc and lead concentrations, respectively one and three times exceed leaching limits for waste, which are removal in inert waste landfills (Table 3). Therefore, the bottom ash cannot be attributed to inert waste. Also, too high concentration (12.2 mg kg^{-1}) of lead is set in the 1 sample, when the limit for non-hazardous waste is 10.0 mg kg^{-1}. Copper concentration was 0.1–2 mg kg^{-1} and did not exceed requirements for inert and non-hazardous waste.

Table 3. MSWI bottom ash eluiate characteristics.

Element, parameter	Sing	Units	I sample	II sample	III sample	IV sample	V sample	VI sample	2003/33/EC	
									Inert waste	Non-hazardous waste
Copper	Cu	mg kg^{-1}	2	1	1	1	0.1	0.1	2	50
Lead	Pb	mg kg^{-1}	12.2	2.4	2.5	0.3	0.1	0.2	0.5	10
Zinc	Zn	mg kg^{-1}	5	2	2	< 0.4	1	1	4	50
Electrical conductivity	SEL	µS cm^{-1}	8000	8750	9130	6260	5820	8240		Must be evaluated
pH			12.7	12.6	12.6	12.5	12.4	12.6		> 6

Based on the experience of other countries municipal waste bottom ash is classified as non-hazardous waste, however, these wastes are classified into categories according to the ability to use them in civil engineering. MSWI bottom ash which is used in civil engineering in Germany, must comply Z2 category leaching values (according to the German federal states Community technical document LAGA 20). Table 4 provides a comparison of research results and ash leaching values limits.

Table 4. Comparison of research results and LAGA 20 limit values for BA.

Element, parameter	Units	I sample	II sample	III sample	IV sample	V sample	VI sample	LAGA20				
								Z0	Z1.1	Z1.2	Z2	Bottom ash eluate permissible values
Cu	µg/l	180	77	63	90	9	6	50	50	150	300	300
Pb	µg/l	**1200**	**250**	**240**	30	6	17	20	40	10	200	50
Zn	µg/l	**500**	210	180	<40	68	72	100	100	300	600	300
SEL	µS cm^{-1}	**8000**	**9130**	**8750**	**6260**	**5820**	**8240**	500	500	1000	1500	6000
pH		**12.7**	**12.6**	**12.6**	**12.5**	**12.4**	**12.6**	6.5–9	6.5–9	6–12	5.5–12	7–13

Note: (blue) - results mismatch for Z2 category limits; (red) - results mismatch for Bottom ash eluate permissible values and Z2 category.

The presented data show that (Table 4) MSWI bottom ash eluate values do not match requirements for Z2 category in the 3 parameters (pH, SEL, Pb). Also, according to Criterion 3 (SEL, Pb, Zn), it exceeds the permitted municipal waste bottom ash leaching (eluate) values.

It can be concluded that the bottom ash is not suitable for use in road and building construction elements. First of all, it must be reduced by leaching of heavy metals. At the moment, the cheapest and the most effective technology is a natural aging (weathering) of the bottom ash. When the bottom ash is affected by precipitation like rain, a chemical reaction starts which generates a calcium carbonate ($CaCO_3$) and calcium aluminate hydrates. These substances act as a binder; they stabilize (curing process) bottom ash constituent components and reduce the leaching of heavy metals. Also, the other technologies such as washing, vitrification at high temperatures can be used.

4. Conclusions

- The six samples of bottom ash chemical (elemental and oxide) composition analysis show that the chemical composition of the time is a little variable. The main elements of the bottom ash are silica (57 ± 2 %), calcium (16 ± 2.5 %), iron (8 ± 3.2 %), sodium (5 ± 1 %) and aluminium (5 ± 0.5 %) oxides;
- As MSWI bottom ash eluates analysis demonstrates, it can be concluded that MSWI bottom ash does not meet requirements for inert waste (2003/33/EC), but it meets requirements for the waste removal in non-hazardous waste landfills. Changing concentration of the environmentally hazardous heavy metals indicates that the need for continuous slag ash eluate test is important;
- In order to improve municipal waste bottom ash quality parameters and reduce the potential negative impact on the environmental components it is recommended full separation of ferrous and non-ferrous metals as well as bottom ash natural aging in the storage area with a hard coating and a waste water (surface water and leachate) collection system.

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