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Fractional Composition of Nickel, Manganese and Iron in Municipal Solid Waste Incineration Bottom Ash

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ABSTRACT
Fractions of Ni, Mn and Fe in the municipal solid waste incineration (MSWI) bottom ash were investigated. Three fractions of studied metals were separated according to the BCR procedure (acid soluble and exchangeable, reducible and oxidizable). Pseudo-total metal content, pH and dry mass in all samples were evaluated. The share of nickel in fractions followed the sequence: F4 (79.0%) > F3 (9.0%) > F1 (7.5%) > F2 (4.5%), for manganese: F4 (60.4%) > F1 (16.8%) > F2 (13.2%) > F3 (9.6%) and for iron: F4 (74.4%) > F2 (12.6%) > F3 (12.5%) > F1 (0.5%). Mobile pool of metals (F1-F3) contained the least of nickel (21.0%) and the most of manganese (39.7%).

Keywords: nickel, manganese, iron, metal fraction, MSWI bottom ash, BCR method.

INTRODUCTION

Total output of the municipal solid waste in the European Union is equal to 245Mt and 26% is incinerated. Incineration process is mostly intended for energy recovery, reduction of waste volume and disposal of hazardous organic compounds (Keber et al. 2020). Bottom ash is comprised mainly of metals (around 10%) and mineral fraction (around 90%) which includes combustion products and non-combustible materials (Huber et al. 2021). Heavy metals and other harmful constituents might have negative effects on the environment (Kumar and Singh 2021), including humans which are the part of it (Staszewski et al. 2015). High metal content in the soil could affect the microbial activity and amount as well as diversity of microorganisms, thus the organic matter decomposition, enzyme activities and mineralization of nitrogen (Alamdari et al. 2022). Nevertheless, waste processing and then utilisation as secondary material is still the best solution (Phua et al. 2019). At present, the processed bottom ash is mainly utilised in Europe as construction material (Cho et al. 2020, Vateva and Laner 2020, Kizinievič et al. 2022). This residue is more often utilised as the raw material in comparison with fly ash; therefore, its influence on the environment must be considered (Wu et al. 2016).

The total content of heavy metals in municipal solid waste incineration (MSWI) bottom ash cannot be the source of information about the threat what they create, since it is tantamount to a statement that all the metal forms are available in the same degree (del Valle-Zermeño et al. 2017). To identify the metal fractions in MSWI bottom ash, the sequential extraction procedure is used (Gonzales et. al 2019, Haberl and Schuster 2019, Kitamura et al. 2022). Each fraction refers to a chemical form of metal which might be released into environment under different conditions (Jabłońska-Czapla et al. 2014). In the present research, the BCR method was used. This method was worked out to standardize the sequential extraction procedures (Pöykiö et al. 2013), since many schemes have been developed, which makes it difficult to compare the study results worldwide.

During incineration, the heavy metals included in municipal solid waste are cumulating mainly in the bottom ash (Phoungthong et al. 2016). According to del Valle-Zermeño et al. (2017) the finest fractions of bottom ash (particle size <4mm) contain the largest amounts of heavy metals, mainly in the form of metal oxides. The authors stated that leaching potential of metals from that fraction might be higher, as compared to the larger fractions. The highest contents of Ni and Mn were noted in fraction ≤2mm. The goal of this study was the assessment of Ni, Mn and Fe fractions in this particular size fraction.

MATERIALS AND METHODS

Bottom ash sampling

Fresh bottom ash was collected from municipal solid waste incineration plant located in the city of Białystok, Poland (23°10'07" E, 53°09'30" N). Figure 1 shows the incineration plant scheme. Twenty samples were gathered, roughly 1 kg each, over a period of 49 days in the year 2016. Municipal solid waste was incinerated at temperature of 700°C in the stoker-fired furnace. Annually, the plant is incinerating around 120,000 t of sorted and mixed wastes. During the sampling period, 11650 t was incinerated. Magnetic separation was used to recover the ferrous metals prior to sampling. The bottom ash was cooled down with the water. The collected samples were sieved through a 2 mm sieve and crushed with a mortar.

Physicochemical properties

Dry mass was defined at 105°C by drying until the constant mass was reached. The pseudo-total content of Ni, Mn and Fe was assayed by the flame atomic absorption spectrometry (FAAS), using the iCE 3500 apparatus (Thermo Scientific, Waltham, MA, USA), after microwave-assisted digestion (0.5 g + 3 cm^3 $HClO_4$ + 10 cm^3 HNO_3) with the use of Ethos Easy system (Milestone, Sorisole, Italy). The pH was assessed potentiometrically, after 24 hours, in water suspension (25 cm^3 H_2O + 5 g of ash sample).

Evaluation of metal fractions

Fractional composition of nickel, manganese and iron in the studied bottom ash was assessed with the BCR method, which was accelerated by the usage of ultrasonic processor Sonics VCX 130 (Sonics & Materials, Newtown, USA). Three operationally defined fractions were isolated (Figure 2).

The FAAS method was used to assess the fractional composition of Ni, Mn and Fe. The percentage of studied heavy metals in fractions with reference to the pseudo-total content and the summary amount in F1 – F3 fractions (mobile pool) were calculated. The residual fraction (F4) was defined according to the following formula: F4 = pseudo-total content – (F1+F2+F3).

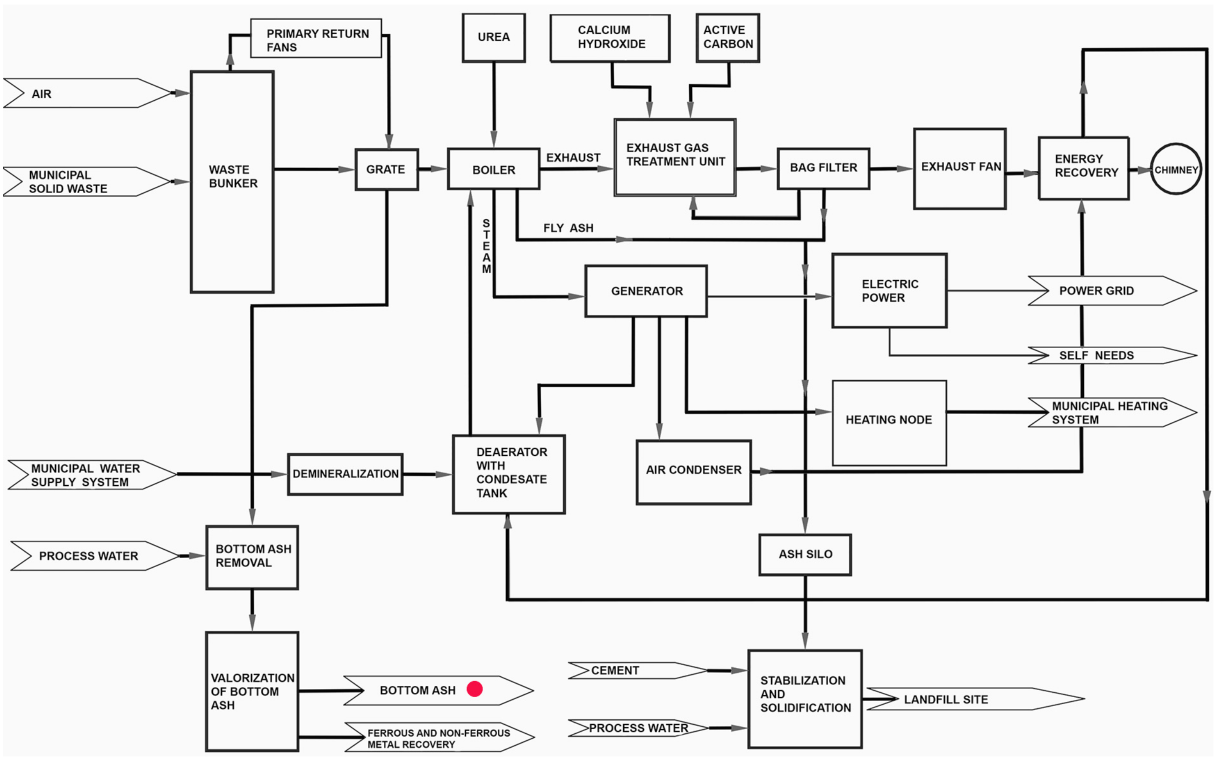

Figure 1. Scheme of the municipal solid waste incineration plant in Białystok (sampling point marked with a red dot)

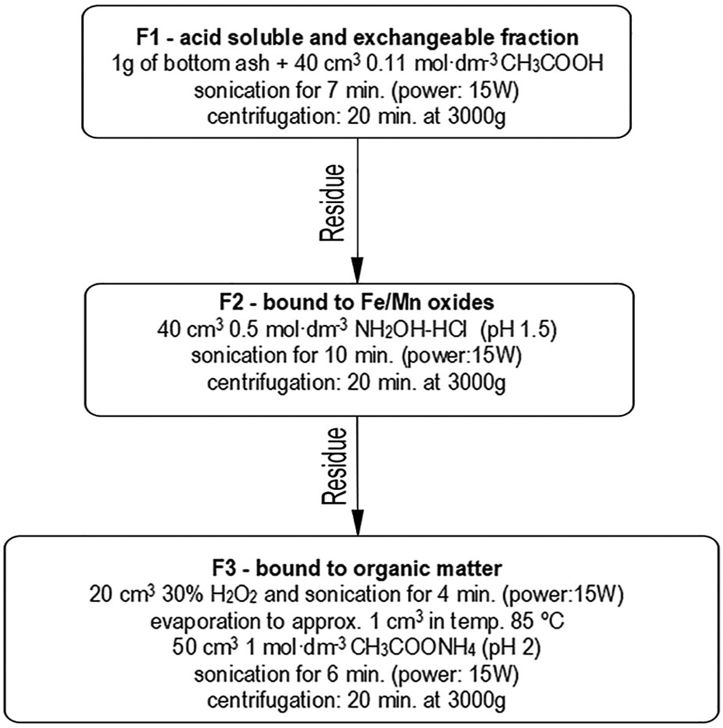

Figure 2. The BCR sequential extraction procedure

RESULTS AND DISCUSSION

Physicochemical characteristics of bottom ash

The details about pH and dry mass of the studied MSWI bottom ash samples can be found in the earlier article (Łukowski and Olejniczak, 2020).

The content of heavy metals in bottom ash depends on the operating conditions of the incineration plant (type of furnace, temperature of incineration, feed rate, burning time, etc.) and the load of waste materials (Wu et al. 2016). Manganese and iron are considered as a non-volatile elements, while nickel as semi-volatile (Yang et al. 2016, Zhu et al. 2021), for that reason Mn and Fe remain mainly in the bottom ash during the incineration process (Wu et al. 2016). The present study results confirm such a claim. In the investigated samples, the highest amounts of iron (5091.8 $mg \cdot kg^{-1}$) and the lowest of nickel (57.70 $mg \cdot kg^{-1}$) were found. The manganese content amounted 727.4 $mg \cdot kg^{-1}$ on average, by the range 540.9-917.0 $mg \cdot kg^{-1}$. Similar results were obtained by Yao et al. (2010) who studied bottom ash from six incineration plants. They noted 410.8–1245.9 $mg \cdot kg^{-1}$ of Mn and 13.4–63.9 of Ni. The results of the current research are confirmed also by Dou et al. (2017). The authors, based on the data from different countries, emphasized that MSWI bottom ash contains approximately10–1000 $mg \cdot kg^{-1}$ of Ni and 100–10000 $mg \cdot kg^{-1}$ of Mn. Conversely, some authors stated clearly higher contents of discussed elements. Vateva and Laner (2020) reported 141.3 $g \cdot kg^{-1}$ of Fe, 2562 $mg \cdot kg^{-1}$ Mn and 701 $mg \cdot kg^{-1}$ of Ni in the predominant mineral part of fraction ≤ 2 mm. Chimenos et al. (2003) noted in the fine fraction 113.3 $g \cdot kg^{-1}$ of Fe, 1285 $mg \cdot kg^{-1}$ of Mn and 177 $mg \cdot kg^{-1}$ of Ni.

Metal fractions in bottom ash samples

The F4 fraction contained the most of nickel, 47.20 $mg \cdot kg^{-1}$ on average (Table 1). It comprised 79.0% of pseudo-total content (Figure 3), at the range from 64.9 to 89.0%. It was at the same time the highest percentage among all the fractions of investigated heavy metals. The F2 fraction gathered 4.5% of nickel. It was the lowest share in that fraction in comparison with the rest of studied metals. The percentage of Ni in the F1 and F3 fractions was similar, 7.5 and 9%, respectively. The metals in the F1 fraction are the most mobile and available. They can be released easily from the MSWI bottom ash particles under slightly acidic or even neutral conditions by the ion-exchange process. The F3 fraction includes the metals bonded with sulfides and organic matter (Pöykiö et al. 2016). Fulvic or humic acids, which are the part of the organic fraction of the soil, when exposed to oxidizing conditions, can

Table 1. The pseudo-total content of Ni, Mn, Fe and amount in the fractions

Metal	Sample	Min–Max	Mean ± SD, n = 20
Ni (mg·kg^{-1} DM)	Pseudo–total content	18.95–144.17	57.70 ± 35.16
	F1	1.20–7.89	3.79 ± 1.94
	F2	0.45–6.23	2.56 ± 1.69
	F3	0.47–8.21	4.14 ± 1.79
	F4	20.36–126.77	47.20 ± 32.03
Mn (mg·kg^{-1} DM)	Pseudo–total content	540.9–917.0	727.4 ± 111.4
	F1	98.2–141.9	119.1 ± 13.1
	F2	75.3–132.5	93.8 ± 14.0
	F3	46.5–94.6	67.9 ± 14.7
	F4	231.4–658.3	446.6 ± 120.2
Fe (mg·kg^{-1} DM)	Pseudo–total content	4572.6–5507.6	5091.8 ± 282.3
	F1	11.7–42.7	24.1 ± 9.1
	F2	412.1–753.1	645.5 ± 104.4
	F3	511.1–754.7	634.4 ± 56.7
	F4	3399.8–4238.7	3787.8 ± 234.3

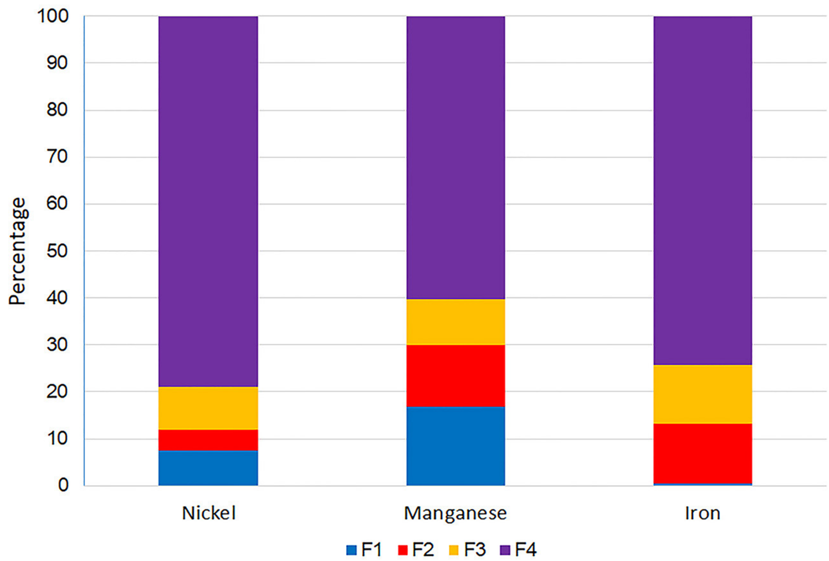

Figure 3. Fractions of Ni, Mn and Fe in bottom ash

release these elements into the solution (Bruder-Hubscher et al. 2002).

Nickel mobile pool, 21.0% at the range from 11.8 to 35.1%, was the least among studied metals. Taking into account its pseudo-total content, it can be said that nickel leachability does not represent a high threat to the environment, since Ni toxicity for most humans, animals and plants might emerge at the level of 100 mg·kg^{-1} of soil (Boostani et al. 2022).

The highest content of manganese, 446.6 mg·kg^{-1} on average, was stated in the F4 fraction. It was 60.4% of pseudo-total content, within the range of 42.8–71.9%. A little less Mn in this fraction, about 50%, was noted by Yao et al. (2010) in bottom ash samples from four incineration plants. The authors also found approximately 25 to 55% of Mn in carbonate and exchangeable fraction, 12.7–36.9% in Fe/Mn oxides fraction and no more than few percent in organic matter fraction. Otherwise, in the present research, the percentage of Mn in the F1 and F3 fractions was lower and amounted to 16.8 and 9.6%, respectively. The share in the F2 fraction (13.2%) was similar. The metals in this fraction, under appropriate redox potential and pH, can be released and incorporated into the food chain (Long et al. 2022). Decomposition of Fe/Mn oxides occurs readily under natural conditions (Shi et al. 2020). Bruder-Hubscher et al. (2002) found in bottom ash, which was stored for a year, mostly lower amounts of manganese in particular fractions according to the BCR method, as compared to the current study. Only in the organic matter fraction, they found more manganese (13–25%).

The mobile pool of manganese amounted to 39.7%, at the range from 28.1 to 57.2%, and was the highest among the studied elements. It means that the potential release of Mn into the soil and ground water was also the highest.

Moreover, Fe, similarly to Ni and Mn, was present mostly in the F4 fraction (3787.8 $mg \cdot kg^{-1}$). This fraction includes heavy metals, which might be released only under very acidic conditions or due to the activity of microorganisms (Zimmerman and Weindorf 2010, Łukowski and Wiater 2011). Thus, residual fraction is considered as most stable and not harmful to the natural environment (Wielgosiński et al. 2014). The percentage of iron amounted 74.4% and ranged from 71.2 to 79.7%. Gonzales et al. (2019) found 97 wt% (relative amount of weight) of iron in the discussed fraction. According to the authors, it means that Fe was mostly associated with the vitreous phase and aluminosilicate minerals, which indicated its slight leachability. In exchangeable fraction they noted 3 wt% and Fe/Mn oxides fraction contained below 5 wt% of iron. In the present investigations, the F1 and F2 fractions accumulated 0.5 and 12.6% of Fe, respectively. Mobile pool contained 25.6%, at the range from 20.3 to 28.8%.

CONCLUSIONS

The amount of manganese in acid soluble and exchangeable fraction, which is the most mobile and available, was the highest among the studied heavy metals. Moreover, the highest percentage was noted in mobile pool. This confirms that the manganese contained in MSWI bottom ash may be leached out into the soil and groundwater much easier, as compared to the other studied elements. Fractions F1–F3 gathered the least of nickel, which means that this metal was characterized by the lowest solubility. In the most mobile fraction (F1), the lowest content of iron was stated. The share of heavy metals in the residual fraction, which is unavailable under normal environmental conditions, always exceeded 60%. Despite such a high percentage in this fraction, the studied elements still might represent the threat to the environment, due to the large utilisation scale of bottom ash as a construction material.

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