

Increasing the utilization potential of fly ashes from fluidized bed combustion by mechanical treatments

Abstract

The increasing use of biomasses in the production of electricity and heat delivers an increased amount burning residue, fly ash that disposal is becoming more and more restricted and expensive. Therefore, there is a great interest to utilize fly ash in other means than disposal. This study aimed to determine the suitability of air jet sieving and air classification to separate detrimental elements, such as sulfate, chloride and some heavy metals, into fine fractions. Furthermore, the effect of fly ash deagglomeration on classification selectivity was studied. Experiments were conducted using two fly ash samples originated from fluidized bed combustion of peat, forest residues and wastes. The results showed that it is possible to significantly improve the utilization potential of fly ashes using air jet classification. In classification, calcium is concentrated into a fine fraction and silica and aluminum are concentrated in coarse fractions. Sulfate, chloride and studied detrimental metals (cadmium, copper, lead and zinc) enrich to the fine fraction with high selectivity. Deagglomeration had a minor effect, as it slightly improved the classification selectivity for some elements.

Keywords: bioenergy; fractionating; grinding; processing; sieving; sustainability

1. Introduction

Biomass is a sustainable energy source used to produce electricity and heat. In the near future, its share will increase due to political goals, such as the EU 20-20-20 target (European Union 2009), of decreasing CO₂ emissions originating from fossil fuels and increasing renewable energy use. Biomass fly ash is the residue produced from combustion of biomass in power plants, paper mills and other biomass burning facilities. In Finland, biomass, i.e., wood, bark and other forest residues, is commonly co-fired with peat, which works as a supporting fuel that evens out the fluctuation in heat capacity and ensures sufficient heat generation in cold seasons. Sometimes, minor amounts of recycled wood and recycled fuel (REF) are used as well.

Among combustion methods, fluidized bed combustion is efficient and common due to its ability to utilize low-grade fuels with fluctuating quality, composition and moisture content, or mixtures of fuels, in-situ capture of SO_x and low NO_x emission (Patel et al. 2017). However, the fly ash originating from fluidized bed combustion of biomasses has limited utilization potential unlike ashes from pulverized combustion of coal. Annually, 500,000 tons of fly ash originating from fluidized bed combustions of peat and wood are produced in Finland (Korpijärvi et al. 2009). Over 1.5 Mt of fly ash are produced in Europe annually (van Eijk et al. 2012), and this amount is expected to increase. The numbers evidently show that fly ash generation is a critical issue in many countries especially when considering that disposal is becoming more and more restricted and expensive, and therefore applications in which fly ashes could be utilized efficiently are in demand.

Some fly ashes can be utilized in soil improvement (Pedersen 2003), earth construction (Ohenoja et al. 2016b, a), concrete application (Rajamma et al. 2009, 2015) and as a fertilizer (Dahl et al. 2009; Ingerslev et al. 2011; Budhathoki and Väisänen 2016) if their properties comply with regulations. The regulations are country specific although international regulations exist as well. For example, the Finnish national legislation MMM 24/11 (Finnish Ministry of Agriculture and Forestry 2011), which came into effect in 2011, regulates the utilization of ash as a field or forest fertilizer and sets minimum acceptable content for Ca, the sum of phosphorous and potassium, and maximum content for detrimental metals, as shown in Table 1. Fly ashes used in earth construction must to comply

with Government Degree 591/2006 concerning the recovery of certain wastes in earth construction amended by Government Degree 403/2009 (Finnish Ministry of the Environment 2009). For concrete application, the European standard SFS-EN 450-1 (EN 450-1 2012) sets the maximum values for chloride and sulfate; however, it should be noted that the use of fluidized bed combustion fly ashes in concrete is not standardized in EN 450-1, which applies to ashes originating from pulverized combustion where the coal content must be over 60% or over 50% when coal combustion takes place with pure wood. However, these limit values still provide a guideline for fly ash utilization in concrete.

Table 1. Finnish limits for fly ashes to be used as a fertilizer (MMM 24/11), in earth construction (Government Degree 403/2009) and in concrete application (SFS-EN 450-1).

	Limit content for field fertilizers	Limit content for forest fertilizers	Limit content for earth construction	Limit content for concrete application (SFS-EN 450-1)
Ca [%]	>6	>6	-	-
P+K [%]	>2	>2	-	-
As [mg/kg]	<25	<40	<50	-
Ba [mg/kg]	-	-	<3000	-
Cd [mg/kg]	<2.5	<25	<15	-
Cr [mg/kg]	<300	<300	<400	-
Cu [mg/kg]	<600	<700	<400	-
Hg [mg/kg]	<1.0	<1.0	-	-
Mo [mg/kg]	-	-	<50	-
Ni [mg/kg]	<100	<150	-	-
Pb [mg/kg]	<100	<150	<300	-
V [mg/kg]	-	-	<400	-
Zn [mg/kg]	<1500	<4500	<2000	-
Cl [%]	-	-	-	< 0.1
SO ₃ [%]	-	-	-	< 3.0

Quite commonly, excessive content of heavy metals can prevent the use of fly ashes in the aforementioned applications. Manskinen studied the fuel composition of two different ashes and two different ash fractions (bottom and fly ash) originating from fluidized bed combustion and concluded that only bottom ash is possible to utilize as it is (Manskinen 2013). For other ash fractions, the heavy metal contents were too high. Generally, the cadmium content in fly ash produced from wood combustion is especially known to often exceed regulations (Narodoslawsky and Obernberger 1996; Pedersen 2003). Cadmium is considered the most harmful of all heavy metals because it remains in the soil, becomes enriched in food chains and is toxic to organisms (Orava et al. 2006). In addition to heavy metals, the utilization of fly ash in concrete applications is limited by chlorides and sulfates. The presence of chlorides and sulfates in cementitious materials can reduce durability through deterioration of the microstructure (Rajamma et al. 2009).

Classification or fractionation is one interesting option to improve fly ash utilization since there is a strong relationship between fly ash particle size and total heavy metal content (Davison et al. 1974; Obernberger et al. 1997; Camerani et al. 2002). An electrostatic precipitator can be used as a classifier in addition to its initial function of collecting fly ash particles from flue gases (Orava et al. 2006; Van de Velden et al. 2008; Świetlik et al. 2012). Quite often, an electrostatic precipitator consists of two or three units connected in a series. Based on the achieved results, the concentrations of detrimental metal are at their lowest in the first collector chamber and highest in the last chamber (Orava et al. 2006). However, detrimental metal content can be too high even at the first collector chamber and therefore classification afterwards is necessary. This can be done using an air classifier (Zhang et al. 2011; Lanzerstorfer 2015b, a), the centrifugal SPLITT method (Camerani et al. 2002) and sieving (Erdoğan and Türker 1998; Chindaprasirt et al. 2004; Itskos et

al. 2010). There is only one study (Camerani et al. 2002) concerning the classification of fly ashes from fluidized bed combustion although the results for other ashes indicate that method to be promising. For example, lead content in the coarse fraction could be reduced to 60% by separation of 20% fines (Lanzerstorfer 2015b). Therefore, this study aimed to establish whether the utilization of fly ashes from fluidized bed combustion of peat, forest residues and wastes can be improved by mechanical classification of sulfate, chloride and some detrimental metals as well as determine the effect of fly ash deagglomeration on classification selectivity. The research was carried out at the University of Oulu in Finland during the year 2015. The regulations referred in this paper were valid in 2017 when this manuscript was written.

2. Materials and Methods

2.1 Fly ashes

Fly ashes from two different Finnish power plants were studied. Both of the power plants have bubbling fluidized bed boilers. However, the power plants use different mixtures of fuels, and therefore the fly ash samples differed in properties. Fuel composition of the first fly ash sample (FA1) was 70% forest residues and 30% peat, while in the second fly ash sample (FA2) the composition was 60% forest residues, 30% recycling waste and 10% sludge from the papermaking industry. Fly ash samples were collected from the first electrostatic precipitator unit.

The chemical composition of both fly ashes was mainly CaO and SiO₂, but the fly ashes also contained high percentages of Al₂O₃ and Fe₂O₃ (Table 2). FA2 had a high sulfate content of 6.9 %. FA2 contained more detrimental elements than FA1, which is because FA2's power plant used recycling waste as fuel. Trace elements reported were cadmium (Cd), copper (Cu), lead (Pb) and zinc (Zn) since those elements were the most critical for FA1 and FA2 according to Finnish regulations (see Table 1). A huge difference between the ash samples can be seen: for FA2, Cd and Zn contents were four times higher, Cu content was 32 times higher and Pb content was 20 times higher compared to FA1. Both fly ashes had similar particle size: the median particle size for FA1 was 45.0 μm and 46.5 μm for FA2.

Table 2. Chemical and physical properties of studied fly ashes.

	FA1	FA2
Loss on ignition 950°C [%]	3.2	0.7
Particle size d ₅₀ [μm]	45.0	46.5
CaO [%]	16.0	23.0
SiO ₂ [%]	52.2	39.4
Al ₂ O ₃ [%]	11.0	12.1
Fe ₂ O ₃ [%]	4.8	4.9
Na ₂ O [%]	2.1	2.9
K ₂ O [%]	2.9	2.4
P ₂ O ₅ [%]	1.7	1.3
SO ₃ [%]	1.7	6.9
Cl [%]	0.1	0.4
Cd [mg/kg]	2.2	9.7
Cu [mg/kg]	58	1890
Pb [mg/kg]	26	510
Zn [mg/kg]	500	2210

2.2 Methods

2.2.1 Analysis of fly ashes

The particle size distribution of the fly ash samples reported as a volumetric median size (d_{50}) was measured with the laser diffraction technique (Beckman Coulter LS 13320) using the Fraunhofer model and the wet procedure using water. A specific surface area measurement was based on the physical adsorption of gas molecules on a solid surface using Micrometrics ASAP 2020, and the results were reported as BET isotherm. The main chemical components of fly ash were determined using an Omnia Pananalytix Axiosmax 4 kV X-ray fluorescence (XRF) from a melt-fused tablet. The melt-fused tablet was produced from 1.5 g of fly ash melted at 1150 °C with 7.5 g of X-ray Flux Type 66:34 (66% LiB_4O_7 and 34% LiBO_2). Trace elements were measured with inductively coupled plasma atomic emission spectroscopy (ICP-OES) from wet digested samples. The microwave-assisted wet digestion was performed using a 3:1 ratio of HNO_3 and HCl acid mixture for 0.5 g of fly ash at 175°C according to EPA3051A (United States Environmental Protection Agency 2007).

2.2.2 Low intensity grinding

The original fly ash samples FA1 and FA2 were ground by an impact mill with one rotating and one stationary pin disc (Laboratory Fine Impact Mill 100 UPZ-II from Hosokawa Alpine, Augsburg, Germany). The aim of the grinding procedure was not to reduce median particle size but only to break down the agglomerates formed during transportation and storing due to electrostatic forces. The pin mill was operated with the lowest possible rotational speed of 2000 rpm corresponding to a tip speed of 15 m/s in order to achieve the deagglomeration effect on the fly ash particles. After deagglomeration, two new samples for classification were achieved: FA1_G, and FA2_G. More information about the mill type used in this study can be found in (Salman AD et al. 2007).

2.2.3 Classification

Two different classification methods were used to fractionate FA1 and FA2 samples into fine and coarse fractions. The first method was an air jet sieving (e200 LS) with a 20 μm wire screen. Another method was using an air jet classifier (Multiprocessing System, Hosokawa Alpine, Augsburg, Germany), as presented in Fig 1. Classification rotor speed and air flow can be changed to adjust fine fraction share, and by using classification rotor speeds of 2000 and 4000 rpm and air flows of 75 and 85 m^3/h , fine material shares from 6 to 45 % were achieved. As an operational principle, the amount of coarse fraction increases when classifier rotor speed is increased and air flow is decreased. Parameters are still highly dependent on the material properties: particle size and shape affect classification results, and therefore each material before and after deagglomeration different classification parameters were studied. In both instances, the achieved coarse and fine fractions were collected for weighing and elemental analysis.

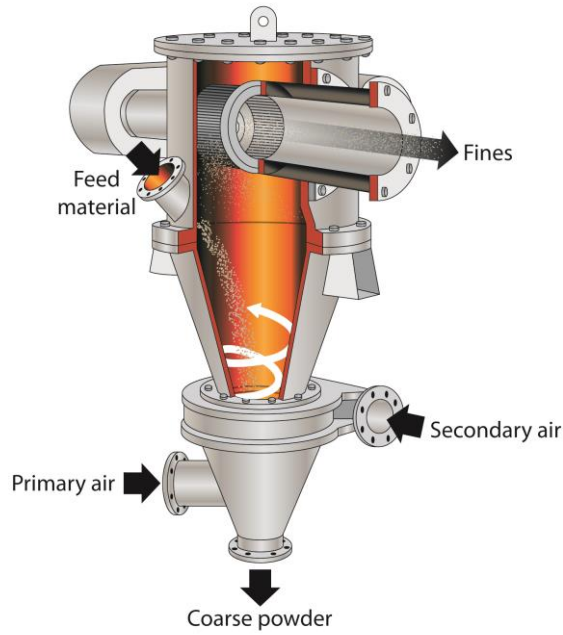


Fig 1. Classification procedure with an air jet classifier.

2.2.4 Selectivity calculations

Fine material share was calculated according to the general mass reject ratio:

$$RR_m = \frac{m_{Fi}}{m_F} \cdot 100, \quad (1)$$

where RR_m is fine material share [%] and m_{Fi} and m_F stand for the mass of fines and feed, respectively.

The calculation of removal efficiency was based on the following equation:

$$E_r = \frac{m_{Fi} \cdot x_{Fi}}{m_F \cdot x_F} \cdot 100 = \left[1 - \left(1 - \frac{RR_m}{100} \right) \cdot \frac{x_C}{x_F} \right] \cdot 100, \quad (2)$$

where E_r is the removal efficiency of a component [%], x is content of elemental analysis and C stands for coarse. The calculated removal efficiencies were plotted against the fine material share, and the selectivity curve introduced by Nelson (Nelson 1981) was fitted to them. The curve fitting was done by adjusting Nelson's selectivity index in the following equation so that the best fit was achieved:

$$E_r = \frac{RR_m/100}{1 - Q_N + Q_N \cdot RR_m/100} \cdot 100, \quad (3)$$

where Q_N is Nelson's selectivity index. Representative curves can be prepared with the Nelson's selectivity index, but the selectivity index shown this way is not uniform. A positive selectivity index ($0 < Q_N < 1$) refers to enrichment of the component to the fine, and the ideal selectivity is described as $Q_N = 1$. In the case of a split flow, $Q_N = 0$, the content values for coarse and fine are equal. A negative selectivity index ($-\infty < Q_N < 0$) is a result of component enrichment in the coarse fraction. To achieve uniform and infinite range ($0 < Q < 1$) for the selectivity index, Karnis' selectivity index (Karnis 1997) was used:

$$Q_K = \frac{x_1}{x_2} \quad (4)$$

where Q_N is Karnis' selectivity index and x_1 is content of elemental analysis given for the fraction in which the content lower and x_2 is for the fraction in which the content is higher. Karnis's selectivity index has a finite range but it does not indicate in which fraction (fine or coarse) a component has been enriched. The interdependence of Nelson's and Karnis' selectivity index is:

$$Q_K = 1 - \frac{1}{1 - Q_N} \quad (5)$$

3. Results and Discussion

3.1 Classification of main components

Air jet classification separates components into fine and coarse fractions. According to mass balance, when some elements concentrate on a fine fraction, some elements must concentrate on a coarse fraction. Fig 2 shows the classification results for the main components, where CaO can be seen to concentrate in a fine fraction with a moderate selectivity ($Q=0.55$), whereas SiO₂ and Al₂O₃ concentrated in a coarse fraction. Selectivity of SiO₂ was 0.50, but for Al₂O₃ the selectivity was not as clear. For FA1, there was no selectivity, and the content in fine and coarse fractions were the same, but for FA2, Al₂O₃ concentrated clearly in a coarse fraction with a selectivity of 0.5. These results comply with those of an earlier study (Zhang et al. 2011; [Ohenoja et al. 2018](#)).

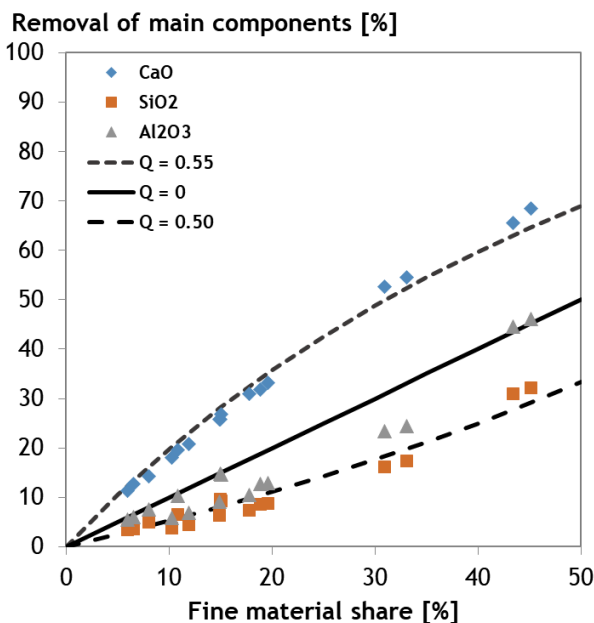


Fig 2. Fractionation of main components during fly ash classification. Flow split, i.e., when the content for fine and coarse fractions are equal, is indicated by a black line ($Q = 0$). At the upper side of this line, elements are concentrating into a fine fraction and at the underside of this line elements are concentrating into a coarse fraction.

The concentration of SiO₂ and Al₂O₃ in coarse and CaO in fine fractions is an advantage when concerning concrete applications according to SFS-EN 450-1 (EN 450-1 2012) since the sum of

Al+Si+Fe increases. However, for fertilizing and earth construction purposes this is a disadvantage since fertilizer should contain over 6% of Ca, and calcium is an important component of the self-hardening reactions (Illikainen et al. 2014) used in earth construction.

3.2 Classification of sulfate and chloride

Classification of sulfate and chloride into fine fractions can improve utilization of coarse fractions in concrete applications, and guidelines were taken from the European standard SFS-EN 450-1 (EN 450-1 2012). In other words, chloride content must be less than 0.1% and sulfate less than 3%. Typically, biomass fly ashes contain significant amounts of chloride and sulfate. It is suggested in the literature that the performance of fly ash–cement binders could be improved by the removal or control of sulfates and chlorides since the presence of these in cementitious materials can reduce durability through deterioration of the microstructure (Rajamma et al. 2009). When comparing the chemical composition of FA1 and FA2 (Table 2) to regulations (Table 1), it can be noticed that the sulfate content of FA1 is at an acceptable level but is remarkably high for FA2. The chloride content for FA1 is at the limit level and is also too high in FA2. However, these limits are only suggestions since the use of fluidized bed combustion fly ashes in concrete has not yet been standardized.

Sulfate selectivity to concentrate into a fine fraction is shown in Fig 3. In all cases, SO_3 enriches into a fine fraction with a high selectivity. At lower fine material shares (5%–25%), selectivity is around 0.60, whereas at higher fine material shares (30%–45%) selectivity is even higher $Q=0.80$. These results comply with those of an earlier study (Ohenoja et al. 2018). Deagglomeration had a negligible effect on classification of sulfate. For FA1 the difference was within experimental limits, but for FA2 the selectivity was the same.

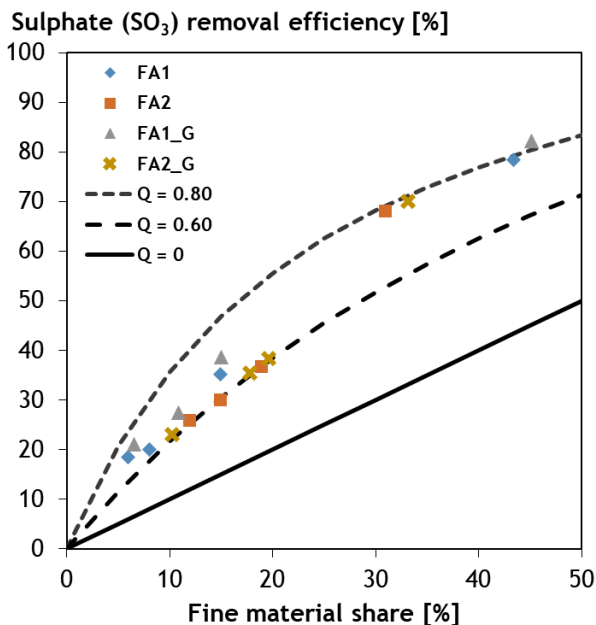


Fig 3. Fractionation of sulfate during fly ash classification. Flow split is indicated by a black line ($Q = 0$), and points above this line show concentration into fine fractions.

Chloride enriches into a fine fraction with a high selectivity (Fig 4). Chloride fractionates were distinctly different between the fly ash samples: FA1 gained only a moderate selectivity of 0.45, whereas FA2 acquired a selectivity of 0.80. This is most probably due to the difference in initial chloride content between the ashes: FA1 had a much lower initial amount of chloride than FA2 did.

These results comply with those of an earlier study (Ohenoja et al. 2018). Deagglomeration slightly improved the selectivity in both ashes. This refers to how chloride is most probably on the fly ash particle surfaces and therefore improved selectivity was obtained after deagglomeration.

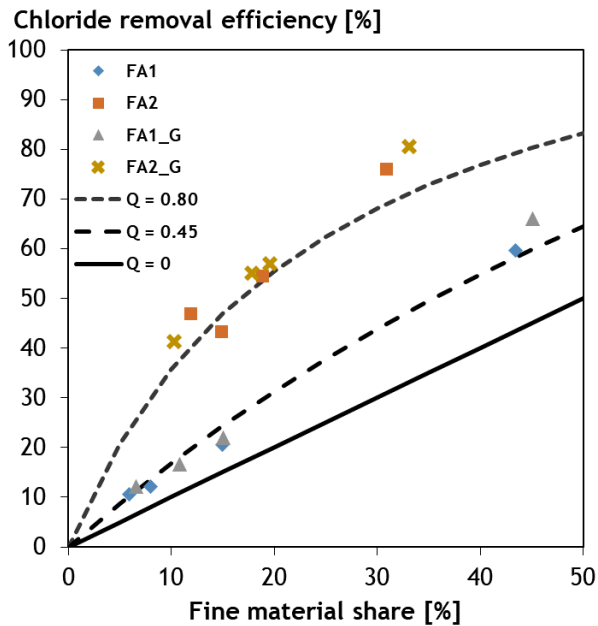


Fig 4. Fractionation of chloride during fly ash classification. Flow split is indicated by a black line ($Q = 0$), and points above this line show concentration into fine fractions.

3.3 Classification of detrimental metals

Fly ash utilization in earth construction, soil stabilization or fertilizing is limited by regulations, which set minimum concentration values for detrimental metals, as shown in Table 1. Elements reported here include Cd, Cu, Pb and Zn since these elements were the most critical for the studied fly ashes according to Finnish regulations presented in Table 1. FA1 originated from combustion of peat and forest residues and can therefore be used as fertilizer. Since the contents of detrimental elements were rather low (Cd is near the limit in the FA1 sample studied here), FA1 could even be used as a field fertilizer. However, since FA2 is originated partly from combustion of recycled waste, its utilization as a fertilizer is impossible (Finnish Ministry of Agriculture and Forestry 2011), but earth construction is a potential area where FA2 can be used. However, in the original FA2, copper, lead and zinc contents were higher than the limit values for earth construction.

All the detrimental metals examined in this study enriched in fine fractions. Cadmium enriched into a fine fraction with a high selectivity (Fig 5). FA1 had lower selectivity, $Q=0.68$, at a lower fine material share (from 5 to 20%) and higher selectivity, $Q=0.77$, at a higher fine material share (45 %). FA2 had higher selectivity, $Q=0.77$, and selectivity was not influenced by fine material share. Deagglomeration slightly improved the selectivity of FA1. For both studied fly ashes, cadmium content was below earth construction (15 mg/kg) and forest fertilizer (25 mg/kg) limit.

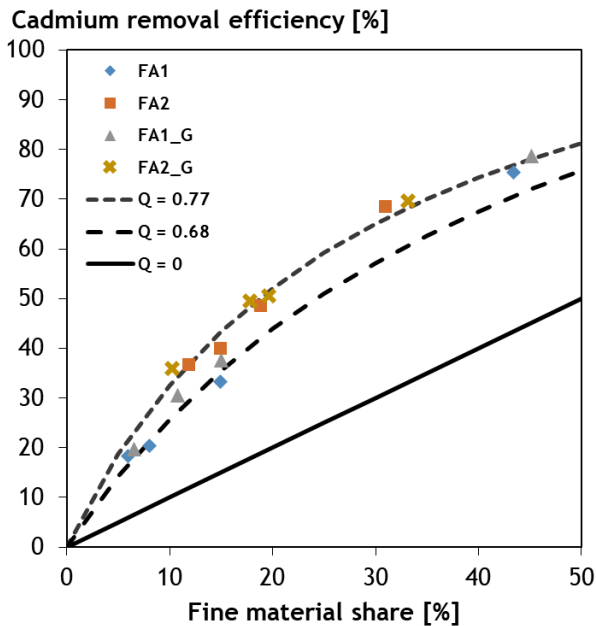


Fig 5. Fractionation of cadmium during fly ash classification. Flow split is indicated by a black line ($Q = 0$), and points above this line show concentration into fine fractions.

Copper removal efficiency was dependent on fuel type (Fig 6). In all cases, copper enriched into a fine fraction. FA1 had higher selectivity, $Q=0.65$, than FA2, $Q=0.35$, regardless of fine material share. Deagglomeration slightly improved the selectivity of FA2. Overall, the copper content of FA2 was a challenge: even by removing 33% fine material, the copper content was still over the limit for earth construction (1620 mg/kg vs 400 mg/kg). Therefore, one option for future study is to utilize this type of fly ash in alkali activation to stabilize heavy metals (Yliniemi et al. 2015).

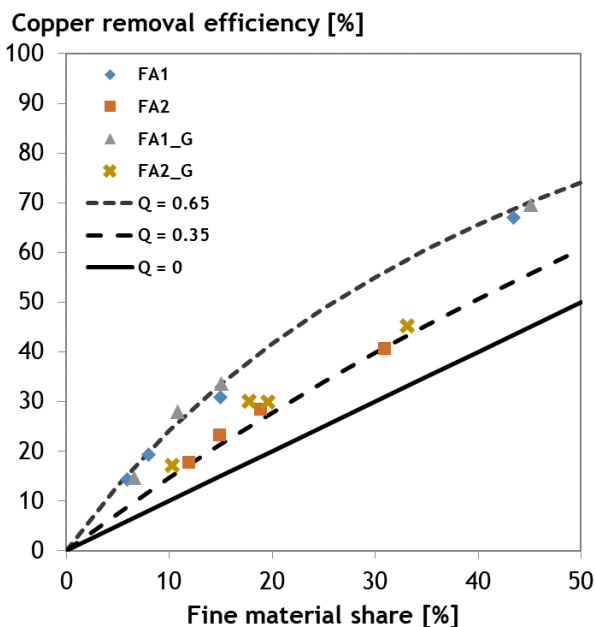


Fig 6. Fractionation of copper during fly ash classification. Flow split is indicated by a black line ($Q = 0$), and points above this line show concentration into fine fractions.

Lead removal efficiency was good for both fuel types (Fig 7). FA1 had a lower selectivity, $Q=0.67$, at a lower fine material share (from 5 to 20%) and a higher selectivity, $Q=0.75$, at a higher fine material share (45%). FA2 had a selectivity between $Q=0.67$ and $Q=0.75$ in all fine material shares

studied. Deagglomeration improved the selectivity of both ashes. For FA2, by removing 30% fine material using air jet classification, lead content under the earth construction limit (300 mg/kg) was achieved. This result is in well-accordance with results of an earlier study (Lanzerstorfer 2015b).

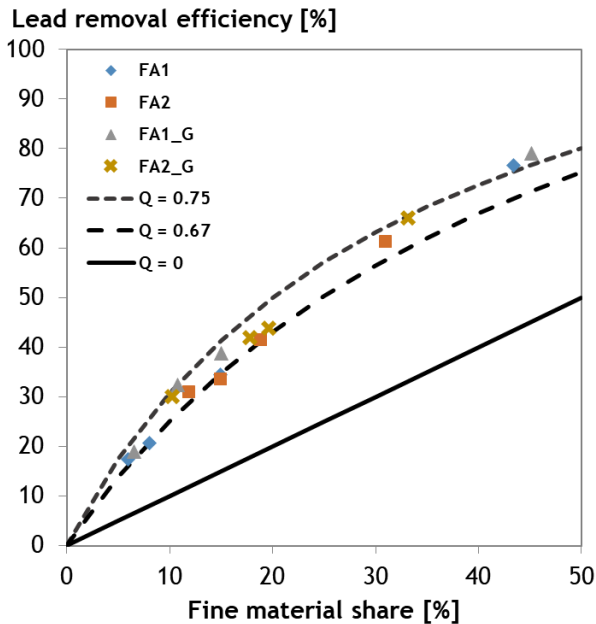


Fig 7. Fractionation of lead during fly ash classification. Flow split is indicated by a black line ($Q = 0$), and points above this line show concentration into fine fractions.

Excellent removal efficiency was also obtained for zinc (Fig 8), which enriches into a fine fraction with high selectivity. FA1 had a selectivity of $Q=0.60$ in all the fine material shares studied, and deagglomeration evidently improved the selectivity at fine material shares of 8 and 18%. FA2 had a selectivity between $Q=0.65$ and $Q=0.70$ at all studied fine material shares. Deagglomeration improved the selectivity of FA2 at a lower fine material share, but it seems that deagglomeration worsened the selectivity at high fine material shares (30%). For FA2, by removing 10% of the fine material, a zinc content under the limit for earth construction (2000 mg/kg) can be achieved.

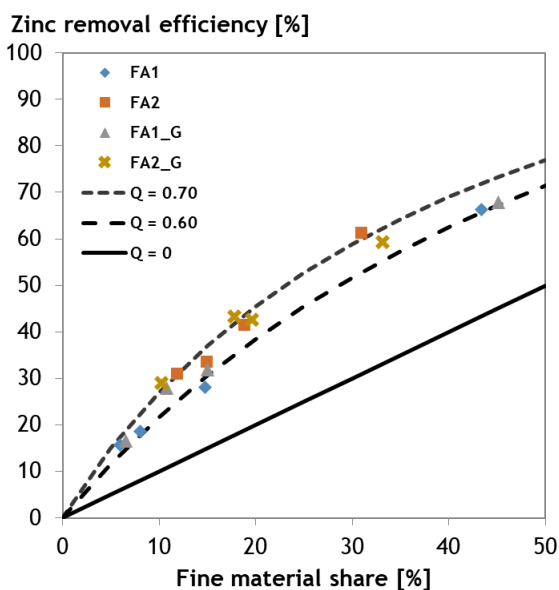


Fig 8. Fractionation of zinc during fly ash classification. Flow split is indicated by a black line ($Q = 0$), and points above this line show concentration into fine fractions.

As shown, air jet classification is an effective method to separate detrimental elements from fly ashes originating from fluidized bed combustion with different fuel compositions. Air jet classification is especially recommended for fly ashes that will be used in earth construction and concrete applications since CaO concentrates into fine fractions, which is not desired in fertilizer applications. However, high calcium fly ashes may contain so much calcium that even after classification the calcium content is at acceptable level. The removal efficiency of detrimental components was studied using Nelson's and Karnis' selectivity indices, which have been used previously in screening, cleaning, fractionation and flotation studies (Nelson 1981; Karnis 1997; Hautala et al. 2009; Körkkö 2012). These are powerful tools, as they point to component removal independent of the mass reject ratio, whereas cleanliness and removal efficiency, for example, are dependent on the mass reject ratio. According to the results of this study, low intensity grinding having only a deagglomeration effect is not a necessary to improve classification selectivity since it has only minor effect to selectivity results. In the future, higher intensity grinding to reduce median particle size can be tested.

4. Conclusions

The studied fly ashes differed in their fuel composition, and therefore, in their chemical composition. FA2 contained much more detrimental metals, including sulfate and chloride, since it originated from a plant that uses recycling waste as 30% of its fuel. However, both ashes behaved similarly in the air jet classification process. CaO ended up in fine fractions and SiO₂ and Al₂O₃ ended up in coarse fractions. Sulfate, chloride and the studied detrimental metals (cadmium, copper, lead and zinc) enrich into fine fractions with a high selectivity. If 30 mass-% is removed as a fine fraction from FA2, sulfate and chloride contents are under concrete limits (<3% and 0.1%, respectively) and, cadmium, lead and zinc contents are below earth construction limits (15 mg/kg, 300 mg/kg and 2000 mg/kg, respectively). The content of detrimental elements in FA1 were originally low, nevertheless, it behaved accordingly to FA2 in air jet classification. Overall, the results of this study suggest it is possible to significantly improve the utilization potential of fly ashes in concrete, fertilizer and earth construction applications by using air jet classification.

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