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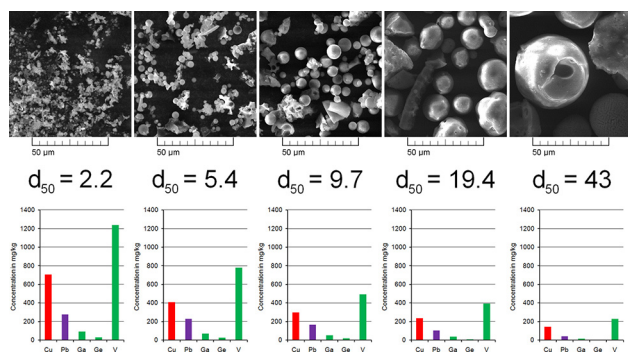
Fly ash from coal combustion: Dependence of the concentration of various elements on the particle size



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GRAPHICAL ABSTRACT



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ABSTRACT

The dependence of the distribution of various elements on the particle size of coal combustion fly ash is of special interest for use of the fly ash in ultrafine cement, or for recovery of valuable elements from fly ash. Therefore, electrostatic precipitator fly ash from a coal fired power plant was split into particle size classes with mass median diameters of 2.2 μm , 5.4 μm , 9.7 μm , 19 μm and 43 μm by means of an air classifier. The size dependence for the elements Al, As, Ba, Br, Ca, Ce, Cl, Co, Cr, Cu, Fe, Ga, Ge, Hg, K, Mg, Mn, Mo, Na, Nb, Ni, P, Pb, Rb, S, Sb, Se, Si, Sn, Sr, Ta, Ti, Th, Tl, U, V, W, Y, Yb, Zn, Zr, and for total carbon, was determined. For several components a considerable dependence of the concentration on the particle size was observed, which can be approximated by power functions. Substantially higher concentration with decreasing particle size was found for total carbon, As, Cu, Ga, Ge, Hg, Sn, Pb, Tl, V and Zn, whereas the concentration of the main mineral components Si, Al, Fe, Ca, K, Na, Mg, Mn and Ti was slightly lower in the fine particles. The increased concentrations have to be considered for critical elements like Pb and Cu when air classified ultrafine fly ash is used in ultrafine cement. Air classification might also be an important pre-processing step in a recovery process for trace elements like Ga, Ge and V.

1. Introduction

In the combustion of coal most of the mineral content remains in the form of ash. The coarse fraction of the ash is discharged at the bottom of the furnace, while the fine fly ash fraction has to be separated from the

combustion off-gas. The proportion of the fly ash in the total amount of ash depends on the type of combustion process and the operation conditions. In pulverized coal combustion the fraction of fly ash is typically three quarters. The current annual production of coal fly ash worldwide is estimated at around 500 million tones [1]. This huge

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amount of fly ash, released throughout the world by thermal power plants and other combustion processes using coal as fuel, should be utilized as much as possible because the disposal of such a large quantity of fly ash has become a serious environmental problem. The present-day utilization of fly ash varies widely from country to country. The world average utilization amounts to approximately 16% of the total fly ash. A substantial quantity of fly ash is still disposed of in landfills at a significant cost [1].

The composition of coal fly ashes has been studied considerably [2–8]. The content of carbon in coal combustion fly ash varies widely and depends on the conditions during combustion. Values for the loss on ignition (LOI) are reported from less than 1% to higher than 20% [2,3] [4,5]. Besides the main mineral components Si, Al, Fe and Ca, fly ash also contains some heavy metals and trace elements including As, Cr, Cu, Ga, Ge, Ni, Pb, Rb, Sn, V and Zn. For both the particle size and the particle size distribution of coal fly ash measured results are available in the literature. Mass median diameters d_{50} between 6.8 and 98 μm were reported [3–5], while the maximum particle size is approximately 150–250 μm . The distribution of various elements in size fractions of fly ash obtained by sieving has also been investigated. Classification by sieving is limited in particle size, thus the finest size fraction was 0–20 μm [9], 0–25 μm [10], 0–30 μm [11] and 0–38 μm [12]. However, in the first three studies only the main components were investigated, but neither heavy metals nor trace elements. In the fourth study the focus was on rare earth elements only.

Utilization of coal fly ashes in the cement industry is quite feasible. There are essentially two applications for fly ash in cement and concrete: firstly, the replacement of cement in Portland cement concrete and, secondly, as pozzolanic material in the production of pozzolanic cements. The utilization of fly ash in concrete is regulated, for example in the standard DIN EN 450-1 “Fly ash for concrete - Part 1: Definition, specifications and conformity criteria.” [13], while in the standard DIN EN 197-1 “Cement - Part 1: Composition, specifications and conformity criteria for common cements,” [14] the maximum permissible content of fly ash in various cement qualities is established.

In some fly ashes the carbon content is too high for utilization of the fly ash as a concrete additive. The maximum allowable value of the LOI according to DIN EN 450-1 lies between 5.0% and 9.0% depending on the category [13]. For fly ashes with a higher content of unburned carbon and therefore a higher LOI, beneficiation processes can be applied for the reduction of the carbon content. Increased values of the LOI are typically found in fly ashes from combustion with low NO_x production [15,16]. A process used for separation of unburned carbon from the mineral-rich fraction is dry triboelectrostatic beneficiation [17,18] [19]. Some other methods tested for fly ash beneficiation are ultrasonic column agglomeration, flotation and sieving [18,20,21].

Cement as well as fly ash contains some heavy metals which can be present in different compounds. In one study the leachability of some heavy metals with water from hardened fly ash cement and hardened cement was compared [22]. It was found that the leachability of Cu and Pb was substantially higher from the concrete containing fly ash, although the Pb content of the cement was higher compared to the Pb content of the fly ash and the Cu content of the cement, whereby the fly ash was the same. In other leaching tests with fly ash containing concrete, oxyanion-forming elements, especially As and Sb, were found to be critical [23,24].

Ultrafine cement is used for the production of very high performance concrete. It is characterized by an extremely fine grain size with a d_{50} of approximately 5 μm . The addition of ultrafine fly ash, preferably with spherically shaped particles, helps to improve workability of the concrete paste [16,25]. Fly ash with the required grain size can be produced in two ways: the one method is known as “stripping”, whereby the ultrafine fraction is extracted from the fly ash by means of air classification. The capacity of this method is limited because only part of the fines might be extracted without seriously reducing the quality of the remaining coarse product. The second method requires an

Table 1
Data of the size fractions produced.

	Mass fraction in %	Mass median diameter d_{50} in μm	d_{10} in μm	d_{90} in μm
Fly ash	–	17.6	3.4	30
Size fractions				
1	7.3	2.2	0.61	5.9
2	13.5	5.4	2.5	13.2
3	20.3	9.7	5.2	22
4	35.3	19.4	9.6	39
5	23.6	43.2	25	77

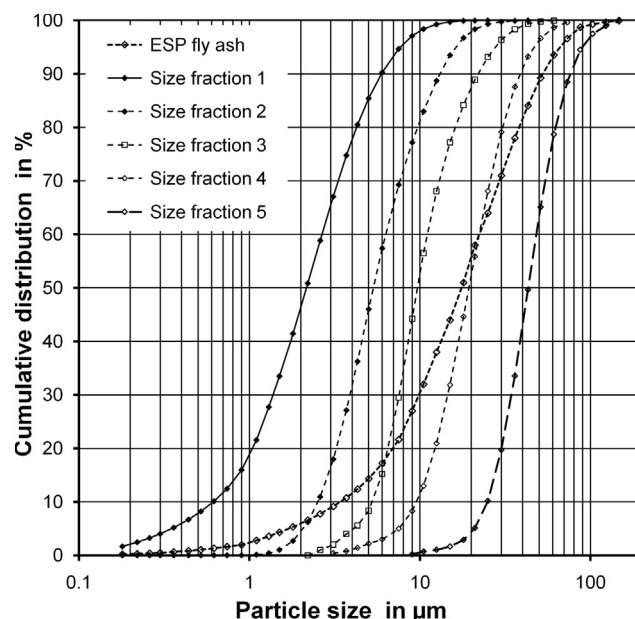


Fig. 1. Particle size distribution of the ESP fly ash and the dust size fractions produced.

ultrafine milling step which, on the one hand, increases production capacity of ultrafine fly ash, but on the other hand causes significantly higher power consumption [26]. In one study, high volume ultrafine fly ash cement composites were investigated. For this purpose, raw fly ash with a mean particle size of 15 μm was ground to produce ultrafine fly ash of a mean particle size of 8.1 μm [27]. In other studies, fly ash from coal combustion was classified by means of air classification to obtain ultrafine fly ash for special applications [25,28–31]. Generally, the finer the grain size, the higher is the strength of the concrete produced. A comparison of the use of ultrafine fly ash in concrete produced by air classification and by milling showed comparable results [16]. Both fly ashes improved the compressive strength and fluidity to a similar extent. However, the grinded fly ash had a substantially smaller particle size and the particle shape of the two ultrafine fly ashes was different. During milling the shape changes from round to angular, while classification does not affect the particle shape. In contrast, when the fly ash is milled the chemical composition of the ultrafine fly ash is identical to the average composition of the original fly ash, whereas in the case of extraction of the ultrafine fraction by air classification the chemical composition of this fraction might be different. However, only few data about the varying composition of coal fly ash with the particle size are available [9–12].

Besides the utilization of coal combustion fly ash in the cement industry, the fly ash can also be used for several other applications: for example, in soil amelioration [8,32], as base course in road construction [8], as mine-filling material for reclamation of abandoned mines [1,8], and for the solidification and stabilization of hazardous wastes

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