



Impact of solid fuel combustion technology on valence speciation of chromium in fly ash



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HIGHLIGHTS

- Biomass and high combustion temperature favor enrichment of fly ash with Cr(VI).
- Biomass combustion favor enrichment of fly ash with water soluble Cr(VI).
- Alkali compounds and Fe oxides play a key role in shaping Cr(VI/III) speciation.
- Cr(VI) level in coal fly ash drops in order: PCC boiler \gg FBC boiler \approx SF boiler.
- An influence of combustion technologies on Cr speciation in fly ash was explained.

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ABSTRACT

Fly ash (FA) generated in real furnaces was used to evaluate the impact of the kind of the solid fuel burnt and combustion technology on chromium speciation, particularly the presence of Cr(III) and Cr(VI) forms as well as readily and hardly leachable chromium(VI) species in FAs. The FAs originated from a pulverized coal combustion boiler (PCC boiler), a fluidized bed combustion boiler (FBC boiler), a stoker-fired boiler (SF boiler), a municipal solid waste incinerator (MSWI), a cement rotary kiln (CRK) and a modern domestic boiler (DB). The speciation analysis of chromium was carried out by means of extraction followed by catalytic cathodic stripping voltammetry with adsorption of Cr(III)-DTPA complexes (CCSV-DTPA) for determination of Cr(VI) and AAS was used for determination of Cr content.

It has been revealed that the antagonistic action of alkali metal compounds and iron oxides plays a crucial role in shaping valence speciation of chromium. According to the proposed transformation path of oxidation of Cr(III) to Cr(VI), hard coal combustion in an SF boiler, an FBC boiler or a domestic boiler will generate FAs with a low Cr(VI) level. Replacing fuel with biomass should create favorable conditions for generating FA enriched with Cr(VI). Relatively high concentrations of Cr(VI) can also be expected in FA generated in the process of high-temperature combustion of coal in PCC boilers.

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1. Introduction

Chromium speciation in fly ash has been studied for a number of years and were focused on the mode of chromium occurrence as well as the mechanism of chromium transformation in a combustion chamber. The determination of chromium(III) and chromium(VI) has been carried out directly in a FA sample by using non-destructive techniques [1–5] or indirectly by extraction followed by determination of released chromium species [2,6–8]. The interest in chromium valence speciation in FA stems from a significant difference in toxicity and bioavailability of the

Cr(III)- and Cr(VI)-species. Cr(III) is a bioelement, whereas particulate Cr(VI) is regarded as a factor which increases the severity of ongoing allergic asthma and alters its phenotype [9]. The environmental impact and health effects are related to chromium(VI) load in the environment.

The scale of the problem is large, although it cannot be determined accurately, as Cr(VI) emission is inventoried only in relatively few countries. Within the last twenty years the EU (15) countries produced annually 40 Tg of FA on average, of which 92% was utilized in 2006. The construction industry and underground mining used 20 Tg (concrete addition – 26.7%, cement raw material – 25.8%, road construction and filling applications – 22.5%, blended cement – 11.3%, concrete blocks – 7.1%, infills – 4%, and others – 2.6%) [10]. The remaining FA was used in land reclamation and

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restoration. In that same year in the USA 72 Tg of FA was generated, of which 32 Tg was utilized (the construction industry – 60% and mining and earthworks – 40%) [11]. If we assume that 3 mg/kg is the expected value of Cr(VI) content in FA [2,4,7,8,12] and that utilization of only a half of the FA generated creates conditions for Cr(VI) release, the environment in Europe is loaded annually with 60 Mg of Cr(VI). Although this value is high, it seems to be correct. For comparison, total annual emissions of Cr(VI) in the US were 44 Mg in 2002 [13]. Proportionally lower Cr(VI) emissions to air were found in the UK – 28.6 Mg/year (2008) [14], Australia – 2.5 Mg/year (2011) [15] and Canada – 1 Mg/year (2010) [16].

In order to reduce the Cr(VI) pollution burden on the environment, it is crucial to control oxidation of Cr(III) to Cr(VI) taking place during combustion of solid fuels in such a way that it is possible to reduce the Cr(VI) content or, at least, to limit Cr(VI) elution from FA.

Although Cr valence speciation in various coals and coal combustion products has been covered quite extensively in the literature, and moreover recently a number of papers have been published in which the authors proposed a mechanism of possible chromium transformations [3,5,6,17,18], there is still a lack of a general approach which would explain the essence of shaping Cr speciation in FA generated in real furnaces based on various combustion technologies.

The aim of our work is to examine the speciation of Cr(III/VI) and speciation of Cr(VI) (water leachable and hardly leachable fractions) in FAs depending on which combustion technology of solid fuel has been used. FAs produced in pulverized fuel boilers, stoker-fired boilers, a fluidized bed boiler, an up-draught combustion

domestic boiler, a Krüger grate furnace and a cement rotary kiln have been investigated. An attempt has been made to prepare an outline of a chromium transformation model in real solid fuel fired boilers which may explain the differences in chromium valence speciation observed in the generated FAs.

2. Experimental

2.1. Sampling and samples preparation

FA samples (1 kg) were collected from various parts of dust arresting systems. Pulverized fuel combustion FA samples were collected from each of the three fields of an electrostatic precipitator system. Both units, in which hard coal was fired and biomass was co-fired ($C/B = 9:1$), were of the same type and worked in the same coal-fired power plant. The samples were denoted as PCC/FA-I/1-3 and PCC/FA-II/1-3 for coal and coal/biomass combustion, respectively (Table 1). The stoker-fired coal fly ash samples were collected from collection tanks of the multicyclone and a battery of cyclones in two heating plants (I and II) equipped with the same type of stoker-fired boilers. The samples were denoted as SF/FA, 1 – multicyclone and 2 – battery of cyclones and the subscripts *a* and *b* stand for fraction $>63 \mu\text{m}$ (coarse fraction) and $\leq 63 \mu\text{m}$ (fine fraction), respectively. Three FA samples denoted as FBC/FA-1, FBC/FA-2 and FBC/FA-3 were collected from the ash hopper of the ESP in the FBC boiler system which was supplied with hard coal from three coal mines, one from Lublin Basin (1) and two from Upper Silesian Basin (2 and 3). In this installation dry limestone was injected into the flue gases before the collection of particulate

Table 1
Kind of furnace, fuel, dust collection device and sampling points.

No	Fly ash sample	Furnace	Fuel	Temperature of burning zone	Dust collection device	Sampling point ^a /remarks	
1	PCC/FA-I/1	Pulverized-fuel boiler	Hard coal energy value – 22 MJ/kg, ash – 20%, sulfur – 1.1%. Particle size <0.1 mm. Hard coal (ditto) + wood waste (10%) energy value of biomass – max. 18 MJ/kg	1800–1935 °C	ESP	1st-field ESP	
2	PCC/FA-I/2					2nd-field ESP	
3	PCC/FA-I/3					3rd-field ESP	
4	PCC/FA-II/1					1st-field ESP	
5	PCC/FA-II/2					2nd-field ESP	
6	PCC/FA-II/3					3rd-field ESP	
7	SF/FA-I/1a	Stoker-fired boiler	Hard coal energy value – 23.2 MJ/kg, ash – 15.1%, sulfur – 0.63%, moisture content – 11.8%	1100 °C	1. Multicyclone	$\varphi > 63 \mu\text{m}$ (42%)	
8	SF/FA-I/1b					$\varphi \leq 63 \mu\text{m}$ (58%)	
9	SF/FA-I/2a					2. Cyclone	$\varphi > 63 \mu\text{m}$ (52%)
10	SF/FA-I/2b					$\varphi \leq 63 \mu\text{m}$ (48%)	
11	SF/FA-II/1a	Stoker-fired boiler	Hard coal energy value – 21.9 MJ/kg, ash – 19.5%, sulfur – 0.62%, moisture content – 10.2%	1100 °C	1. Multicyclone	$\varphi > 63 \mu\text{m}$ (40%)	
12	SF/FA-II/1b					$\varphi \leq 63 \mu\text{m}$ (60%)	
13	SF/FA-II/2a					2. Cyclone	$\varphi > 63 \mu\text{m}$ (57%)
14	SF/FA-II/2b					$\varphi \leq 63 \mu\text{m}$ (43%)	
15	FBC/FA-1	Fluidized-bed boiler	Hard coal energy value – 21.1 MJ/kg, ash – 21.1%, sulfur – 1.04%	850–865 °C	ESP preceded by dry limestone injection	Collection tank	
16	FBC/FA-2					Hard coal energy value – 22.1 MJ/kg, ash – 21.4%, sulfur – 0.52%	
17	FBC/FA-3					Hard coal energy value – 22.9 MJ/kg, ash – 20.7%, sulfur – 0.42%	
18	DB/FA-1	Up-draught combustion domestic boiler	Pine wood briquette		Lack	Dead space of flue	
19	DB/FA-2						Birch plywood briquette
20	MSWI/FA	Krüger grate furnace (incinerator)	Sorted municipal wastes	1000–1100 °C	Bag filter preceded by hydrated lime injection		
21	CRK/FA-1	Cement rotary kiln	Hard coal energy value – 27 MJ/kg, ash – 7.0%, sulfur – 1.2%, moisture content – 8.5% + alternative fuel/wood waste	Max. 2000 °C	1. Dust chamber 2. Two parallel bag filters	1st bag filter	
22	CRK/FA-2						
23	CRK/FA-3					2nd bag filter	

^a If different than the one given in the dust collection device column.

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