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Chemical speciation and leaching characteristics of hazardous trace elements in coal and fly ash from coal-fired power plants



Shilin Zhao^{a,b}, Yufeng Duan^{a,*}, Jincheng Lu^a, Rajender Gupta^b, Deepak Pudasainee^b, Shuai Liu^a, Meng Liu^a, Jianhong Lu^a

^a Key Laboratory of Energy Thermal Conversion and Control of Ministry of Education, School of Energy and Environment, Southeast University, Nanjing 210096, China

^b Department of Chemical and Materials Engineering, University of Alberta, Edmonton T6G 1H9, Canada

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ABSTRACT

This work reports the chemical speciation and leaching characteristics of seven hazardous trace elements (HTEs, including Hg, As, Cr, Cd, Ba, Mn, and Pb) in the coal and fly ash samples collected from four coal-fired power plants in China. The physical structure and chemical composition of the fly ash were characterized by the scanning electron microscope (SEM) and the energy dispersive X-ray spectrometer (EDX). The chemical speciation of HTEs in the coal and fly ash was measured by the modified three-step sequential extraction method, proposed by the European Community Bureau of Reference (BCR). Leaching characteristics of HTEs in the fly ash were investigated by a single batch leaching test. The concentration of HTEs in solid and liquid samples was determined by the direct mercury analyzer DMA 80 (Hg in solid) and the inductively coupled plasma-mass spectrometry (ICP-MS). Results show that for the coal, concentration of Hg, As, Cr, Cd, Ba, Mn, and Pb is 0.06–0.22, 0.63–4.01, 8.91–13.09, 0.06–0.15, 108.67–229.21, 49.94–100.24 and 6.74–26.38 mg/kg, respectively. Mercury is mainly in the residual form while Cd and Ba are primarily in reducible form. Compared with other HTEs, manganese in water/acid soluble and exchangeable fraction has the large percentage. For the fly ash, the concentration of Hg, As, Cr, Cd, Ba, Mn and Pb is 0.17–1.26, 5.15–25.74, 43.25–64.61, 0.56–0.70, 777.05–970.70, 163.83–831.47 and 28.94–119.57 mg/kg, respectively. Mercury and chromium are mainly in the residual speciation. Arsenic and manganese in water/acid soluble and exchangeable form have high ratio with value of 7.27–58.60% and 6.14–62.27%, respectively. Cadmium and barium are primarily in the reducible form. Based on the risk assessment code, manganese in the coal can pose high or very high risk on the environment. Leaching concentration of Cr in some fly ash is higher than permissible limits and the pH value of leaching solution for the fly ash is alkaline. Considering huge fly ash production from coal combustion and complex landfill conditions, some suitable disposal measures to minimize the risk on the environment are needed.

1. Introduction

With increasing poisoning accidents related with hazardous trace elements (HTEs) happened in recent years, the adverse effects of HTEs on human health and eco-environment are emphasized [1–4]. The U. S. Clean Air Act Amendments, European Union and the Canadian Environmental Protection Agency have listed some HTEs, such as Hg, As, Pb, etc., as the prime environmental concerns [5–7]. Coal combustion for power generation is considered as one of the major emission sources of HTEs [8–11]. The field tests on coal-fired power plants [12–15] have shown that the most of HTEs are mainly distributed in fly ash removed from the electrostatic precipitator (ESP) or electric fabric filter (EFF). With huge electricity generation on the global scale, a considerable

amount of fly ash, such as India: 112 Tg/y, China: 100 Tg/y, USA: 75 Tg/y, Germany: 40 Tg/y, UK: 15 Tg/y and Poland: 4 Tg/y, are produced [16]. It has been reported that 40–100% of the fly ash is mainly reused, such as in concrete and cement manufacturing; while the rest is disposed of in lagoons and landfills [17–19]. If not properly disposed, the HTEs can be leached out from the fly ash and may contaminate soil, groundwater, and surface water [20–22], which has caused widespread concern worldwide, especially in the USA and Europe [23–28].

Lots of researchers have investigated the leaching behavior of HTEs in fly ash. Gong et al. [29] used column leaching tests to study the leaching behavior of HTEs in a typical ash dump in Guizhou province, China. Results showed that concentration of Cd remained stable after 16 days, but Zn was leached out significantly in the short-time

* Corresponding author.

E-mail address: yfduan@seu.edu.cn (Y. Duan).

compared to Cd and Pb. Jones et al. [30] adopted synthetic ground-water leaching procedure (SGLP) and long-term leaching (LTL) to explore the leaching characteristics of HTEs (Al, As, Ba, Cr, Mn, Pb, etc.) in bottom ash, economizer fly ash and fly ash. Most HTEs were found to be leached out from fly ash compared to the mill rejects and bottom ash. In addition, concentration of most HTEs in the leachates increased with increasing SGLP and LTL leaching duration. Dutta et al. [31] investigated the effects of pH and solid-liquid ratio on the leaching of HTEs (Mn, Cr, As, Pb, etc.) in four fly ash samples from different coal-fired power plants in India, result of which indicated that much higher mobility of the elements was observed at low pH while leachability of Cr decreased with increasing solid-liquid ratio from 0.05 to 0.20. Sandeep et al. [32] studied leaching behavior of HTEs (As, Se, Cr, Pb, etc.) in two fly ash samples at various pH, leachate solution temperatures and leaching methods, revealed that As had the highest leaching rate (1.5 and 2.4 $\mu\text{g/g}$) in the two fly ash samples while leachate solution temperature increased metal's leachability. In addition, most of the metal's leachability in toxicity characteristic leaching procedure (TCLP) was higher than that in batch leaching tests. It can be found that all these studies were focused on the influence of leaching methods, pH, solid-liquid ratio, leaching duration and leachate solution temperatures on the mobility of HTEs in fly ash. In fact, the chemical speciation of HTEs in fly ash is a very important internal factor for determining the leaching behavior. However, the studies related to this are very few [16,33]. In addition, it is of great importance to know the chemical forms of HTEs in coal due to the increasing coal-washing rate which rose from ~22% to ~60% since 2002 in China [34]. The knowledge on chemical form of HTEs in coal can help to predict the removal efficiency of HTEs from coal and concentration of HTEs in the wastewater during coal washing.

Based on the deficiencies in the previous studies, the chemical speciation and leaching characteristics of HTEs in fly ash were investigated in this work, which also included the occurrence of HTEs in coal. The coal and the corresponding fly ash (removed from ESP or ESP + FF (fabric filter)) samples were sampled from four different coal-fired power plants in China. The sequential extraction procedure proposed by the European Standard, Measurements and Testing (SM&T) program, formerly the Community Bureau of Reference (BCR), was applied to obtain the chemical form of HTEs in the coal and fly ash. The scanning electron microscope (SEM) equipped with an energy dispersive X-ray spectrometer (EDX) was adopted to characterize the physical and chemical properties of the fly ash surface. A single batch leaching test was used to acquire the leaching characteristics of HTEs in the fly ash. The main contents include as follows: (1) Concentration and chemical speciation of HTEs in coal; (2) Morphological features and elemental composition of fly ash; (3) Concentration and chemical speciation of HTEs in fly ash; (4) Risk assessment code; (5) Leaching characteristics of HTEs in fly ash. The main objective is to provide a reference for the subsequent disposal of fly ash and coal washing.

2. Experimental section

2.1. Sample collections

The detail information of the four studied coal-fired power plants in China is shown in Table 1. The coal and the corresponding fly ash (removed from ESP or ESP + FF) samples were collected from the four power plants. The ash in each field of ESP or ESP + FF would be collected and transported into the ash hopper by air or plunger pump, in this process ash is mixed uniformly. To ensure the ash samples be representative, the fly ash was taken from the ash hopper. During the field sampling, the coal and fly ash samples were collected from power plants every thirty minutes. The whole sampling time for the coal and fly ash lasted for about two hour. The samples collected every thirty minutes were mixed thoroughly and evenly for representativeness. The coal and fly ash samples corresponding to the four power plants were

Table 1

The detail information of the four studied coal-fired power plants.

| No. | Output | APCDs | Location |
|---------|--------|----------------------------|----------------------------------|
| Plant-1 | 660 MW | SCR + ESP + WFGD + WESP | Dingzhou City, Hebei Province |
| Plant-2 | 350 MW | SCR + (ESP + FF) + WFGD | Nantong City, Jiangsu Province |
| Plant-3 | 100 MW | SCR + ESP + WFGD | Zibo City, Shandong Province |
| Plant-4 | 320 MW | SCR + ESP + WFGD | Zhangjiakou City, Hebei Province |

Note: APCDs: Air pollution control devices; SCR: Selective catalytic reduction; ESP: Electrostatic precipitator; WFGD: Wet flue gas desulphurization; WESP: Wet electrostatic precipitators.

defined as Coal-1, Coal-2, Coal-3, Coal-4, and FA-1, FA-2, FA-3, FA-4, respectively.

2.2. Chemical speciation and HTEs concentration determination

The modified three-step sequential extraction method, proposed by the European BCR [35] was used to acquire the chemical speciation of HTEs in the coal and fly ash in this study. The content of HTEs in the sample was divided into four fractions, as shown in Table 2. The preparation method for the solution listed in the table was same as that in Nemati et al. [36]. Concentration of HTEs in the extracting solution after each step was determined by the inductively coupled plasma-mass spectrometry (ICP-MS, 7700x, US Agilent Technologies Co., Ltd.). For HTEs in solid samples (coal, fly ash and the residue at step 4), content of Hg was determined by the direct mercury analyzer DMA 80 (Milestone GmbH, Germany). The concentration of other HTEs (As, Cr, Cd, Ba, Mn, Pb) in coal, fly ash, and the residue at step 4 was determined by ICP-MS after digestion in a mixture of acids (HCl, HNO₃, HF, HClO₄).

Water/acid soluble and exchangeable fraction (F1): Composed of interchangeable HTEs, which was related with carbonates and those soluble in water or under slight acidic condition.

Reducible fraction (F2): Composed of HTEs bound to Fe and Mn oxyhydroxides that might be released if the environmental conditions changed from oxic to anoxic.

Oxidizable fraction (F3): Composed of HTEs associated with organic matter and sulfides, which could be leached out under oxidizing condition.

Residual fraction (F4): Composed of HTEs strongly bound to the structures of crystalline and amorphous phases, which could be only extracted by aqua regia digestion. Thus, HTEs in this fraction were unlikely to release, unless they have undergone extreme weathering condition.

HTEs in water/acid soluble and exchangeable fraction were considered to be easily leachable to the environment. HTEs in reducible and oxidizable fraction were thought to be potentially leachable to the environment. The fraction including the water/acid soluble and exchangeable, reducible and oxidizable fraction was defined as the mobile fractions, which represented the HTEs associated with the mobile phase. HTEs in the mobile fraction were considered to be easily leached out into the environment, which affected bioavailability, biological activity and interactions among organisms and their environment [37].

To minimize the experimental error, all the measurements were conducted repeatedly, and all the data given in this paper are the average value.

2.3. Leaching characteristics of HTEs in fly ash

Many factors, such as liquid/solid ratio, pH value, leaching time, etc. can influence the amount of HTEs leached out from fly ash [30,31,38]. As a baseline, the leaching characteristics of HTEs in the four fly ash samples were conducted by mixing 4 g of the sample with

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