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Study of detailed geochemistry of hazardous elements in weathered coal ashes



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ABSTRACT

The geochemical changes and the behaviors of the hazardous elements during the weathering of coal ash are not well known despite the potential toxicity of the hazardous elements leached from the ashes. In this work, ashes disposed in ash ponds were collected from different depths to investigate the effects of weathering on the geochemical changes and the mobilities of hazardous elements. The mineral phases identified in ash samples are quartz, mullite, K-feldspar, magnetite, illite, and glass phase. The contents of most major ions such as Si, Al, and K are strongly correlated with the changes in the mineral compositions of the ashes. However, the decrease in Ca and increase in loss on ignition (LOI) values with depth at both sites are observed owing possibly to the weathering of ashes. Our results show that leaching of hazardous elements caused by saturation with sea water is very important for the mobility of hazardous elements. Sequential extraction experiments show that high contents of hazardous elements are in the residual fraction, particularly in the raw coal ashes. For weathered ashes, a higher fraction of hazardous elements, especially As is bound to iron and manganese oxides, which increases significantly with depth. This indicates that As can be easily leached owing to the concentrated As on the surfaces of ash grains. Moreover, it can be strongly sorbed on iron oxide and hydroxide or co-precipitated with them. Therefore, magnetite and weathering products such as iron hydroxide can reduce the toxicity of As and other hazardous elements leached out during weathering.

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1. Introduction

A large portion of power generation is from coal firing, wherein the combustion of coal produces a range of residues including coal fly ash (CFA) and bottom ash [1-3]. Many different methods have been developed to utilize or dispose of the large amounts of ash resulting from power generation. Coal ash can be utilized in products such as building materials, zeolite synthesis, wood substitute products, soil stabilization, road base/embankments and ground consolidation, land reclamation, and soil amendments in agricultural applications [4-8]. However, large amounts of ash have been dumped in landfills, CFA basins, and artificial dams, lagoons, and ponds. Coal ash containing high levels of leachable toxic elements can contaminate soil, groundwater, and surface water, which poses a threat to the environment [8-13]. Therefore, many studies were carried out to investigate the leaching characteristics of the hazardous elements from coal ash [11,14-16]. When CFA is dumped in ash ponds, the leaching of hazardous elements can increase owing to the reaction of the ash surface. Because the weathering of CFA also affects the leachability of metals contained in coal ash, several studies have been performed to determine the effects of weathering on the mineralogical changes and leaching of hazardous elements from this ash. Zevenbergen et al. [17] used various mineralogical methods and found that after weathering, clay formed rapidly, and hazardous elements were fixed on noncrystalline clays. Akinyemi et al. [18,19] compared fresh and weathered CFA. They reported that mineralogical and geochemical changes occurred after weathering, and they discussed the possible environmental effects. However, the long-term environmental effects of coal ashes and contaminated soils dumped in landfills are not fully understood [3,20].

The hazardous elements in coal ash are partitioned in different chemical forms, and their toxicity is dependent mainly on the chemical forms rather than the total concentrations [21–23]. Sequential extraction techniques have been developed and applied to coal ashes to provide useful information on the mobility of hazardous elements [24–28]. The objectives of the present study are to







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compare the behaviors of hazardous elements in weathered coal ashes by using X-ray diffraction (XRD), scanning electron microscopy (SEM), chemical analysis, and sequential extraction methods. In addition, the different factors affecting the leaching behaviors of the hazardous elements are discussed.

2. Experimental

2.1. Sample collection

The samples used in this study were obtained from the Seocheon Electric Power Plant, Korea, which annually burns about 0.8 Mt of pulverized anthracite coal (Fig. 1). These ashes are composed of 80% CFA and 20% bottom ash and were collected by electrostatic precipitator and water-filled hopper methods, respectively [26]. The ashes were disposed in ponds formed within an offshore concrete dyke. A currently closed disposal site at which both CFA and bottom ash were disposed was in operation for nine years. When the samples were collected from a new disposal site, the CFA had been recycled, and only bottom ash had been disposed for seven years. At the closed old ash pond, the disposed ashes were covered by soil layers. Five samples at respective depths of 125 cm, 350 cm, 600 cm, 850 cm, and 1100 cm, were collected by using a vibracore device. Soil cover was not present at the new ash pond, from which four samples at respective depths of 50 cm. 250 cm. 500 cm. and 700 cm were collected by using the same device. Currently, bituminous coal is used for the power plant. A fresh CFA sample was also obtained to compare the mineralogical and chemical data.

2.2. Sample preparation and analysis

All of the samples were dried for three days in oven at 30 °C prior to analysis. The mineral compositions of the ash samples were examined by X-ray diffraction (XRD; X'pert APD, Philips,) with Ni-filtered Cu-K α radiation at 40 kV and 30 mA at the Daegu Center of Korea Basic Science Institute. The data were collected from 5° to 50° 2 θ in 0.016° 2 θ steps with a counting time of 1 s



Fig. 1. Location of study site where sampling was conducted.

per step. For quantitative analysis of the mineral compositions, the ash samples were mixed with 20% corundum powder and were ground to less than 5 μ m in preparation for XRD analysis. The diffractometer was a Rigaku Ultima IV equipped with a Cu target operating at 40 kV/30 mA, and the counting time was 2 s per 0.03° steps. The phase compositions of the CFA samples in weight% were determined by using Siroquant software in the region of 14–65° 20. Similar procedures for coal and other materials were previously reported by other works [29–34].

The CFA grains were mixed with Araldite epoxy resin and hardener. The mixture was smeared on a metal stub for scanning electron microscope (SEM) observation. The stub was gently heated to homogenize the mixtures and to reduce the appearance of bubbles. After curing for two days, the mixture was ground on rotating diamond plates followed by polishing with 0.5 μ m diamond paste. The polished surface was coated with carbon and was observed by SEM (VEGA II LMU, Tescan) in the back-scattered electron image mode. The chemical composition was analyzed by an energy dispersive X-ray spectrometer (EDS).

The concentrations of Al_2O_3 , CaO, Fe_2O_3 , K_2O , MgO, MnO, Na₂O, P₂O₅, SiO₂, and TiO₂ in the coal ashes were determined by X-ray fluorescence (XRF; PW 1404, Philips) using the fused glass method.

To analyze the hazardous elements contained in the coal ashes, total digestion methods suggested by the United States Environmental Protection Agency (EPA; modified method 3052) were used. The concentrations of the hazardous elements were examined by inductively coupled plasma (ICP; Ultiam 2C, Jobin Yvon) analysis at the Seoul branch of Korea Basic Science Institute.

To determine the chemical fraction of the hazardous elements, the sequential extraction method modified by Li et al. [35] from the scheme proposed by Tessier et al. [36] was used. This method includes five components for each fraction: exchangeable elements (F1), elements bound to carbonate and specially adsorbed (F2), elements bound to Fe/Mn oxides (F3), elements bound to organic matter and sulfides (F4), and elements in the residual fraction (F5). The concentrations of Pb, Cr, Co, Ni, Cu, Zn, As, and Cd of each chemical fraction were analyzed by using ICP–atomic emission spectroscopy (ICP–AES; Optima 4300 DV; PerkinElmer,) and ICP–Mass Spectroscopy (ICP–MS; X-series, VG Elemental Ltd.) at the Korea Basic Science Institute.

3. Results and discussion

3.1. Mineralogical compositions

The XRD patterns of the raw CFA and the ash samples from old and new pond sites are presented in Fig. 2. The mineral phases identified in those samples are mainly quartz and mullite with small amounts of K-feldspar, magnetite, and illite. In our samples, mullite is the main mineral formed during combustion. However, other minerals such as hematite, calcite, lime, enstatite, dolomite, anhydrite, portlandite, and gehlenite, which have been reported in other coal ashes were not identified in our samples likely owing to the different coal compositions, combustion processes, and temperatures of the coal [1–3,6,19,37–41].

The samples from the old site do not show noticeable changes with one exception: illite was found at lower depths based on the wt% of phase compositions calculated from the XRD patterns (Table 1). The mineral compositions of the samples from the new site are almost the same regardless of depth; however, small amounts of magnetite and illite were noted. The raw CFA sample shows the highest relative intensity of mullite peaks compared with other samples likely owing to differences in the coal. Differences between the XRD patterns and phase compositions of the samples from the two sites are obvious, and the wt% of mullite Download English Version:

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