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Abundances and distribution of minerals and elements in high-alumina coal fly ash from the Jungar Power Plant, Inner Mongolia, China

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ABSTRACT

The fly ash from the Jungar Power Plant, Inner Mongolia, China, is unique because it is highly enriched in alumina (Al_2O_3>50%). The fly ash mainly consists of amorphous glass and mullite and trace amounts of corundum, quartz, char, calcite, K-feldspar, clay minerals, and Fe-bearing minerals. The mullite content in fly ash is as high as 37.4% because of high boehmite and kaolinite contents in feed coal. Corundum is a characteristic mineral formed during the combustion of boehmite-rich coal.

Samples from the economizer were sieved into six size fractions (<120, 120–160, 160–300, 300–360, 360–500, and >500 mesh) and separated into magnetic, mullite + corundum + quartz (MCQ) and glass phases for mineralogical and chemical analysis. The corundum content increases but amorphous glass decreases with decreasing particle size. Fractions of small particle sizes are relatively high in mullite, probably because mullite was formed from fine clay mineral particles under high-temperature combustion condition. Similarly, fine corundum crystals formed in the boiler from boehmite in feed coal. The magnetic phase consists of hematite, magnetite, magnesioferrite, and $MgFeAlO_4$ crystals. The MCQ phase is composed of 89% mullite, 6.1% corundum, 4.5% quartz, and 0.5% K-feldspar.

Overall, the fly ash from the power plant is significantly enriched in Al_2O_3 with an average of 51.9%, but poor in SiO_2, Fe_2O_3, CaO, MgO, Na_2O, P_2O_5, and As. Arsenic, TiO_2, Th, Al_2O_3, Bi, La, Ga, Ni, and V are high in mullite, and the magnetic matter is enriched in Fe_2O_3, CaO, MnO, TiO_2, Cs, Co, As, Cd, Ba, Ni, Sb, MgO, Zn, and V. The remaining elements are high in the glass fraction. The concentration of K_2O, Na_2O, P_2O_5, Nb, Cr, Ta, U, W, Rb, and Ni do not clearly vary with particle size, while SiO_2 and Hg decrease and the remaining elements clearly increase with decreasing particle size.

1. Introduction

According to the report released by Coal Industrial Society of China in January 2009, China produced a total of about 2.716 billion tons of coal in 2008, 7.65% higher than last year. Chinese power plants consumed 1.09 billion tons of coal and generated 81.8% of the total electricity in 2005 (Zhou, 2006). The demand for coal as a major energy resource in China is projected to increase in the near future. Meanwhile, 293 million tons of coal combustion residues were generated by Chinese power plants in 2005 (Zhou, 2006) and the amount of that will increase every year.

Environmental problems can be caused by coal mining, transportation, storage and utilization, among which coal combustion is most significant (Meij and te Winkel, 2009). Toxic elements released during coal combustion have influenced normal growth of animals and vegetation, and human health in some countries and areas (Finkelman et al., 2002; Dai et al., 2005). On the other hand, certain elements are concentrated in fly ash that can be potentially of industrial value (Seredin, 1996; Seredin and Finkelman, 2008). Vassilev and Vassileva (2007) indicated that the abundance and modes of element occurrence play a leading role in specification and utilization of fly ash. Trace-element partitioning in pulverized-coal boilers is primarily dependent on the feed coal, boiler types, and pollution control system operating conditions (Guedes et al., 2008; Mardon et al., 2008; Huggins and Goodarzi, 2009).

Minerals are major carriers of elements in coal combustion residues. Minerals in coal combustion residues can be primary, secondary and tertiary in origin (Vassilev and Vassileva, 1996). Research on mineral behaviors during coal combustion can provide useful information for boilers to run safely and economically.

The purpose of this paper is to determine the mineralogical and chemical compositions of high-alumina fly ash generated at the Jungar Power Plant, Inner Mongolia, China. Both the coals from the Jungar

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coalfield and the fly ash from the Jungar Power Plant are unique because the raw coals are highly enriched in boehmite (Dai et al., 2006a, 2008) and the fly ash is high in alumina (Al_2O_3 > 50%). Because of the discovery of high Al and Ga contents in both coal and fly ash, the management of the Heidaigou Surface Mine, which is owned by Shenhua Group Zhungeer Energy Corporation Limited, has financially supported us to conduct a research project to investigate the modes of occurrence of Ga and Al in coals and to develop technology to extract the metals from associated combustion residues.

2. Samples and methods

The location of the Junger Coalfield was described by Dai et al. (2006a). Feed coals, fly and bottom ashes of the Jungar Power Plant were sampled in a period of 5 days (from June 20 to 24, 2006). Samples of pristine fly ash were collected from the economizer of the power plant with a special sampling device because the flue gas along with fly ash was disposed through wet ash concentration units at this power plant. Samples of wet fly ash (WA) were also collected at the ash concentration units and bottom ash was collected at the bottom hoppers.

Fly ashes from economizer were screened using sieves of 120 (120 μm), 160 (96 μm), 300 (48 μm), 360 (41 μm), and 500 mesh (25 μm). Wet fly ash was not used because of its pozzolanic nature, and even after being dried, particles could be conglutinated with each other. In addition, since it had reacted with water in the ash concentration units, some new minerals may be formed in wet fly ash, such as calcite formed from lime and gypsum formed from anhydrite.

All the feed coals were analyzed for moisture, ash yield, and sulfur content. Loss on ignition of fly ash was determined by heating the sample at 1050 °C for at least 1 h. Epoxy-bound pellets were made from fly ash samples and prepared to a polish with 0.05-μm alumina.

The minerals in fly ash samples were quantified by powder X-ray diffraction method, based on the method described in Petroleum and Gas Standard of China (SY/T 6210-1996: Quantification methods of total clay and other minerals in sedimentary rocks). The XRD pattern was recorded over a 2θ interval of 3°–70°, with a step size of 0.02°. A scanning electron microscope equipped with an energy-dispersive X-ray spectrometer was used to study the surface characteristics and determine the distribution of elements in fly ash. The accelerating voltage was 20 KV and the beam current was 10^{-10} A. X-ray fluorescence spectrometry (XRF) was used to determine the oxides of major elements, including SiO_2, Al_2O_3, CaO, K_2O, Na_2O, Fe_2O_3, MnO, MgO, TiO_2, and P_2O_5. Inductively coupled plasma mass spectrometry (ICP-MS) was used to determine trace elements in coal and ash samples that were first subjected to microwave digestion in ultrapure concentrated HNO_3 using high pressure quartz vessels. However, Hg, Se and As were determined by atomic fluorescence spectrometry (AFS) after dissolution by $HNO_3 + HClO_4 + HCl$. Fluorine was determined by pyrohydrolysis in conjunction with a fluoride ion-selective electrode, following the method described in Chinese National Standard GB/T 4633-1997.

In order to study chemical and mineral compositions in different phases of fly ash, the fly ash was separated into magnetic, mullite + corundum + quartz (MCQ), and glass phases. The magnetic materials were first separated by a hand magnet. The nonmagnetic fraction was then treated with 4% HF to dissolve the glass and the solid residues were mainly mullite, quartz, and corundum.

3. Results and discussion

3.1. Properties of feed coal at the power plant

The No. 6 coal (the uppermost Taiyuan Formation of Pennsylvanian age) of the Jungar Coalfield, Inner Mongolia, has a thickness between 2.7 and 50 m and an average thickness of 30 m (Dai et al., 2006a, 2008). The No. 6 coal at the Heidaigou Mine, Jungar Coalfield, is divided into 7 benches which are ZG6-1 to ZG6-7. Petrology and geochemistry of the coals have been reported (Dai et al., 2006a). The vitrinite random reflectance of the No. 6 coal is 0.58% (Dai et al., 2006a). Only mined coal from benches ZG6-5 to ZG6-7 is delivered to the power plant after having been washed. Coal from benches ZG6-1 to ZG6-2 is largely discarded because of a high ash yield. Coal mined from benches ZG6-3 to ZG6-4 is sold to other markets because of good qualities. The No. 6 coal is remarkably high in boehmite (33.7% of the total minerals). The origin of boehmite in coal is related to transportation of aluminum compounds from weathered and oxidized bauxite crust into the basin during peat deposition (Dai et al., 2006a).

The Jungar Power Plant generates about 200 MW of power and 380 Kt of fly and bottom ashes every year. The fly ash collected by ash concentration units accounts for 90% of total combustion residues which are piped into a landfill 3 km away to the northeast of the power plant.

Table 1 is a list of moisture, ash yield, and sulfur content in samples (C20–C25) of feed coal collected daily at the power plant in a period of 5 days and the average values of coal from the Heidaigou Mine. The feed coal of the power plant is low-sulfur and high volatile bituminous in rank (the average volatile matter is 33.5% on a dry and ash-free basis; Dai et al., 2006a) from the Heidaigou Surface Mine in the Jungar Coalfield. No significant variation of these characteristics is noticed between individual feed coals. The average ash yield of feed coal at the power plant is 33.0%, which is much higher than the average ash yield of coals from the Heidaigou Mine (17.2%) because the power plant uses the middling from the preparation plant from the lower benches (ZG6-5 to ZG6-7) of coals only from the mine.

Although the most boehmite-rich coals (ZG6-3 to ZG6-4) were excluded from the preparation plant, boehmite content in feed coal is still as high as 21.1% (mineral matter basis). Other minerals in feed coal are mainly 71.1% kaolinite, 3% gypsum, 2.5% calcite, and 1.9% quartz (mineral matter basis; Chen, 2005). The raw coal is enriched in aluminum and gallium (Dai et al., 2006a,b, 2008). Consequently, Al and Ga are highly concentrated in the fly ash (Dai et al., 2006b; Zhang et al., 2006).

3.2. Minerals and amorphous glass in fly and bottom ashes

3.2.1. Minerals in pristine bottom and fly ashes

As in most cases, both fly ash and bottom ash contain an important proportion of amorphous aluminosilicate glass, minerals, and char (Table 2). Fly ashes from the economizer and wet ash concentration units have much lower loss on ignition than the bottom ash. Minerals in bottom and fly ashes identified by XRD include mullite, corundum, quartz, and minor amounts of calcite and K-feldspar. The mullite and corundum contents in bottom ashes are much lower than those in fly ash. The dry fly ash from the economizer has a lower mullite and a higher glass content than the wet ashes from concentration units (Table 2). Fly ashes are composed of spherical, semispherical, and

Table 1
Moisture, ash yield, and sulfur content of feed coal from the Jungar Power Plant (wt.%).

Samples	M_{ad}	A_d	$S_{t,d}$
C20	3.62	36.9	0.33
C21	3.51	34.1	0.41
C22	3.35	33.2	0.42
C23	3.13	30.5	0.43
C24	2.91	30.2	0.39
Average feed coal	3.3	33.0	0.4
Heidaigou Mine[a]	5.19	17.7	0.73

M, moisture; ad, as-determined; A, ash; d, on a dry basis; S_t, total sulfur. C20–C24, feed coal collected daily for a period of 5 days.
[a] From Dai et al. (2006a).