

Available online at www.sciencedirect.com

ScienceDirect

www.elsevier.com/locate/jes

JES
 JOURNAL OF
 ENVIRONMENTAL
 SCIENCES
www.jesc.ac.cn

Distribution and mode of occurrence of uranium in bottom ash derived from high-germanium coals

Yinglong Sun¹, Guangxia Qi^{1,2}, Xuefei Lei¹, Hui Xu¹, Lei Li¹, Chao Yuan¹, Yi Wang^{1,*}

1. Department of Environmental Engineering, School of Environment, Tsinghua University, Beijing 100084, China.

E-mail: sunyinglong.007@163.com

2. Department of Environmental Science and Engineering, School of Food and Chemical Engineering, Beijing Technology and Business University, Beijing 100048, China

ARTICLE INFO

Article history:

Received 25 May 2015

Revised 16 July 2015

Accepted 17 July 2015

Available online 26 September 2015

Keywords:

Coal bottom ash

Uranium

Radioactivity

Tessier sequential extraction

Magnetic separation

ABSTRACT

The radioactivity of uranium in radioactive coal bottom ash (CBA) may be a potential danger to the ambient environment and human health. Concerning the limited research on the distribution and mode of occurrence of uranium in CBA, we herein report our investigations into this topic using a number of techniques including a five-step Tessier sequential extraction, hydrogen fluoride (HF) leaching, Siroquant (Rietveld) quantification, magnetic separation, and electron probe microanalysis (EPMA). The Tessier sequential extraction showed that the uranium in the residual and Fe–Mn oxide fractions was dominant (59.1% and 34.9%, respectively). The former was mainly incorporated into aluminosilicates, retained with glass and cristobalite, whereas the latter was especially enriched in the magnetic fraction, of which about 50% was present with magnetite (Fe₃O₄) and the rest in other iron oxides. In addition, the uranium in the magnetic fraction was 2.6 times that in the non-magnetic fraction. The experimental findings in this work may be important for establishing an effective strategy to reduce radioactivity from CBA for the protection of our local environment.

© 2015 The Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences.

Published by Elsevier B.V.

Introduction

Lignite is often used as fuel for many small to middle scale pithead power plants, and in some cases, as a raw resource for Ge smelters (Papastefanou, 2010; Dai et al., 2014c). However, in Lincang, Yunnan province, China, the average radioactivity of uranium (U) in lignite can reach 87.1 Bq/kg, much higher than that of the other types of coal (such as low-rank coals, middle-rank coals and high-rank coals) (Xiong et al., 2007; Yu, 2007). After burning, the natural radioactivity level of coal combustion ash is 4–10 times higher than that of the feed coals (Bhangare et al., 2014; Tripathi et al., 2013), which may be extremely dangerous for the surrounding environment and

human health. For example, the enrichment and transformation of radionuclides in coal fly ash and bottom ash has already caused secondary pollution, and has negatively impacted the local environment and human health in Yunnan province, China (Yu, 2007).

Some late Permian coals are highly enriched in uranium (Dai et al., 2008, 2013a, 2013b, 2015a). Unfortunately, the radioactivity of their combustion residue (e.g., bottom ash) has not yet been studied in a great detail. In comparison with the abundant use of fly ash in construction materials (Dai et al., 2012; Camilleri et al., 2006; Eze et al., 2013; Lima et al., 2012), the coal bottom ash (CBA) is still stocked in piles close to coal fields, and could generate negative impacts on the surrounding environment including air,

* Corresponding author. E-mail: yi_wang@tsinghua.edu.cn (Yi Wang).

soil, ground water and human health (Bartořová and Klika, 2014; Lanzerstorfer, 2015; Liu et al., 2011). Therefore, it is essential to process the relatively highly radioactive CBA prior to its reutilization and to minimize its negative influence on the ambient environment.

As there are many uranium-rich coals in China and other countries, reducing radioactivity is becoming important. The main method to extract uranium from coal bottom ash is acid leaching. The experimental results of Paul and Seferinođlu indicated that nearly 80% of uranium in coal ashes was leached with sulfuric acid after 14 days (Seferinođlu et al., 2003; Paul et al., 2006), due to its predominant occurrence in uranium-organic compounds in the original coal. El-Hamid et al. (2014) reported that more than 97.1% leaching of the uranium in petroleum ash could be achieved, using a high sulfuric acid concentration (200 g/L) with 6% vol.% MnO₂ oxidant and 6 hr of agitation. However, direct acid leaching of uranium from many other coal bottom ashes is difficult. Lei et al. (2014) were only able to leach less than 20% of the uranium from their samples. Zielinski et al. (2007) compared the leaching conditions of uranium and arsenic in coal ash and found that leaching of arsenic with a carbonate buffer solution was rapid and efficient (the leaching rate was 49%). In contrast, U barely leached (7%) in 2 weeks. Most explanations for the low leaching efficiency of uranium in CBA involve the relative insolubility of uranium residing in particles within a glassy matrix (Zielinski et al., 2007; Zielinski and Budahn, 1998).

Thus, extraction of uranium from coal ashes greatly differs with coals and regions, but a uniform standard extraction method has not been developed for uranium-rich bottom ash (Zhang et al., 2008). Different uranium extraction methods, which depend on the combustion conditions (e.g., combustion temperatures, categories of raw coal, furnace types) and modes of occurrence of uranium in raw coals, would lead to different leaching efficiencies. Therefore, to effectively extract uranium and reduce radioactivity from CBA, the distribution and mode of occurrence of uranium in bottom ash must be known.

The purpose of this work is to investigate the distribution and mode of occurrence of high uranium bottom ash. The samples were supplied from Lincang, southwestern China, and a number of extraction and analytical techniques were utilized including a five-step Tessier sequential extraction, hydrogen fluoride (HF) leaching, Siroquant (Rietveld) quantification, magnetic separation, and electron probe microanalysis (EPMA). The experimental findings in this work are not only important for understanding the distribution of uranium in bottom ash, but also for establishing an effective strategy to reduce radioactivity in CBA and protect the local environment.

1. Materials and methods

1.1. Samples and reagents

The CBA samples were obtained from two different germanium (Ge) smelters in Lincang, Yunnan Province, China (samples no. 1 and no. 2). The samples were crushed with a ball mill, and then passed through a 500-mesh standard sieve (<25 μm in diameter). The fine powder samples were dried at 105°C in a forced air oven to constant weight and stored in a

desiccator until further use. The CBA was characterized as a uranium-rich (374 ppm) material with low-level radioactivity (gross alpha decay (α) of 3.08 Bq/g, and gross beta decay (β) of 11.83 Bq/g). The two samples had no significant differences in terms of components and characteristics, therefore, they were combined together for further investigation.

1.2. Tessier sequential extraction of uranium in the coal bottom ash

The combined samples were analyzed below. Tessier sequential extraction procedures were used to fractionate the uranium in the CBA into five components: exchangeable, bound to carbonates, bound to iron and manganese oxides, bound to organic matter, and remaining in residue (Tessier et al., 1979). The experimental procedures were analogous to those described in the literature (Wan et al., 2006; Smeda and Zyrnicki, 2002; Landsberger et al., 1995; Bódog et al., 1996). Briefly, (1) the CBA was extracted at room temperature for 3 hr with a sodium acetate solution (1 mol/L CH₃COONa, pH 8.2) under continuous agitation; (2) the residue from (1) was leached at 50°C with a 1 mol/L sodium acetate solution adjusted to pH 5.0 with acetic acid (CH₃COOH). Continuous agitation was maintained for 5 hr; (3) the residue from (2) was extracted with a 0.04 mol/L NH₂OH–HCl solution in 25% (V/V) acetic acid. The extraction occurred at 60°C under continuous agitation for 8 hr; (4) a solution of 0.02 mol/L HNO₃ and 30% H₂O₂ adjusted to pH 2 with HNO₃ was added to the residue from step (3), and the mixture was heated at 85°C for 2 hr under continuous agitation. NH₄Ac was then added and the sample was heated again to 65°C for 6 hr under continuous agitation; and (5) the residue from (4) was digested with a mixture of HF, HNO₃ and HClO₄ for total metal analysis. Each step was repeated four times, and the leachate was collected separately to measure the concentration of uranium and other major metals.

1.3. HF leaching of the coal bottom ash residue

A total of 5 g of the residual Tessier fraction was placed into a 150 mL Teflon beaker, and 80 mL of an HF solution (10%, 8%, 6%, 4%, or 2%, V/V) was added. The resulting suspension was magnetically stirred at 500 r/min for 20 min at room temperature, and then the slurry was centrifuged. The supernatant was collected and the residue washed with 80 mL of distilled water. The supernatant and washing solution were mixed and diluted up to 500 mL, and then a 10 mL-aliquot was withdrawn for a uranium content analysis. The solid residue was further washed twice, dried, and the weight of the residue was recorded. The leaching experiment was performed in duplicate.

1.4. Magnetic separation of the coal bottom ash

A total of 20 g of CBA was added to 2.0 L of distilled water, and the slurry was stirred vigorously with a magnetic rod (3000 G). This procedure was repeated until no more magnetic fraction adhered to the magnet. Then the residual parts were further subjected to a wet-type high intensity magnetic separator with a magnetic field intensity of 15,000 to 20,000 G to collect the weakly magnetic fractions. The weakly magnetic, strongly

Download English Version:

<https://daneshyari.com/en/article/4453660>

Download Persian Version:

<https://daneshyari.com/article/4453660>

[Daneshyari.com](https://daneshyari.com)