Available online at www.sciencedirect.com

ScienceDirect

Procedia Environmental Sciences 31 (2016) 589 – 597

Procedia Environmental Sciences

The Tenth International Conference on Waste Management and Technology (ICWMT)

Extraction of uranium in bottom ash derived from high-germanium coals

Yinglong Sun[a], Guangxia Qi[a], Xuefei Lei[a], Hui Xu[a,b], Yi Wang[a,1]

[a]*Department of Environmental Engineering, School of Environment, Tsinghua University, Beijing 100084, China*
[b]*Northwest Institute of Nuclear Technology，Xi'an 710024，Shanxi，China*

Abstract

The concentration of uranium (374mg/kg) and radioactivity in coal bottom ash (CBA) from high-germanium containing coals were much higher than that of other bottom ashes generated from normal coals. The radioactivity from daughter radionuclides generated by uranium decays. Apparently, the much higher radioactivity in this kind of solid waste may present a potential danger to the ambient environment and human health. However, the recovery of uranium as resources and removal of radioactivity were rarely studied. The distribution and occurrence of uranium in CBA were studied. The experimental results show that uranium mainly exists as incorporated into aluminosilicates (mainly glass phase) (59.1%) and Fe-oxides (34.9%). The extraction of uranium was achieved by Tessier sequential extraction, acid leaching, magnetic separation, mechanical activation combined with alkali activation, as well as calcination with $Cacl_2$ followed by HNO_3 leaching. The results demonstrated that the optimal method of extraction uranium was calcination with $Cacl_2$ followed by leaching with HNO_3. By this method, the recovery of uranium can be as high as up to 95.8%, and removal of gross α and gross β can reach92.9% and 84.9%, respectively. The possible mechanism of uranium recovery by $Cacl_2$ roasting followed by HNO_3 leaching was also investigated in this work. After processing, the radioactive CBA can meet the requirement of the standard of limit on radioactive substance for industrial waste slags used in building materials. It can be concluded that the method of $Cacl_2$ roasting followed by HNO_3 leaching can be used for the resource-oriented utilization of radioactive CBA.

Peer-review under responsibility of Tsinghua University/ Basel Convention Regional Centre for Asia and the Pacific

*Keywords:*Coal bottom ash; Uranium; Extraction; Removal.

* Corresponding author. Tel.: 86-10-6277-3313; fax: 86-10-6277-3313.
*E-mail address:*yi_wang@tsinghua.edu.cn.

Peer-review under responsibility of Tsinghua University/ Basel Convention Regional Centre for Asia and the Pacific
doi:10.1016/j.proenv.2016.02.096

1 Introduction

Some lignites in the world, e.g. South Texas (USA) [1], Ajka valley (Hungary) [2], Figueira County (Brazil) [3], southern Urals and Transbaikalia (Russia) [4], Northern Greece are highly enriched in U [5]. Middle Asia is probably the most abundant area which contains the U-bearing coals [4]. In China, some late Permian coals in south and north China, e.g. Guiding (Guizhou Province) [6], Yanshan (Yunnan Province) [7], Heshan (Guangxi Province) [8],Fusui (Guangxi Province) [9], Yili (Xinjiang Province) are also highly enriched in U [10].However, the radioactivity of their combustion residue (e.g. bottom ash) has not been studied yet. Its impact on the surrounding environment (including air, soil and underwater) and human health may be significant [11]. Hence, it is essential to treat such coal bottom ash with comparatively higher level of radioactivity before reutilization and minimize its influence on ambient environment.

As there are many uranium-rich coals in China and other countries, the removal of uranium and radioactivity is becoming significant. The main method to extract uranium from coal bottom ash is acid leaching.The experimental results of Pauland Seferinoğluindicated that, nearly 80% of uranium in coal ashes was released to the solution after leaching with sulfuric acid for 14 days [12,13], because the metal predominantly existed as uranium organic compounds in the original coal. Hamid reported that leaching of uranium from petroleum ash could achieve more than 97.1% applying the optimum leaching condition [14]. However, direct leaching of uranium from many other coal bottom ashes by acid solution was verified difficult, only less than 40% of uranium was leached [15]. Zielinski compared the leaching conditions of uranium and arsenic in coal ash [16], and found that leaching of arsenic with the carbonate buffer solution was rapid and efficient (leaching rate was 49%), in contrast, U was hardly leached (7%) in two weeks. Most explanations for the low leaching efficiency of uranium in CBA were the relatively insolubility of uranium residence in particles within glassy matrix [17], but lack of direct evidences.

In conclusion, extraction of uranium from coal ashes greatly differed from coals and regions, but there hasn't been a unified extraction method applied for global uranium-rich bottom ash. Different uranium extraction methods, which depended on the different 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 remove radioactivity from CBA, the distribution and mode of occurrence of uranium in bottom ash should be performed.

Yunnan province in southwestern China is an important lignite producing area, the reserves of lignite in Yunnan accounts for 11.9% of total amounts in China [18]. The average radioactivity of uranium in lignite is higher than that of the other kinds of coal (e.g., 87.1Bq/kg in lignite produced from Yunnan Province, China) [19], sometimes even up to 624.4 Bq/kg [20]. After its burning, the natural radioactivity level of coal combustion ash is 4-10 times higher than that of feed coals [21,22]. Hence, it is essential to treat such coal bottom ash with comparatively higher level of radioactivity prior to reutilization and to minimize its influence on ambient environment.

As discussed above, the purpose of this work was to investigate the extraction and recovery techniques based on distribution and the mode of occurrence of uranium in bottom ash with comparatively higher radioactivity in Lincang, southwestern China.

2. Materials and Methods

2.1. Samples and reagents

The CBA samples were obtained from two different germanium (Ge) smelting factories in Lincang, Yunnan Province, China (sample No.1 and No.2). The samples were crushed into powder by milling with a ball mill, and then were 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 for further use. The CBA was characterized as a uranium-rich (374 ppm) waste with low-level radioactivity (gross α of 3.08 Bq/g, and gross β of 11.83 Bq/g).

2.2. Tessier sequential extraction of uranium in CBA

Tessier sequential extraction procedures were used to take the uranium fractionation in CBA into five fractions: the exchangeable, bound to carbonates, bound to iron and manganese oxides, bound to organic matter, and