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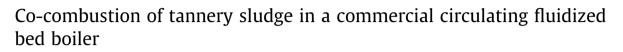
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Dong Hao, Jiang Xuguang*, Guojun Lv, Chi Yong, Yan Jianhua

State Key Laboratory of Clean Energy Utilization, Zhejiang University, Hangzhou 310027, China

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ABSTRACT

Co-combusting hazardous wastes in existing fluidized bed combustors is an alternative to hazardous waste treatment facilities, in shortage in China. Tannery sludge is a kind of hazardous waste, considered fit for co-combusting with coal in fluidized bedboilers. In this work, co-combustion tests of tannery sludge and bituminous coal were conducted in a power plant in Jiaxing, Zhejiang province. Before that, the combustion behavior of tannery sludge and bituminous were studied by thermogravimetric analysis. Tannery sludge presented higher reactivity than bituminous coal. During the co-combustion tests, the emissions of harmful gases were monitored. The results showed that the pollutant emissions met the Chinese standard except for NOx. The Concentrations of seven trace elements (As, Cr, Cd, Ni, Cu, Pb, Mn) in three exit ash flows (bottom ash in bed, fly ash in filter, and submicrometer aerosol in flue gas) were analyzed. The results of mono-combustion of bituminous coal were compared with those of co-combustion with tannery sludge. It was found that chromium enriched in fly ash. At last, the leachability of fly ash and bottom ash was analyzed. The results showed that most species were almost equal to or below the limits except for As in bottom ashes and Cr in the fly ash of co-combustion test. The concentrations of Cr in leachates of co-combustion ashes are markedly higher than that of coal mono-combustion ashes.

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1. Introduction

Hazardous wastes need our concern as they are more harmful to environment and human health than other wastes. In 2013, 31.57 million tons industrial hazardous wastes was generated in China, of which 25.7% were stored temporarily, waiting for disposal according to the 2012 Environmental Statistics Annual Report by the Chinese Environmental Protection Department. The risk and challenge is growing with rising hazardous waste generation and the lack of disposal capacity in China (Duan et al., 2008).

Co-combustion in fluidized bed combustors (FBC) has been proved effective to dispose of many kinds of wastes. This technique has two clear advantages. On the one hand, widely existing experience in combustion technology can be applied to wastes as potential fuels; on the other hand, existing FBC facilities are promising to burn a great variety of wastes. However, influences of co-combustion on operation and emissions may be a potential drawback and should be investigated.

Previous studies Huotari and Vesterinen (1996), Philippek and Werther (1997), Laursen and Grace (2002), Tsai et al. (2002),

* Corresponding author. E-mail address: jiangxg@zju.edu.cn (X. Jiang).

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Wagland et al. (2011) and Leckner et al. (2004) have investigated co-combustion in FBC with a variety of materials like biomass, MSW, RDF and sewage sludge. Several co-incineration experiments of hazardous wastes in FBC have also been reported and hazardous wastes co-firing included automotive shredder residue and bioferment residue (Van Caneghem et al., 2010; Jiang et al., 2012). Gaseous emissions of major pollutants (CO, NO_X, and SO₂) during co-firing were studied. Varol et al. (2014) tested co-firing of Turkish lignites and wood chips in a circulating bed combustor and investigated changes in emissions of CO, NO, and SO₂ as a function of excess air ratio. Leckner et al. (2004) studied co-combustion of sewage sludge and coal/wood in two circulating fluidized bed (CFB) units, a laboratory scale plant and a pilot scale CFB boiler respectively. Emissions of gaseous pollutants were tested and they met the emission standards of the EU. The Influence of air staging on emissions was also studied. Lee et al. (2010) studied the effect of co-combustion ratio on the emissions of NO_x and SO₂ during co-combustion of refuse derived fuel and Korean anthracite in a commercial CFB boiler and obvious changes of emissions were not observed. The Content of nitrogen in waste materials had a significant influence on N₂O and NO_X emissions. A reduction of N₂O emissions was observed by researchers during co-combustion of coal and mature biomass or RDF with low nitrogen content

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(Desroches-Ducarne et al., 1998; Liu et al., 2002; Boavida et al., 2003); in contrast co-combustion of wet sludge with higher content of nitrogen with bituminous coal in an circulating FBC led to an increase in N₂O (Philippek and Werther, 1997; Werther and Ogada, 1999). Ash formation and characteristics during co-firing in FBC was another research focus. Elled et al. (2010) surveyed the fate of potassium, which led to deposits on heat transfer surfaces during co-firing of municipal sewage sludge with biomass containing high amounts of potassium and chlorine. The addition of municipal sewage sludge was found to be beneficial to control alkali chlorides and corrosive deposits. Davidsson et al. (2007) performed co-firing experiments in a 12 MW CFB boiler at Chalmers University of Technology, Sweden with biofuel, coal and sewage sludge and examined effects of feed composition on bed ash and fly ash. The results also showed that sewage sludge eliminated alkali chlorides in flue gas and deposits. Pettersson et al. (2008) carried out co-combustion tests on the same unit as Davidsson et al. They applied the Chemical fraction method to predict and characterize fly ash properties. Heavy metals in ashes are significant with respect to assessing environmental risks. Elled et al. (2007) investigated the fate of trace elements during cocombustion of wood and municipal sewage sludge in a fluidized bed boiler. The experimental data agreed fairly well with the prediction of volatility by thermodynamic equilibrium calculations for most trace elements. Amand and Leckner (2004) studied metal emissions from co-combustion for sewage sludge and coal/wood in fluidized bed. Emission of trace metals from co-combustion and enrichment in ashes were investigated to see the effect of the sludge share in the fuel mixture on the fate of trace metals. Despite all previous studies mentioned above, there is little literature on the co-combustion of tannery sludge and coal in fluidized bed combustors.

Tannery sludge (TS) is a kind of hazardous waste generated from the tanning process (Chuan and Liu, 1996) and classified under the code of HW21-193-001-21 in China Hazardous Waste List, as enacted in 2008. It is considered to be a potentially hazardous waste that can be co-combusted with coal in FBC. Hence, it is necessary to investigate the effects of co-combustion of coal and TS, and the operation of co-combustion of coal and TS in FBC and their pollutants emitted should be considered.

Thermogravimetric analysis (TGA) is an effective and fundamental method to investigate the thermochemical behavior (combustion or pyrolysis) of materials. In this work, TGA was conducted for TS and coal in oxidation conditions to characterize their combustion behavior. Furthermore, a co-combustion test of TS and coal was performed in an industrial circulating bed boiler (CFB) of a power plant in JiaXing city and mono-combustion of coal was also performed as a reference test. The effects of co-combustion on operation, emission and fate of heavy metals were studied.

2. Experimental

2.1. Materials

TS is from tannery factories in JiaXing and surrounding area. Bituminous coal in the power plant is used as auxiliary fuel. The proximate analysis, calorific value and ultimate analysis were carried out for both TS and auxiliary fuel (Table 1). The proximate analysis was determined by the Coal Industry Analysis Method (GB/T 212-2008). The lower heating value was quantified by the Chinese standard method of Determination of Calorific Value of Coal (GB/T 213-2008). The ultimate analysis was performed with an elemental analyzer 1ECO-CMNS932. The contents of F and Cl were obtained by the combustion–hydrolysis/ion chromatography (IC) method. Digested Samples of TS and coal were analyzed for heavy metals (Hg, As, Cd, Cr, Ni, Cu, Pb and Mn) by ICP-AES (inductively coupled plasma-atomic emission spectrometry).

Compared with bituminous coal, TS has a higher volatile matter content, with negligible fixed carbon so that the ignition of TS is earlier than that of bituminous coal, as follows from thermogravimetric analysis. A High content of heavy metals in TS is a risk. Of particular concern is the content of chromium which attains 0.32 wt%. Chromium is a risk to the environment and human health. According to previous research, chromium is the second most abundant inorganic ground-water contaminant at hazardous waste sites (Blowes, 2002) and hexavalent chromium is recognized by the International Agency for Research on Cancer and by the US Toxicology Program as a pulmonary carcinogen (Barceloux, 1999).

2.2. Thermogravimetric experiment

Thermogravimetric analysis is performed on a Mettler Toledo TGA/SDTA851° thermo analyzer, with a temperature range of 25–1100 °C, an accuracy of ± 0.25 °C, and a repeatability of ± 0.15 °C. About 10 mg sample was used in a single test. The TS samples were on an air-dry basis, while the coal sample was on an as-received basis. As volatile evolution ended below 1000 °C for most hazardous wastes (Jiang et al., 2010, 2009; Tao et al., 2010), the experimental temperature range was set from 25 °C to 1000 °C, at a heating rate of 30 °C/min. To study the effects of different heating rate, experiments at a heating rate of 10 and 50 °C/min were also

Table 1

Proximate and ultimate analysis, heat value and heavy metals content of TS and coal (on air dry basis).

Sample	Proximate analysis (%)						Lower heating value (J/g)	
	Moistur	e	Ash	Volatile		Fixed carbon		
TS Bituminous coal	0.93 4.40		20.02 18.77	78.90 33.17		0.15 43.66	13,483 22,595	
Sample	Ultimate analysis (%)							
	С	Н		Ν	S	0	F	Cl
TS	42.97	3.	32	0.03	0.83	31.90	0.01	0.7
Bituminous coal	58.48	3.	88	0.95	0.96	12.56	0.02	0.00
Sample	Heavy metal content (ppm)							
	Hg	As	Cd	Cr	Ni	Cu	Pb	Mn
TS	UD	931.5	18.13	3230	31.60	123.2	89.47	311.5
Bituminous coal	UD	148.1	3.794	159.2	82.55	38.56	69.52	11.3

UD - undetected.

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