



Chemical–mechanical bromination of biomass ash for mercury removal from flue gases

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ABSTRACT

Current approaches for mercury removal have been focusing on injection of powdered activated carbon sorbents into the flue gas stream. In particular, brominated activated carbon has been proven at pilot scale and plant trials to be effective at removing mercury from flue gases of coal-fired power plants. However, activated carbon is a costly source material. Using an industrial solid waste from biomass combustion as an alternative source material to produce sorbents for mercury emission control is an attractive option. This paper describes a novel chemical–mechanical bromination process for production of mercury sorbent from a biomass combustion ash, which is normally considered a solid waste. The chemical–mechanical brominated ash was characterized and tested for mercury capture and release at high temperatures. The brominated biomass ash was found to effectively capture mercury up to 390 °C in lab scale tests. The tests of the new brominated sorbent in a 375 MW coal fired power plant showed promising performance.

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1. Introduction

Mercury is a toxic substance which is emitted to the atmosphere through many sources, including coal-fired power plants. Airborne mercury becomes deposited into rivers, lakes and oceans, and bioaccumulates in the form of methyl-mercury, moving up the food chain as it is consumed by various aquatic species [1]. The primary source of mercury exposure in humans is through eating contaminated fish, which can cause neurological disorders and is especially detrimental to fetal brain development [1,2]. Some of the diseases which can be caused by mercury exposure are cerebral palsy, behavioral issues, memory loss and insomnia, along with problems with the immune, gastrointestinal and cardiovascular systems [1]. Bioaccumulation of methyl-mercury also impacts other animals such as fish, birds and mammals, causing impaired growth, behavioral abnormalities, lower reproductive success and even death [2]. The toxic effects of mercury have been largely realized in the last 20 years, leading to regulations at the state and federal levels of government. In Alberta, Canada, a regulation has been set in place to capture mercury at a minimum level of 70%, to be implemented in 2010 and proposals must be submitted by December 31, 2011 outlining the plan for continuous improvement (targeting a minimum of 80% capture) [3].

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Injecting a powdered activated carbon-based sorbent into flue gases of coal-fired power plants is one technology which has been studied for reducing mercury emissions [4–6]. For coals with sufficient chlorine content, a homogeneous reaction occurs between the gaseous chlorine and mercury, producing HgCl_2 . This understanding has led to work involving the addition of halogens, such as chlorine or bromine, to the activated carbon sorbent. Impregnating the halogen components on the sorbent has been found to increase its effectiveness of mercury removal (with higher removal efficiency and lower injection rates), especially for coal containing low levels of chlorine [4–6]. A review of various studies completed on brominated carbon sorbents by Yang et al. [6] described a faster adsorption of elemental mercury (Hg^0) on brominated sorbents than on non-brominated carbon sorbents. The same study showed that the sorbent capacity was slightly reduced when the temperature increased, while the capacity of the carbon sorbent improved as the amount of bromine on the sorbents increased. Tests at various power plants showed that the brominated activated carbon is very efficient at removing mercury [6].

In Alberta, at least one power plant has proposed to use the technology of injecting brominated activated carbon to meet the regulation requirements of 70% mercury removal, set in place by the government [3]. However, use of activated carbons as source materials is costly [7]. In biomass combustion based power plants, a large amount of ash with high carbon content up to 40–50% is produced as a solid waste. It would be attractive to use this waste

as the source material to produce sorbents for mercury emission control. The objective of this study is to develop an environmentally benign and economically practical sorbent for mercury emission control of flue gases from coal-fired power plants using biomass combustion wastes and liquid bromine as source materials. This sorbent is prepared using a novel chemical–mechanical bromination procedure developed in our research laboratory, and intended to be used as an injectable powdered sorbent. The brominated biomass ash is characterized and exposed to real flue gases in an online coal-fired power plant.

2. Experimental

The received biomass ash had a broad size distribution, and was ball-milled to prepare samples for bromination and mercury breakthrough tests. The biomass ash was placed in a tumbler charged with 1" steel balls. The tumbler was then capped and rotated for 3 h on a pair of mechanical rollers. The particle size distribution after tumbling was close to normal distribution with $d_{0.1} = 4.3 \mu\text{m}$, $d_{0.5} = 17.0 \mu\text{m}$, and $d_{0.9} = 53.4 \mu\text{m}$ [8].

2.1. Novel bromination procedure

Bromination was achieved through chemical–mechanical activation of biomass ash using a tumbler containing liquid bromine, biomass and glass beads, as shown schematically in Fig. 1. This chemical–mechanical bromination process featured two major benefits: good contact was established between the bromine and ash particles; and the ash particles were ground to an appropriate size convenient for transportation and injection.

A 10-L carboy containing 6-mm glass beads and liquid bromine was used for the chemical–mechanical bromination. Biomass ash was placed in the carboy at a glass beads to biomass ash ratio of 7 [8]. The carboy was tightly sealed and rotated on a set of rollers for 30 min. The resulting brominated ash was separated from the mixture using a 3.35 mm sieve before thermal treatment in a vacuum oven at 200 °C to ensure the stability of loaded bromine on biomass ash for safe storage, transportation and applications.

2.2. Mercury injection tests

Mercury pulse injection tests were performed in an argon carrier gas stream running at 40 mL/min, with the experimental setup described elsewhere [7]. A precisely weighed 40 mg of sample was placed in a borosilicate glass tube (4-mm i.d.) with quartz wool to

keep the powdered sorbent from escaping the tube. The tube was then placed inside a GC oven to control the mercury breakthrough tests at the desired temperatures from 20 to 400 °C. Once the oven reached the desired temperature for 5 min, the downstream GB trap was heated to remove any bromine or mercury liberated from the brominated biomass ash during heating. The GB trap consisted of gold-sputtered silica beads inside a quartz tube, and was used to preconcentrate trace amount mercury in the purge gas before its analysis. After cooling the GB trap, 200 μL of air saturated with Hg^0 at room temperature between 16 and 22 °C, corresponding to a Hg^0 concentration of 10.08–16.61 $\text{pg}/\mu\text{L}$ [9], was injected upstream of the sorbent. Mercury that was not captured by the sorbent leaked through the sorbent and was captured in the GB trap by amalgamation with the sputtered gold. Five minutes after the initial injection, the GB Trap was quickly heated, using a voltage applied to a heating wire, to above 400 °C to release the mercury from the GB trap to a Cold Vapor Atomic Fluorescence Spectrophotometry (CVAFS) detector (Tekran model-2500), where the amount of mercury passed through the sorbent was determined accurately. Mercury breakthrough for pulse injection tests has been previously defined [7] as the amount of mercury which is not captured by the sorbent, usually described as a percentage of the mercury injected. The goal is to have a low mercury breakthrough (or high capture) value.

Calibrations were completed at each temperature by injecting a known volume of Hg^0 saturated air, with a blank borosilicate glass tube containing quartz wool (no sorbent present). The test at each temperature was repeated several times to ensure repeatability of results. Three tubes (one blank, two samples) can be inserted into the GC oven for testing at each temperature. The materials used in constructing this experimental setup are all in Teflon, except the borosilicate glass tube.

2.3. Flue gas exposure tests

The brominated biomass ash was exposed to real flue gases at a 375 MW Alberta power plant using the method described by Liu et al. [7]. The selected power plant was burning Alberta subbituminous coal, and the exposure point was upstream of the electrostatic precipitator (ESP). The duct gas temperature was approximately 130 °C at the time of analysis, with a velocity of 17.9 m/s. Raw biomass ash, brominated biomass ash, commercial activated carbon (Norit FGL) and commercial brominated activated carbon (Norit Darco HgLH) were all tested for mercury capture in the flue gases. A portion of each sorbent was separated into a sampling container without being exposed to the field flue gases and used as the “field blank” sample. This “field blank” sample was tested for mercury content and compared with the sorbent exposed to mercury in the flue gases. The amount of mercury captured on the sorbent was determined by wet digestion of the samples followed by analysis using a PSA Millennium Merlin mercury analyzer based on the CVAFS principle.

2.4. Sorbent characterization

Scanning Electron Microscope (SEM) and X-ray Photoelectron Spectroscopy (XPS) at the Alberta Centre for Surface Engineering and Science (ACES) were used to characterize the sorbents. XPS analysis was conducted on the Br-Ash, Norit Darco HgLH and Raw Ash samples in order to compare bromine concentrations and carbon content in the sample. SEM analysis was also completed on the raw biomass ash before grinding, the brominated biomass ash and the commercial activated carbon sorbents. The sorbents were further characterized by measuring the BET surface area at the Integrated Nanosystems Research Facility (INRF) at the National Institute of Nanotechnology (NINT). Thermal Gravimetric

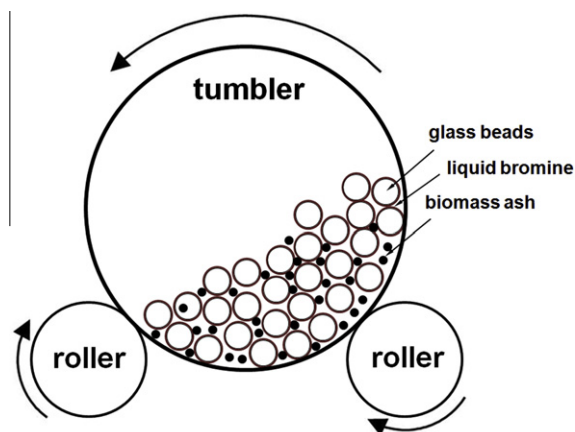


Fig. 1. Chemical–mechanical bromination of biomass waste ash in a laboratory tumbler [8].

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