

Study on removal of elemental mercury from simulated flue gas over activated coke treated by acid



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ABSTRACT

This work addressed the investigation of activated coke (AC) treated by acids. Effects of AC samples, modified by either different acids (H₂SO₄, HNO₃ and HClO₄) or HClO₄ of varied concentrations, on Hg⁰ removal were studied under simulated flue gas conditions. In addition, effects of reaction temperature and individual flue gas components including O₂, NO, SO₂ and H₂O were discussed. In the experiments, Brunauer–Emmett–Teller (BET), X-ray photoelectron spectroscopy (XPS) and Fourier transform infrared spectroscopy (FTIR) were applied to explore the surface properties of sorbents and possible mechanism of Hg⁰ oxidation. Results showed that AC sample treated by HClO₄ of 4.5 mol/L exhibited maximum promotion of efficiency on Hg⁰ removal at 160 °C. NO was proved to be positive in the removal of Hg⁰. And SO₂ displayed varied impact in capturing Hg⁰ due to the integrated reactions between SO₂ and modified AC. The addition of O₂ could improve the advancement further to some extent. Besides, the Hg⁰ removal capacity had a slight declination when H₂O was added in gas flow. Based on the analysis of XPS and FTIR, the selected sample absorbed Hg⁰ mostly in chemical way. The reaction mechanism, deduced from results of characterization and performance of AC samples, indicated that Hg⁰ could firstly be absorbed on sorbent and then react with oxygen-containing (C–O) or chlorine-containing groups (C–Cl) on the surface of sorbent. And the products were mainly in forms of mercuric chloride (HgCl₂) and mercuric oxide (HgO).

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1. Introduction

Mercury is known as an extremely hazardous substance because of its volatility, persistence and bioaccumulation. Methyl mercury, one organic transformation product of mercury, can be easily absorbed by human beings and have a hyper toxicity to nervous system [1,2]. The mercury in coal, in spite of its low concentration, is emitted directly and has become the major source of mercury in the atmosphere because of the large amount of coal burned. In order to prevent growing mercury pollution, “Minamata Convention on Mercury” was signed by 92 countries on 10 October 2013 to reduce the emissions of mercury and mercury compounds, while measures to control discharges from point source categories such as coal-fired power stations were put forward [3].

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During the combustion, mercury is released into exhaust gas as elemental mercury (Hg⁰) vapor, subsequently part of the vapor can be transformed into oxidized forms (Hg²⁺) and particle-bound atoms (Hg^p) [4]. Commonly used air pollution control devices are effective for Hg²⁺ and Hg^p. For example, Hg^p can be easily removed through particulate matter (PM) control devices such as electrostatic precipitators (ESP) and fabric filters (FF). Water-soluble Hg²⁺ can be captured in wet flue gas desulfurization systems (WFGDs) with high efficiency [5–7]. Nevertheless, Hg⁰ is difficult to be removed by using methods mentioned above because of its high volatility and nearly insolubility in water. Thus, exploitation of methods to capture Hg⁰ is the key access to control the mercury emission [8].

Numerous technologies have been employed to remove elemental mercury from exhaust gas. Therein, activated carbon injection is widely adopted by coal-fired plants to control mercury emission. The carbon is injected into exhaust gas stream and recycled by ESP or in a baghouse [9]. In addition, activated carbons treated by metal oxides or chloride, such as TiO₂, CuCl₂, CeO₂, MnO₂, CeCl₃, V₂O₅, etc. [10–14], are extensively used in the

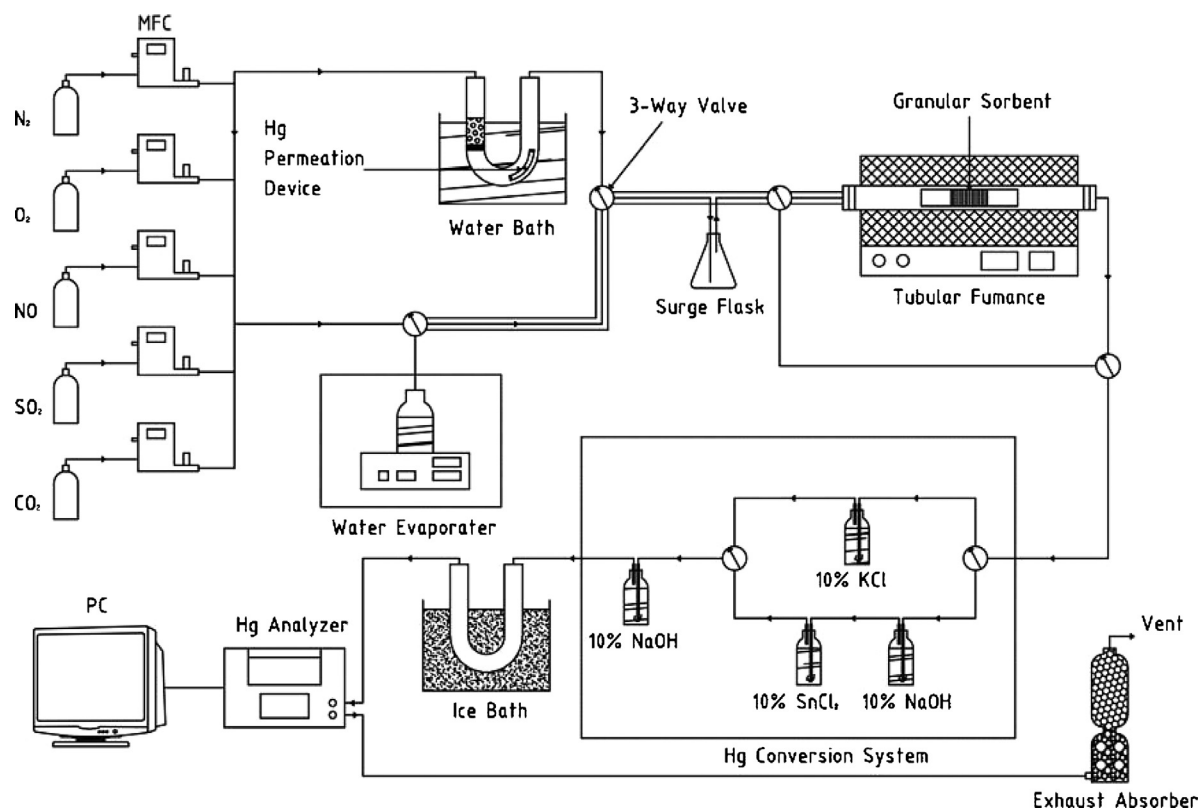


Fig. 1. Schematic diagram of the experimental setup.

process of Hg^0 settlement, in which Hg^0 is transformed into Hg^{2+} and then can be captured in WFGDs. However, these technologies cost tremendous capital, material and manpower, which hinders their wide applications. Besides, as an additive of concrete, the recycle activated carbon would cause secondary pollution due to Hg^0 release.

Activated coke (AC) is a kind of porous activated carbon-based adsorbent which is not adequately activated or retorting. Compared with activated carbon, AC possesses a higher mechanical strength and could be regenerated more conveniently. Furthermore, the price of AC is relatively inexpensive. Meanwhile, AC and activated carbon were similar in some aspects, such as the structure characteristics, adsorption and catalytic properties, physical and chemical stability [15]. In our previous studies [10,12,16], nitric acid (HNO_3) was used for pretreatment of catalysts. Oxidative acid could react with surface carbon-based substances and thus the structure of AC could be modified with the addition of oxygen-containing groups of acidic or alkaline which could improve the adsorption ability of sorbents. Additionally, massive researches indicated that chlorine played an important role in Hg^0 capture [12,17] since it could promote the conversion of Hg^0 to Hg^{2+} and Hg^{2+} was easy to be removed through solutions. Perchloric acid (HClO_4), the strongest inorganic acid, was widely used as oxidant in electropolishing and medical industry. Singh et al. [18] used HClO_4 to investigate the thermolysis of salts and demonstrated the oxidation through the reaction with ammonia. In this study, HNO_3 , HClO_4 and sulfuric acid (H_2SO_4) were engaged into the activation on AC. Under simulated flue gases (SFG) conditions, the performance of modified AC on Hg^0 capture was evaluated through experiment in a tubular electric furnace. Various characterization methods were applied to study the structure and physicochemical properties of AC. Mechanism involved in Hg^0 oxidation was investigated as well.

2. Experiment

2.1. Samples preparation

The target sorbent in our study was a kind of standard commercial AC (columnar granules with a length of 7–9 mm and a diameter of 5 mm) made by Inner Mongolia Kexing Carbon Industry Limited Liability Company. Acid activated AC samples were prepared in steps as follow: at first, HNO_3 , HClO_4 , H_2SO_4 of 4.5 mol/L were added into beakers filled with AC granules, respectively. And the liquid–solid ratio was 2.5 ml/g. Secondly, HClO_4 treated AC sample series were produced in the same way as above, and the concentrations of HClO_4 varied from 1.5 mol/L to 7.5 mol/L. Then, all solid–liquid mixtures were stirred well and stood in darkness for 12 h. After acidification, all samples were washed by deionized water to neutral and dried in an electric blast oven at 95 °C for 6 h. Finally, the AC samples were cooled down to room temperature and stored in a desiccator. The HNO_3 treated sample was noted as AC_N as well as the H_2SO_4 treated sample was noted as AC_S . Moreover, the HClO_4 treated samples were noted as AC_X , where X (valued of 1.5, 3, 4.5, 6, 7.5) represented the molar concentration (mol/L) of HClO_4 . Additionally, the blank sample was synthesized as a contrast following the methods above except for being immersed in deionized water, denoted as AC_F .

2.2. Experimental device and steps

The performance of acid activated AC was investigated on a bench-scale fixed bed system as shown in Fig. 1. All individual SFG components released from gas cylinders were controlled accurately by mass flow controllers. The rate of water vapor generated in evaporation plant was regulated by temperature controller. An elemental mercury permeation tube (VICI Metronics, USA) was

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