



Adsorptive removal of gas-phase mercury by oxygen non-thermal plasma modified activated carbon

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HIGHLIGHTS

- A non-chemical modified method of AC was investigated for Hg⁰ adsorption.
- Oxygen non-thermal plasma could promote mercury adsorption on activated carbon.
- The oxygen-containing groups of modified ACs surface were increased.
- External diffusion and chemisorption controlled Hg⁰ adsorption on modified ACs.

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ABSTRACT

The effects of oxygen non-thermal plasma modification on the surface properties and mercury removal performance of activate carbon (AC) were investigated. The raw and modified ACs were characterized by N₂ adsorption/desorption, scanning electron microscope (SEM), Boehm titration and Fourier Transform Infrared Spectrometer (FTIR). The results showed that the surface textual properties of the modified ACs were slightly damaged, but the surface chemistries such as the content of surface oxygen-containing groups were significantly changed. The elemental mercury removal performance of raw and modified ACs were also evaluated in a quartz tube fix-bed reactor under N₂ atmosphere. The results indicated that the AC treated with oxygen non-thermal plasma had better elemental mercury removal performance compared to the raw AC. This reason was attributed to the oxygen non-thermal plasma treatment increasing the ester groups (C–O), carbonyl groups (C=O) and adsorption activate sites on AC surface, which played an important role in the adsorption of elemental mercury. The adsorption kinetic of raw and modified ACs could be best described by the Pseudo-first-order model and Pseudo-second-order model, which implied that external diffusion and chemisorption were the control step in the mercury adsorption process of raw and modified ACs.

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1. Introduction

Mercury emission from anthropogenic resources, such as coal-fired power plants, municipal solid waste combustors and medical incinerators, has become a global environmental problem because of its hypertoxicity, chemical stability, bioaccumulation to human health and long-distance migration [1,2]. Compared to the other emission sources, coal-fired power plant is considered as the largest anthropogenic resource of mercury emission [3,4]. According the coal consumption in China in 2010, the mercury emission from coal combustion was estimated to be approximately 134 t [5].

Therefore, mercury has become the fourth pollutant following SO₂, NO_x and dusts in the process of coal combustion.

Mercury emitted from coal combustion flue gas usually contains three forms: elemental mercury (Hg⁰), oxidized mercury (Hg²⁺) and particle-bound mercury (Hg^p) [6]. It was reported that the majority Hg²⁺ and Hg^p could be removed effectively by typical air pollution control devices such as electrostatic precipitator (ESP), fabric filter (FF) and wet flue gas desulphurization (WFGD) [7–9]. However, the Hg⁰, which frequently constitutes the majority of mercury species in coal combustion flue gas, can hardly be removed because of its high chemical inertness and poor solubility [10]. Hence, it is very important to effectively control elemental mercury emission from combustion flue gas.

Powder activated carbon (PAC) injection has been considered as one of the most promising methods to remove elemental mercury

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from fuel gas. However, some crucial drawbacks, such as high carbon-to-mercury ratio, poor adsorption capacity and high cost, constraints wide application of this technology [11,12]. In order to promote mercury adsorption capacity and reduce the dosage of sorbent, numerous studies on chemical impregnation of AC have been carried out [13–17]. The impregnated element included sulfur, iodine, chlorine and bromine, which act as the active sites for mercury adsorption. However, the chemical impregnation is a complex procedure with a relatively high cost and the impregnated element may re-release into the flue gas to form new pollutants. Therefore, how to simplify the chemical treatment process, reduce the cost of modified AC and prevent the secondary pollution of impregnated element have been new challenges.

It is well known that mercury adsorption on AC is mainly determined by the surface textural properties and the surface chemistries [18,19]. Research from Li et al. [20] has indicated that the oxygen functional groups on AC surface could enhance the mercury removal efficiency. It has been reported that the non-thermal plasma could increase the amount of oxygen functional groups, which enhanced the adsorption of copper ion [21], iron ion [22] and nitrophenol [23] on AC. Ref. [24] reported that the technology of non-thermal plasma in air could increase the oxygen and nitrogen function groups on the activate carbon surface, and enhance the mercury removal efficiency. Compare to other chemical treatment methods, the non-thermal plasma treatment on AC is a promising method, because it does not need chemical solution and hardly damages the surface textural properties of AC. However, the systematic studies on the adsorption kinetics and mechanism of elemental mercury on the AC modified by oxygen non-thermal plasma are limited and further research is needed.

In this study, AC was modified by dielectric barrier discharge non-thermal plasma in oxygen. The effects of oxygen non-thermal plasma irradiation on the surface properties of AC were investigated. The surface properties of AC were characterized by N_2 adsorption/desorption, SEM, FTIR and Boehm titration. The adsorption of elemental mercury on raw and modified ACs were investigated systematically, and the adsorption kinetics and mechanism were analyzed by using Pseudo-first-order, Pseudo-second-order and Weber–Morris model.

2. Experimental section

2.1. Sample pretreatment

A commercial activated carbon (Shanghai Xinhui Activated Carbon Co., Ltd. China) was ground and sieved to different meshes. The AC with a particle size of 60–80 mesh was selected as the sorbent for experiments. Then, the selected AC was washed with deionized water for ten times to remove the ultrafine particles and dried at 105 °C for 24 h to eliminate the moisture.

2.2. Modification of AC by oxygen non-thermal plasma

The experimental apparatus of oxygen non-thermal plasma irradiated AC was shown in Fig. 1. The non-thermal plasma was generated by a dielectric barrier discharge (DBD) reactor, which had two quartz parallel-plates and two stainless plates. The size of quartz and stainless plates were, respectively, 100 × 40 mm (barrier thickness 2 mm) and 80 × 40 mm (barrier-to-barrier gap 8 mm). The quartz plates and the stainless plates were used for barrier materials and electrodes, respectively. 1.0 g of AC sample was evenly spread at the lower barrier surface of the reactor and the high purity oxygen was injected into the DBD reactor with the flow rate of 100 ml/min.

The oxygen non-thermal plasma was generated in the DBD reactor by using an alternating current with an audio-frequency of 25 kHz and a peak voltage of 30 kV. The power consumption of the non-thermal plasma reactor was estimated through voltage and current traces, which were stored in a digital oscilloscope (Tektronix, TDS3034B, USA) with the help of two high-voltage probes. The sample AC was treated by the oxygen non-thermal plasma for 10 min, 20 min and 40 min respectively, which were named as AC-O10, AC-O20 and AC-O40. The raw AC was named as AC-raw.

2.3. Fixed-bed mercury adsorption experiment

The mercury adsorption capacity evaluation of AC-raw, AC-O10, AC-O20 and AC-O40 were performed in a fixed-bed reactor at 298 K under N_2 atmosphere. The experiment apparatus of mercury adsorption test was shown in Fig. 2, which mainly includes Hg^0 vapor generator, temperature control system, fixed bed adsorption reactor and Hg^0 analyzer. A mercury permeation device was used as a source of Hg^0 , designed to produce constant Hg^0 vapor at the specified temperature. High purify nitrogen was supplied as carrier gas and a mass flow controller was used to control the flow of nitrogen at 200 mL/min. The initial mercury concentration was adjusted to 50 $\mu g/m^3$ by varying the temperature of mercury permeation device. The reactor was a quartz tube, and enclosed in a temperature-controlled oven, whose diameter was $\phi 18 \times 2$ mm. The elemental mercury concentration in simulated flue gas at outlet of the reactor was continuously measured by a continuous mercury vapor analyzer (VM3000, Mercury Instruments, Germany), which is based on cold vapor adsorption spectrometry.

In adsorption experiments, 200 mg activated carbon was packed into the quartz glass tube. The total flow rate of simulate flue gas was controlled at 2.0 L/min. All experiments were duplicated under the same experimental conditions. During each test, the initial Hg^0 concentrations (named as C_{in}) and the outlet Hg^0 concentrations (named as C_{out}) were measured. The Hg^0 removal efficiency of raw and modified ACs were calculated according to the Eq. (1):

$$Hg^0 \text{ removal efficiency}(\%) = \frac{C_{in} - C_{out}}{C_{in}} \times 100 \quad (1)$$

The Hg^0 accumulative adsorption amount per unit quality of AC was calculated according to the following equation:

$$q = \frac{Q_v}{m} \int_0^t (C_{in} - C_{out}) dt \quad (2)$$

where Q_v is flow rate of simulated flue gas, m^3/min ; m is AC mass, g; t is adsorption time, min; q is Hg^0 accumulative adsorption amount per unit quality of AC, $\mu g/g$.

3. Results and discussion

3.1. Discharge characterization of DBD reactor

The discharge characterization and power consumption of DBD reactor were valued by using Lissajous method, which was obtained on a digital oscilloscope screen by plotting with the Y-axis of transported charge and X-axis of applied voltage, because Lissajous diagram could reflect that the charged particles were distributed on barrier surface [25,26]. Fig. 3 shows the V–Q Lissajous diagram of DBD. It was clear from the figure that the Lissajous diagram was an approximate ellipse, which indicated that charged particles were uniformly distributed in the whole non-thermal reactor. The energy W per one discharge cycle T could be calculated as follows [27]:

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