

Regenerable Ag/graphene sorbent for elemental mercury capture at ambient temperature



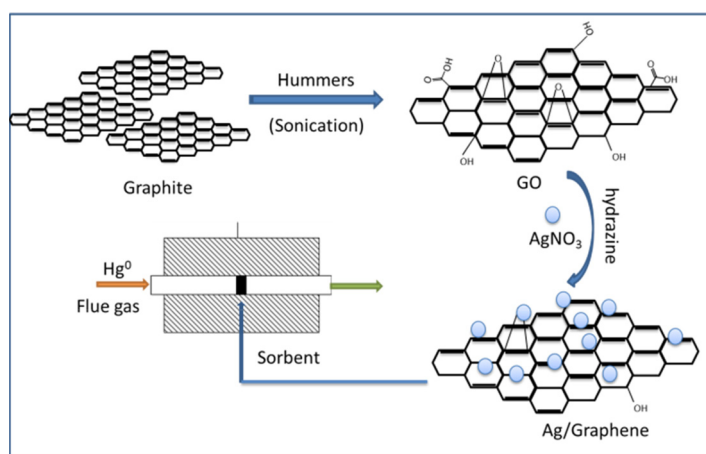
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HIGHLIGHTS

- Ag/graphene composite was synthesized by a facile method.
- The Hg^0 capacity of Ag/graphene was 4.2 mg/g at ambient temperature (25 °C).
- The acid gases had no effect on Hg^0 removal efficiency.
- Mercury could be collected by thermal separation after adsorption.

GRAPHICAL ABSTRACT



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ABSTRACT

Elemental mercury (Hg^0) is a widespread concern due to its high toxic and long residence time in the environment. Ag/graphene composite was synthesized as a high efficient and recyclable sorbent for Hg^0 removal. Ag particles were doped on the surface of graphene by a facile in situ chemical synthesis method. FTIR, XRD, and TEM results showed that Ag nanoparticles successfully grew on the surface of graphene layers. The performance of Ag/graphene as a sorbent was tested under a fix-bed reactor and the results showed that it could completely capture Hg^0 at ambient temperature (25 °C). The Hg^0 capacity was as high as 4.2 mg/g. The Hg^0 removal efficiency decreased from 94% to 60% when the temperature increased from 25 to 100 °C, respectively. SO_2 and NO had nearly no effect on Hg^0 removal, but the efficiency dropped by about 4% when O_2 existed. Hg-TPD results showed that mercury can be rapidly and efficiently released from the composite by thermal separation. The adsorption capacity of Hg^0 had no degradation after six cycles. The high Hg^0 capacity and recyclability make the Ag/graphene a candidate for the removal at ambient temperature.

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1. Introduction

Mercury has long been known to be toxic to humans [1]. After decades of hard work, the *Minamata Convention* was adopted in October of 2013, and it is a treaty aimed at controlling the mercury

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emission globally. The convention is expected to take effect in 2016, and it will call for a ban on the emission of mercury in 2018. The strict requirements about mercury control had been published by many countries. Therefore, it is significant to adjust and improve existing technologies or use the add-on technology to meet the new requirements.

In general, mercury is a trace element the environment, it existed stably in the fossil fuels such as coal and metal ores. But large amounts of mercury were transported from the geosphere into the atmosphere and hydrosphere through human activities in recent 200 years. It was reported that mercury emission from anthropogenic emissions greatly exceed natural geogenic sources, approximately 2100 tons of mercury is emitted by human activities annually [2]. Mercury emission from fuel combustion, waste incineration and mining are the main sources, resulting in high background level of mercury in the environment. And, more remarkable, elemental mercury (Hg^0) is one form of mercury, which is very hard to remove. It mainly exists in the atmosphere and the residence time of Hg^0 is several months to years [3]. Coal-fired power plants are regard as the largest emission sources. Hg^0 emission from coal-fired flue gas accounts approximately 35% of the total [3]. After coal-fired power plants, mercury emissions from cement and mineral production are the second largest anthropogenic sources [4,5]. Most of previous studies put more attention on the technologies for Hg^0 control among these major anthropogenic emissions. But mercury, as one kind of important resource, also exists in many artificial products (e.g., battery, thermometer, lamp, dental amalgam, and some electron devices). It was estimated that fluorescent lamp contain 0.72–115 mg of Hg per lamp, 500 mg for a typical fever thermometer [6,7]. In general, Hg^0 evaporated slowly from these broken products under ambient conditions. For these Hg-containing products, most of them were discarded with a low recycle rate. A large amount of mercury released from these products. The data showed that more than 14.1 tons of Hg^0 released into the environment from wasted batteries and fluorescent lamp annually [8].

Mercury is not only a pollutant but also an important resource. The future technologies for Hg^0 control should consider the high removal efficiency, the innocuity treatment and the recycling of resource. Adsorption method for the capture of Hg^0 was a potential technology. The mechanism for capturing Hg^0 over a sorbent can be divided into three sorts: amalgamation, physical adsorption and chemical adsorption [4,9–11]. Up to now, carbon, metals, metal oxides and selenium were often used as the Hg^0 sorbent at low temperature. Activated carbon (AC) was an efficient sorbent support due to its large surface areas and porosity. Sulfur modified AC showed the Hg^0 capacity of 2.6 mg/g at 20 °C along with the surface areas of 1000–1100 m²/g [7,12]. But it is hard to recover the mercury from the sorbent because of the strong interaction between sulfur and mercury. Selenium was also an efficient sorbent for Hg^0 at low temperature. But this sorbent was not widely used because the toxic of selenium. Metal and metal oxides (e.g., CoO_x , MnO_x , FeO_x) are potential sorbents for the removal of Hg^0 at high temperature, they lose the activity for Hg^0 at low temperature. Furthermore, the mercury-contaminated sorbent in the environment could cause mercury secondary pollution. There is a strong motivation to develop a low-cost and recyclable sorbent for Hg^0 capture. The mechanism of Ag amalgamation exhibits good re-generation characteristic for Hg^0 capture. Silver doped on magnetic zeolite nanoparticles were efficient and recyclable sorbent for Hg^0 [13]. Carbon nanotube–silver composite was also synthesized for Hg^0 capture as a novel carbon-based sorbent, Ag was the main active site for Hg^0 adsorption [14]. A new approach for a recyclable sorbent was used by graphene. Graphene is single-atom-thick sheet of carbon first systematically isolated from crystalline graphite in 2004 [15]. Graphene, is currently, the

most thinnest, strongest and stiffest material. Given the excellent in-plane mechanical, structural, thermal, and electrical properties of graphene, it have huge potential application in many fields [16] [17]. Graphene-based materials was used as a barriers for mercury vapor, which exhibit excellent performance for mercury vapor emission [18]. As a new member of the carbon family, there is still no attempt of the Ag/graphene based materials for the removal Hg^0 . The large surface areas make it possible as a good supporter for Ag particles.

In our study, Ag/graphene composite was synthesized by a facile method and used it as a sorbent for Hg^0 . The Hg^0 removal performance was evaluated in a fix-bed reactor. FTIR, XRD, and TEM were employed for the physicochemical characterization of Ag/graphene. We focus on the Hg^0 capture performance and the property of the sorbent's regeneration. The effect of adsorption temperature and different gases were also tested for optimizing the work condition.

2. Experimental

2.1. Sorbent preparation

Graphite oxide was prepared using Hummers method as described elsewhere [19]. In a typical synthesis method, 2 g of graphite and 1 g of NaNO_3 were added into 50 mL of 98% H_2SO_4 , followed by stirring for 1 h. After that, 0.3 g of KMnO_4 was introduced into the mixture and kept stirring for 30 min. Then another 7 g of KMnO_4 was slowly added into the above mixture in 1 h. During this period, the temperature of the solution was kept below 5 °C by ice bath. Subsequently, the solution was heated to 35–40 °C and kept stirring for 2 h. After that, 90 mL of water was slowly added into the above solution in 15 min. Finally, 50 mL of water and 7 mL of H_2O_2 were added into the as-prepared solution, following by an obvious color change from brown to yellow. The above mixture was then filtrated and washed by 10% HCl and deionized water for several times. The product was kept drying at 80 °C for 48 h to give graphite oxide. Two hundred milligrams graphite oxide was added into 250 mL deionized water. Ammonia was used to adjust the solution's pH, the value was about 7.0. Then the above mixture was operated under ultrasonic for 1 h using high-speed stirring for 1 h to give graphene oxide (GO).

Two hundred milligrams of AgNO_3 was dissolved in 15 mL of ethyl alcohol with 5 mL of H_2O . This solution was then added into GO and kept stirring for 10 h. A amount of hydrazine hydrate was added and the mixture was kept at 90 °C for 12 h. When this reduction reaction was finished, the product was filtrated and dried in an oven at 100 °C for 5 h. After that, the solid product was transferred to a muffle furnace and calcined at 250 °C for 5 h. Fig. 1 shows the synthesis process of Ag/graphene.

2.2. Sorbent characterization

Powder X-Ray diffraction (XRD) (APLX-DUO, BRUKER, Germany) was used for the detection of the crystal structure of the as-prepared samples. The scanning range was from 10° to 80° with scanning velocity of 5°/min by using $\text{Cu-K}\alpha$ radiation. The microstructure of the sorbent was analyzed by transmission electronic microscopy (TEM), and TEM image was performed on a JEOLJEM-2010 TEM device. The micrographs were obtained in the bright-field imaging mode at an acceleration voltage of 200 kV. The measurement of FTIR spectroscopy was carried out to characterize the surface properties. The N_2 sorption measurement was performed using Nova-2200 e, and the specific surface area was calculated using the Brunauer–Emmett–Teller (BET) method.

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