



Mercury adsorption from aqueous solution by regenerated activated carbon produced from depleted mercury-containing catalyst by microwave-assisted decontamination

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ABSTRACT

Mercuric chloride supported on activated carbon was widely used as the catalyst for vinyl chloride monomer (VCM) production especially in Asia. The resultant depleted mercury-containing catalyst (DMC) was a hazardous solid waste due to the high mercury toxicity. Microwave heating (MWH) was used to decontaminate DMC. Effects of microwave power and treatment time on mercury removal efficiency were studied. Up to 99.98% of mercury could be recovered under the optimum treatment conditions of 700 W for 20 min with microwave energy consumption being 15.56 kW h kg⁻¹(DMC); meanwhile the treated DMC was a clean regenerated activated carbon (RAC) without mercury leaching toxicity. The obtained RAC was a satisfactory mercury adsorbent with high specific surface area (632.9 m²/g) and abundant surface functional groups. Then the RAC was used to absorb mercury from HgCl₂ solution. Effects of pH, adsorbent dose, agitation time and initial mercury concentration on mercury sorption were investigated. Reduction and ion exchange were mainly responsible for mercury sorption mechanisms. Adsorption process conforms to pseudo-second-order kinetic model and Langmuir adsorption isotherm model. The calculated adsorption capacity of RAC was 109.05 mg/g. The absorbed mercury was efficiently desorbed by MWH. High sorption capacity of RAC maintained after five times of cyclic sorption/desorption. This new process realized the recycling of secondary resource, providing a new idea for cleaner production of VCM in engineering applications.

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1. Introduction

Mercury is listed as one of the priority pollutants according to the US EPA (Zhang et al., 2005). The ecosystem issues caused by mercury pollution have been the concern for many years all over

the world. The notorious Minamata bay event occurred between 1953 and 1956 in Japan is a typical ecological accident caused by mercury contamination (Rio and Delebarre, 2003).

Anthropogenic activities contributed most to the mercury pollution in the ecosystem (Hadi et al., 2015). Anthropogenic sources of mercury can be divided into two groups: primary anthropogenic sources and secondary anthropogenic sources (Pacyna et al., 2010). The primary sources are related to the mobilization and release of mercury of geological origin to the environment, which include minerals mining, exploitation and combustion of fossil fuels. Secondary sources involve the intentional use of mercury in commercial production, such as lamps, dental applications, batteries, electronic devices, chlor-alkali

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production and vinyl chloride monomer (VCM) production. In recent years, carbon-based materials have been proved to be highly-active catalysts for some chemical reactions (Cheng et al., 2015; Huang et al., 2017; Li et al., 2011; Cui et al., 2017). Also, HgCl₂ loaded on activated carbon (HgCl₂/AC) has long been used as the catalyst for VCM production. Early in 2005, the amount of mercury used by VCM production occupied about 20% of global mercury consumption mainly in east and southeast of Asia, among which China was a main contributor (Pacyna et al., 2010). Therefore, plenty of depleted mercury-containing catalyst (DMC) was generated each year, which is a hazardous solid waste due to the high mercury concentration (Liu et al., 2017). With the enhancement of requirements for environment protection, exploitation and import and export trade of mercury resources are regulated strictly, mercury retrieved from DMC has become the main mercury source in VCM industry.

At present, the hazardous DMC is centralized and treated by authorized corporations in China. The choice of treatment technology concerns the long-term development of an enterprise. A traditional process constituted by chemical pretreatment and distillation has been used commercially for a long time (Wang et al., 2016). Generally, it took several hours to distill the pretreated DMC at high temperature (700–800 °C) for recovering mercury. Secondary mercury pollution may be an unavoidable issue due to the long process flow. The residual catalyst support after Hg removal was generally placed in slag dump or used for industry fuel instead of reutilization. Actually the catalyst support was an activated carbon (AC) with good performance. However, it was unable to recover AC by the traditional technology. Therefore, a cleaner and more efficient method for recovering DMC will benefit the sustainable development of VCM industry.

To the best of our knowledge, very few studies associated with treatment of DMC using microwave heating (MWH) were reported. MWH has achieved considerable attention in environmental fields (Jones et al., 2002) due to the distinct advantages compared with conventional heating, such as rapid volumetric heating (Oghbaei and Mirzaee, 2010), selective heating (Harutyunyan et al., 2002), quick start-up and stopping (Ge et al., 2013), higher level of safety and automation (Menéndez et al., 2010). Therefore, MWH was used to decontaminate DMC in this study.

Adsorption technology is considered as the most convenient and simple method to remove mercury from aqueous solution or real industrial wastewater (Awual, 2017). Various adsorbents are developed in recent years (Yu et al., 2016), among which the adsorbents of interest include carbon-based adsorbents (e.g. activated carbon, activated carbon fibers, carbon nanotubes) (Hadi et al., 2015; Hadavifar et al., 2014), magnetic materials (Bao et al., 2017), nanocomposite materials (Awual, 2017), polymers (Xu et al., 2018; Sun et al., 2012), bio-adsorbents (Wang et al., 2018), solid waste derived adsorbents (Mohan et al., 2001; Mahmoud et al., 2016; Bhatnagar et al., 2015). The choice of adsorbents is usually based on the cost, adsorption capacity and regeneration performance. Some emerging materials show excellent mercury adsorption efficiency while there are still many obstacles to achieve their commercial application. Low-cost waste derived AC has been widely employed to absorb mercury from aqueous solution, which is promising for replacing commercial AC or other adsorbents for large-scale application (Huang, 1978; Koshima and Onishi, 1980;

Namasivayam and Periasamy, 1993; Kadirvelu et al., 2004; Inbaraj and Sulochana, 2006; Budinova et al., 2008; Rao et al., 2009; Zabihi et al., 2010). It is worth noting that the DMC treated by MWH is actually a regenerated activated carbon (RAC), which may be a good adsorbent for mercury uptake from mercury-containing aqueous solution.

The objectives of this study were to decontaminate DMC in a cleaner way (i.e., MWH) and use the RAC to remove mercury from HgCl₂-containing aqueous solution. Effects of microwave power and heating time (i.e. energy consumption) on mercury removal were researched. The obtained RAC was characterized by Scanning Electron Microscope (SEM), Brunauer Emmett Teller (BET), Fourier Transform Infrared (FTIR) and X-ray photoelectron spectroscopy (XPS) techniques. For mercury sorption by RAC, effects of pH, adsorbent dose, initial mercury concentration and adsorption time were studied in detail. Also, mercury sorption mechanisms, adsorption kinetics and sorption isotherms were investigated. Cyclic sorption/desorption studies of RAC were conducted.

2. Experimental

2.1. Sample and characterization

The sample, DMC was collected from a qualified enterprise for recovering hazardous mercury-containing solid waste, which is located in Guizhou province, southwest of China. DMC revealed itself as cylindrical solid with diameter and length approximately 3 mm and 2–8 mm, respectively. Concentrations of total mercury in the samples untreated and treated were determined by Cold Vapor Atomic Absorption Spectrometry (CVAAS, Shimadzu, Kyoto, Japan) after digestion using aqua regia-sodium chloride solution. Digestion method was based on Chinese national standard “Low-level mercury catalyst for chloroethylene synthesis” (GB/T 31530-2015), which has been described in our previous study in detail (Liu et al., 2018). Moisture content of DMC (wet basis) is 1.50%, and the contents of fixed carbon, volatile matters and ash are 73.72 wt%, 12.55 wt% and 12.23 wt%, respectively. The sample was dried at 80 °C for 2 h prior to subsequent experiment and characterization. The contents of other main elements were listed in Table 1.

Sequential extraction procedure containing six fractionations proposed by Hall and Pelchat (2005) was used to identify mercury species in DMC before and after microwave treatment. This method was also employed in our previous study (Liu et al., 2017). Toxicity Characteristic Leaching Procedure (TCLP) test was carried out to evaluate the mercury leaching toxicity of treated DMC (USEPA, 2002).

Concentrations and chemical states of potential elements in the samples were analyzed by a Perkin-Elmer PHI 5300 XPS. The XPS spectra were calibrated by C 1s at 284.8 eV. Microstructures of DMC before and after microwave treatment were observed by SEM (XL-30E Philips). A Nicolet avatar 360 FT-IR spectrophotometer was used to detect functional groups using dry KBr pellets in the frequency of 400–4000 cm⁻¹. BET analysis was performed in an autosorb instrument (Quantachrome) using nitrogen adsorption. Isotherms data were collected at 77 K after degassed at 423 K for 4 h. Values of specific surface area and pore size distribution were determined by BET and Density Functional Theory (DFT) model, respectively.

Table 1
Contents of main elements contained in DMC based on chemical analysis %.

C	N	H	Hg	Na	K	Si	Cl	S	Al	Ba	Ca	Fe
68.65	1.00	1.59	1.95	0.15	0.58	2.78	7.9	0.44	1.06	0.49	0.74	0.23

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