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Using H_2S plasma to modify activated carbon for elemental mercury removal

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ABSTRACT

Non-thermal plasma was applied to modify activated carbons (ACs) in the presence of H_2S for improving the elemental mercury (Hg^0) removal performance. The physical and chemical properties of raw and modified ACs were characterized by scanning electron microscopy with energy disperse X-ray spectroscopy, Brunauer-Emmett-Teller and X-ray photoelectron spectroscopy. The Hg^0 removal performance of modified AC was tested by a bench-scale fixed bed. The results indicated that H_2S plasma treatment enhanced S content of raw AC. Moreover, S content and Hg^0 removal efficiency of modified AC increased with the increase of H_2S concentration. The reason was that higher concentration H_2S resulted in more S active sites on modified AC and S active site played a significant role during the Hg^0 removal process. Besides, non-thermal plasma treatment in the presence of H_2S had a weak impact on BET surface area. By further analysis, it could be inferred that the improvement of Hg^0 removal performance was ascribed to the chemisorption rather than physisorption. Combined the results of temperature programmed desorption and XPS analysis, the mechanism of Hg^0 removal was that elemental S on the surface of modified AC reacted with Hg^0 to form HgS.

1. Introduction

Mercury has attracted a great deal of attention due to its toxicity, bioaccumulation and persistence [1,2]. And the emission of mercury can cause many diseases such as immune, sensory and neurological dysfunctions [3]. According to relevant statistical data, the total quantity of anthropogenic mercury emission is about 1000 tons every year and coal-fired power plant has been considered to be the major anthropogenic mercury emission [4–7]. There are mainly three chemical forms of mercury in coal-fired flue gas: oxidized mercury (Hg^{2+}), particulate mercury (Hg^p) and elemental mercury (Hg^0) [8,9]. Hg^{2+} and Hg^p can be easily removed by existing air pollution control devices (APCDs) [10,11]. However, Hg^0 is difficult to be removed due to its low reactivity, low solubility and high volatility [12]. Many methods have been developed to transform Hg^0 into Hg^{2+} or Hg^p [13–16]. Among these methods, activated carbon injection (ACI) technology has been regarded as one of the most efficient ways to capture Hg^0 in coal-fired flue gas [6]. Yet, some crucial factors such as a high C/Hg ratio and large operating costs limit the popularization of ACI technology in developing countries [17]. Therefore, some researchers put forward halogen impregnated methods to modify activated carbon for improving the Hg^0 removal performance [18–21]. Halogen-impregnated AC can remove Hg^0 effectively, but mercury on the surface of modified AC will be released under acid conditions [22,23]. Vidic et al. presented that the adsorption of Hg^0 by sulfur-impregnated carbon was more stable than that by chlorine-impregnated carbon [24]. Thus, Suresh Kumar Reddy heated powdered virgin porous carbon mixed with elemental sulfur at 544 °C for 43 min to promote Hg^0 removal [25]. University of Pittsburgh reported several sulfur forms existed in activated carbon by reacting with H_2S [26–28].Although these modification methods are effective for Hg^0 removal, their process is time-consuming and high energy consumption. Hence, an effective and simple method should be proposed to modify AC for improving the Hg^0 removal performance.

Non-thermal plasma (NTP) can produce high-energy electrons, ions and active radicals to increase the active species on the surface of sorbents [29,30]. It has been regarded as one of the most promising methods to modify sorbents during the past several years [31,32]. Thus, our previous studies applied NTP to modify sorbents for improving the Hg^0 removal performance [8,33]. Meanwhile, other researchers modified AC by NTP to increase the amount of chlorine or oxygen functional groups for enhancing the Hg^0 removal efficiency [34–36]. But a small amount of mercury species on the surface of modified AC were unstable and they may be released under acid conditions [27,28,37]. Mercuric sulfide (HgS) is insoluble in water and it is one of the most stable

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