



Gas-phase mercury removal through sulfur impregnated porous carbon



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ABSTRACT

Gas phase mercury removal is a vital unit operation in gas processing industries. The present work attempts to prepare a sulfur impregnated carbon at optimized experimental conditions and compares its elemental adsorption capacity with the number of commercially available carbon based adsorbents. The effect of adsorption temperature on mercury adsorption capacity has been estimated for the prepared sulfur impregnated carbon. The adsorption capacity was found to increase with increase in adsorption temperature owing to the chemisorption nature of the adsorption. The adsorption isotherms were generated at three different temperatures and were found to close adhere to the Langmuir Isotherm model. The adsorption capacity was found to increase until 140 °C, while decrease beyond, which was attributed to the softening and agglomeration of sulfur. The maximum adsorption capacity of 4325 µg/g was observed at a temperature of 140 °C. A comparison of the relative adsorption capacity of various adsorbent at 30 °C, revealed the adsorption capacity of the sulfur impregnated carbon prepared in the present work much higher than the commercially available carbons. The high adsorption capacities with simple preparation techniques favor the commercial mercury adoption process.

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1. Introduction

Emissions of mercury is a major environmental concern since mercury is considered to be one of the most toxic metals found in the environment and additionally a hazardous air pollutant (HAP) as per the recommendations of the Clean Air Act (CAA) of 1990. Mercury has been recognized to cause both acute and chronic toxicity on the nervous, renal, and reproductive systems [1]. Mercury discharged into the atmosphere persists in the environment and creates long term contamination problem. Furthermore, food chain transport, bioaccumulation, high toxicity to mammals and severe health problems due to ingestion of mercury are well documented even at low levels rendering a strict control of mercury emission mandatory. Although mercury in natural gas is normally at low levels, some sources contain sufficiently high mercury concentrations to cause both safety and health concerns. Abbott et al. [2] have reported presence of much higher mercury levels in individual wells and that the highest known concentration is 4400 µg/m³ (500 ppbv) from a well in Germany. Rios et al. [3] have registered higher levels of elemental mercury in the

Southeast Asian countries, while lower levels in United States and Gulf coast.

Recently mercury emission from flue gas in natural gas industries has become a major environmental concern, necessitating the need for efficient mercury removal technology. Mercury exists in flue gases in various forms as elemental (Hg⁰), oxidized (HgCl₂ or HgO), and particulate (Hg_(p)). Among these elemental mercury removal is quite difficult due to its high volatility, and chemical inertness. Many methods are in practice to control mercury emission which, include adsorption [4] and photochemical oxidation [5–7], however, adsorption using porous carbon is adopted widely in industry [8].

Porous carbon is a well-established adsorbent for elemental mercury removal from natural gas [4,9–20], however virgin carbon is reported to be in-effective for most practical applications. For most practical applications, the adsorption capacity of the porous carbon is significantly improved by surface modification using sulfur, halides, and thiols. Augured by the importance of the problem and potential commercial application, number of reports are available in the open literature, addressing various ways of improving the adsorption capacity of the virgin carbon [21]. These techniques include introduction of various impregnates such as sulfur, iodine, chlorine, bromine on to porous carbon [22–26], zeolites [27], and fly ash [28]. An analysis of the literature provides a clear knowledge on the state of art of the impregnation

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techniques and availability of low cost adsorbents for mercury removal. Porous carbon is an excellent adsorbent for controlling mercury emissions, due to its high specific surface area, pore volume, wide pore size distribution and easy modify the surface characteristics as required.

The adsorption mechanism is well known to be classified either as physisorption or chemisorption. Physisorption involves no chemical reaction between carbon and mercury, where the elemental mercury condenses from vapor phase into liquid on the pores of the porous carbon. These condensed mercury compounds are loosely bonded to the carbon, and the strength of the bond is very sensitive to temperature changes leading to desorption at elevated temperatures. Chemisorption of elemental mercury occurs on the active sites present in porous carbon. The resultant product forms non-volatile species with elemental mercury which remains permanently on to the porous carbon matrix.

Presence of mercury in natural gas is a serious concern to natural gas processing industries and most of it incorporates a mercury removal technique in the process. Due to the high vapor pressure of the elemental mercury it does not condense under the gas processing conditions, being entrained with the non-condensable gas. For efficient distribution, the natural gas is compressed at high pressure which consequently increases the concentration of mercury, is of serious concern to the gas processing industry. High concentrations of mercury in natural gas are well known to cause severe corrosion to downstream aluminum heat exchangers. The presence of mercury additionally is known to contribute to (i) exposure of workers to high levels of mercury during maintenance operations, (ii) corrosion of process equipment due to liquid metal embrittlement or amalgam corrosion, (iii) poisoning of expensive downstream precious metal catalysts such as platinum or palladium, (iv) difficulty in disposal of mercury contaminated equipment, and (v) emissions to the environment.

The magnitude of above problems demands better understanding of the fundamental mechanisms of mercury adsorption, which include the effect of different adsorbents, the effect of operating

temperatures, moisture content, oxygen partial pressure and presence of other compounds on the adsorption capacity and kinetics of adsorption on to activated carbons.

The objective of this work is to identify sulfur impregnation protocol on porous carbon to maximize the elemental mercury adsorption capacity. In addition an attempt to compare the relative adsorption capacities of the carbon developed using process optimization technique [29] with commercially available popular carbons such as CALGON, NORIT, and UOP. Further, the work relates to assessing the effect of operating temperature and mercury concentration on the adsorption capacity. The morphology of the various carbons samples were also assessed through SEM analysis, which was additionally utilized to analyses the adsorption results.

2. Experimental details

2.1. Sample preparation

Sulfur impregnation was performed using elemental sulfur mixed with a fixed amount of powdered virgin porous carbon (Calgon-SGL) available commercially. Experiments were conducted at the standard set of conditions, which were based on the process optimization studies conducted prior to the present work [29]. The process conditions for preparation of sulfur impregnated carbon are, an impregnation temperature of 544 °C, impregnation time of 43 min and carbon to sulfur ratio of 0.53. The prepared sulfur impregnated carbon will be referred in the rest of the paper as sample E11(S).

2.2. Sample characterization

The BET surface area, along with the pore size distribution was estimated using the standard nitrogen adsorption isotherm, Autosorb 1-C, Quanta Chrome Instruments, USA. Prior to analysis, the samples were first dried in an oven at 130 °C overnight and

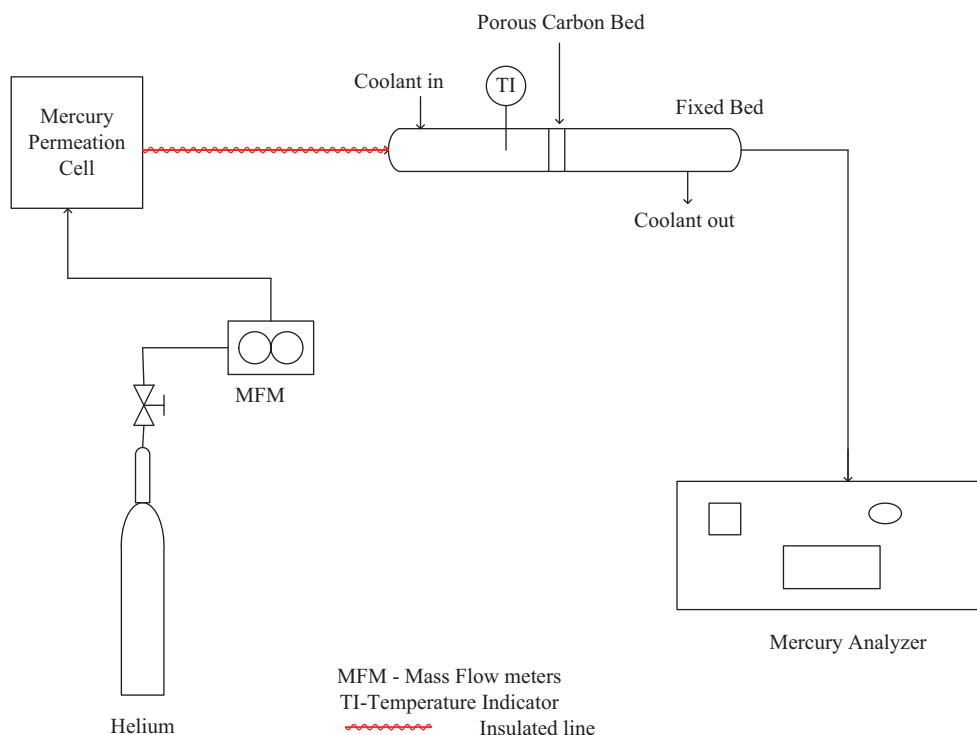


Fig. 1. Elemental mercury adsorption setup.

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