

Optimization of Fixed-Bed Design for Natural Gas Mercury Removal by Sulfur Doped into Porous Activated Carbon

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Abstract

The present work reports the synthesis and application of sulfur doped into porous activated carbon for removing elemental mercury from natural gas using a bench-scale fixed-bed reactor. A series of experiments were carried out to investigate the optimization of Hg⁰ capture. Furthermore, our experimental results about optimum conditions to remove Hg⁰ were 1:10 of sulfur to activated carbon impregnation ratio, 350°C of impregnation temperature, and 3 hours of impregnation time. This research showed that the prepared adsorbents were capable to remove remarkable amount of Hg⁰ (23.615 mg/g) at high adsorption efficiency. This study may serve as reference on natural gas power plants for the removal of Hg⁰ using the same conditions.

Keywords

Porous activated Carbon, Mercury Adsorption Mechanism, Natural Gas, Elemental Sulfur Impregnation

1. Introduction

Natural gas is one of the three major fossil fuels sources of energy including petroleum, and coal. Natural gas is a versatile, clean-burning, and efficient fuel accounted for most of the energy production [1].

While recently, mercury is recognized as a toxic metal, its presence has been a serious concern to natural gas processing plants [2] [3] [4] [5]. Mercury is among the current top environmental challenges due to a rapid industrial growth

that drives technology developments for mercury free hydrocarbons [6] [7] [8]. Some studies suggested that, the exposure to mercury may leads to receive global emphasis through its continuing and serious harm to human health effects ranging from acute to chronic diseases [2].

Consequently, the research about how to remove Hg^0 is critically needed. Mercury appears into three forms in the flue gas including elemental mercury (Hg^0), oxidized mercury (Hg^{2+}) and particulate mercury (Hg^p) [6] [9]-[17].

As recommended by the Clean Air Act (CAA) of 1990 [1] [4]; Hg^0 is considered highly volatile, insoluble and additionally a hazardous air pollutant (HAP). While many adsorbents are effective in removing elemental mercury, activated carbon serves as an adequate carrier for various chemicals [14], which physically react with the mercury and hold it within the adsorbent particles [2]. Activated carbon is a kind of pore structure developed and chemically stable produced from coal and coconut shells [11].

In the previous literatures, the adsorption of elemental mercury using raw activated carbon has been expressed as a good method. Results showed that the elemental mercury adsorption capacity level using raw carbon is typically weak [14] [18].

For getting high Hg^0 removal efficiency, the activated carbon needs to be able to adapt to the complex pore structure with large number of suitable impregnating pores so that the gas processing can be efficiently done [2]. As a result, elemental sulfur impregnated porous activated carbon has been adopted in order to improve and gives more significant Hg^0 adsorption capacity [15] [19].

The sulfur strongly bonded to the activated carbon is more reactive and the mechanism for mercury adsorption is governed by the reaction between active sulfur atoms (S_2 - S_4) which are the macromolecular sulfur broken down though resulting the high elemental mercury adsorption capacity as reported by Yaxuan Yao and his team [14] [18]. The impregnation temperature dictates the predominant form of sulfur allotropes [5] [19].

Even though Hg^0 adsorption capacity increases with sulfur impregnation temperature, at lower impregnation temperatures, sulfur molecules are mainly in the form of rings or long linear chains [13] [14]. Although these molecules will have little steric hindrance for oversized pores, they may form barriers in the medium size pores [14] [20] [21]. As these large sulfur molecules attach to the activated carbon surface, they tend to block the entrance to medium pore openings [20] [21]. It can be concluded that the actual form of sulfur rather than the total sulfur content is a crucial factor governing the chemisorption process [3] [4] [22] [23].

Recent study have showed that the highly adsorptive porous carbon can be prepared by high temperature sulfur impregnation, authors performed a series of experiments for removing mercury from natural gas by employing elemental sulfur doped coconut husk porous activated carbon, this study has found that factors such as the impregnation temperature and impregnation ratio were the most important factors played a critical role for the hole process of mercury re-

removal [14] [15] [19]. The amount of mercury adsorbed by sulfur doped activated carbon found to be uneven to that estimated by the stoichiometry of the reaction which gives HgS [5] [17], which led to describe how some moiety of the doped sulfur does not intervene in the reaction with elemental mercury vapor gas. The unreacted sulfur is considered to be chemically adsorbed and stable [9].

The main objective of this research was to optimize Sulfur doped into Porous Activated Carbon adsorbent preparation focusing on the art of the impregnation technique with high-temperature between 300°C - 500°C for natural gas to remove the Hg⁰ using bench-scale fixed bed reactor.

2. Materials and Methods

2.1. Materials and Chemicals

First, Ceramic boats (25 mL), BSA124S Electronic balance (Sartorius), Desiccator were used in this research. The chemicals used were Coconut husk crushed Activated carbon (Dalian, China), and sulfur sublimed (99.99%) purchased from Tianjin damao reagent Factory (Tianjin, China), ionized water used throughout the whole experiments were prepared in our department.

2.2. Equipments

GSL-1100X Tubular furnace (Nantong Rite Scientific Research Instruments Co., Ltd.), DHG-9070A Electric Drying oven (Shanghai Yiheng Scientific Instruments Co., Ltd.), QM208B Atomic absorption mercury analyzer (made by Suzhou Qing'an Instrument Co., Ltd.), mercury permeation device was made by Dahua Instrument Factory (Shanghai, China), and Nitrogen gas (purified, 99%) was obtained from Chemical Physics Institute (Dalian, China), Jade 6.5, Origin Pro 8, AutoCAD 2016, CASA XPS Software were used in the experiments.

2.3. Preparation of Adsorbents

The experimental study was performed through three steps. Sulfur doped activated adsorbent preparation was carried out using the elaborated techniques.

Impregnating sulfur in activated carbon procedure was developed based on several control parameters. Among them, two most important factors are the impregnation temperature and the initial sulfur carbon ratio in the impregnation furnace.

Different sulfur doped porous activated carbon were prepared and compared. Firstly, a fixed amount of the virgin coconut husk porous activated carbon were crushed in a grinder for 1 hour, ground into 20 × 40 mesh size [9], sieved, then rinsed with ionized water for several times and dried in an oven at 200°C for 12 hours, after they were placed in a desiccator till further use. And then another fixed amount of powdered elemental yellow sulfur high-purity (99.99%) were physically mixed together for the impregnation process in one ceramic bowl and put in tubular furnace for being heated at 250°C - 600°C for 3 - 6 hrs. Experiments were conducted at the standard set of conditions, which were based on the process optimization studies conducted prior to the present work [9].

The pathway made up of mixing raw coconut porous activated carbon placed evenly in a ceramic boat, and a predetermined amount of sulfur in the same ceramic boat with a specific sulfur carbon ratio (1:20 - 1:4). **Figure 1** shows the structure of the experimental system used for the elemental sulfur impregnation on the solid substrate.

An inert atmosphere was made within the tubular furnace then the mix was put under a nitrogen stream with a fixed flow rate of 60 mL/min for 20 min to completely remove traces of oxygen resulting in a certain pressure inside the preparation conditions [24], then the sulfur-doped porous activated carbon was taken out from the tubular furnace. In this way, activated carbon for removal of mercury gas was prepared. Finally, the prepared sulfur doped porous activated carbon cooled to the room temperature ($30^{\circ}\text{C} \pm 10^{\circ}\text{C}$) and was stored in the desiccator to prevent the humidity. The Sulfur content and Sulfurization rate were determined through the following formula [20]:

$$\frac{S}{C} = \frac{M_{\text{sulfur}}}{M_{\text{activated carbon}}} \quad (1)$$

$$\text{Sulfurization rate} = \frac{M_{\text{adsorbed}}}{M_{\text{sulfur}}} \quad (2)$$

where by:

M_{sulphur} is—the mass of added sulfur, g;

$M_{\text{activated carbon}}$ is—the activated carbon added, g;

M_{adsorbed} is—sulfur absorbed by activated carbon, g.

2.4. Physical Characterization

Firstly, the sample of 20 g was sieved between 180 - 200 mesh size before doing characterization. Given that the adsorption capacity of these adsorbents is

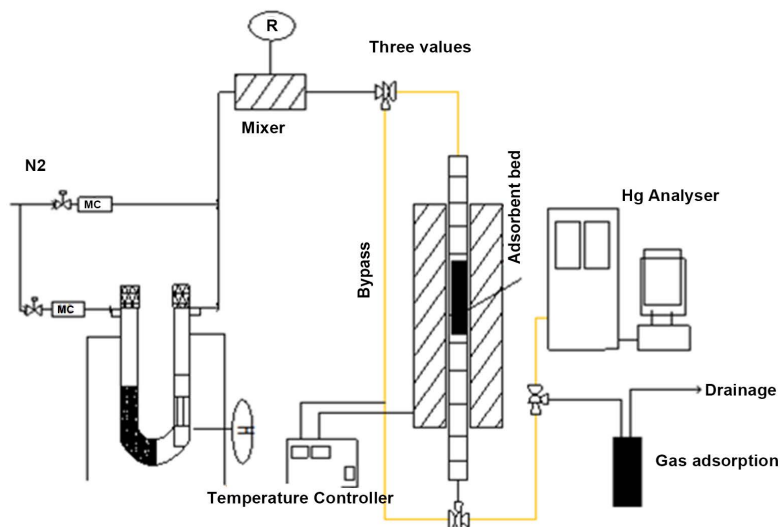


Figure 1. A schematic diagram of mercury removal by sulfur doped activated carbon experimental device. 1-Mixer; 2-Three way valves; 3-Bypass; 4-Temperature controller; 5-Adsorbent bed; 6-Hg Analyser; 7-Drainage; 8-Gas adsorption.

strongly related to the actual forms of sulfur within the carbon particles, the interaction between the carbon and sulfur, and the microstructure of the carbon particles, it is necessary to consider the physical and chemical characteristics at a microscopic level using XRD and XPS techniques [18] [21] [25]. Generally, the sulfur content on the porous activated carbon surface, the yield of sulfurization on activated carbon surface and sulfur bondage were the main parameters used to describe the textural properties of an adsorbent [18] [21] [25].

2.5. Hg⁰ Adsorption Experiment

The mechanism of elemental mercury (Hg⁰) captive at 60°C by sulfur doped porous activated carbon using a bench-scale fixed-bed device, DCW3015 thermostatic bath, mercury permeation device and QM208B Atomic absorption mercury analyzer has been tested. Physisorption and chemisorption are coupled to give the whole Hg⁰ adsorption mechanism. N₂ was used as the carrier gas. Mercury adsorption tests were conducted with activated carbon mass 100 mg, placed inside a tubular reactor having a length of 0.1 m and diameter of 0.0254 m. The reactor was covered with water circulation to maintain the desired adsorption temperature inside the reactor.

A mercury permeation device was used as a source to generate the mercury vapors at the desired Hg⁰ concentration and flow rate. The mercury adsorbed in the porous activated carbon was operated by using automatic mercury analyzer and the mercury adsorption capacity was determined by integrating the area above the breakthrough curve.

In order to evaluate the removal characteristics of sulfur doped activated carbon for Hg⁰ in the natural gas purification, the following equation of Hg adsorption efficiency was employed [21] [23].

Where the Mercury removal efficiency of sulfur doped activated carbon η and the amount of mercury adsorbed q are [24] [25] [26]:

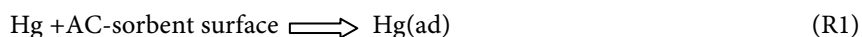
$$\eta = (C_{out}/C_{in}) \times 100\% \quad (3)$$

$$q = \left[C_{in} \int_0^t Q \left(1 - \frac{C_{out}}{C_{in}} \right) dt \right] / m \quad (4)$$

where, (η) is the removal efficiency (%);

(q) is the mercury adsorption capacity (mg·g⁻¹);

C_{out} and C_{in} are outlet and inlet Hg concentration (μg·m⁻³).



3. Results and Discussion

3.1. Yield of Sulfurization

The yield of sulfurization was defined as the weight of the final sulfurized samples to the weight of primary raw activated carbon, in this research 3 g of activated carbon was used for each experiment. To facilitate direct comparison of these prepared mercury sorbents, with the previous ones, the yield of sulfuriza-

tion decreases with the increase of temperature when the impregnation S/C ratio is the same, while the yield of sulfurization increases with the increase of S/C Ratio when the impregnation temperature is the same [5].

According to **Table 1**, at higher temperature like from 500°C used in this study, larger amounts of physically adsorbed sulfur can be vaporized, be converted into sulfur functional groups, and released from the pore surface of activated carbon; therefore, the lower yield would be obtained at this temperature. Compared to the S/C Ratio where the sulfur content on the surface of activated carbon is less, the resulting yield would be smaller, as it is for the case of SAC-400 (1:10).

3.2. Effect of Impregnation Temperature on Hg⁰ Adsorption Capacity

Table 2 illustrates the SAC-300, SAC-350, SAC-400, SAC-450, SAC-500 five different types of elemental sulfurdoped porous activated carbon. From this table, when the ratio is the same, the sulfur content on the activated carbon surface is different after application of high temperature and the highest sulfur content is obtained for SAC-300. According to the previous studies, when the impregnation temperature was low, had a much lower capacity for mercury removal [5] and as the impregnation temperature increased, the adsorption capacity for Hg⁰ removal also increased as suggested by [3].

For the prepared sorbent with 8.33% of sulfur content where the sulfur impregnation temperature reached 500°C, the sulfur content of the prepared sulfur porous activated carbon decreased remarkably, since 444°C boiling point of Sulfur attained Hg⁰ adsorption capacity decreased due to the high amount of Sulfur evaporated.

As the temperature increased, the new bond between Sulfur and Activated Carbon was formed, and then Hg⁰ adsorption capacity increased like for the case of SAC-400(1:5) and SAC-450(1:5).

3.3. Effect of Impregnation Ratio on Hg⁰ Adsorption Capacity

The sulfurization rate was calculated and included into **Table 2** and **Table 3**. The low sulfurization rate shows that most of the sulfur did not react with activated carbon. Sulfurization rate decreased with a decrease in pore volume, which is likely due to reduced accessibility for the reaction with activated carbon. The rising rate shows the combination of sulfur and porous activated carbon.

Owing to the results given in **Table 3**, it can be interpreted that the Hg⁰ adsorption capacity is related to the sulfur content, sulfur bondage and porous structure chemistry of Activated Carbon which is the key factor that intervenes for the synthesis of the sorbent. When S/C is low, sulfur tends to spread to activated carbon. Internally, resulting in adsorption of sulfur functional groups attached to the pores of Activated carbon. A part of sulfur contained on the outer surface of activated carbon. Also, with the increase of S/C ratio, at 450°C (1:10)

Table 1. Yield of sulfurization for different prepared SAC adsorbents.

Sample	Impregnation temperature (°C)	S/C Ratio	Yield of sulfurization
SAC-350	350	1:10	1.094
SAC-400	400	1:10	1.088
SAC-400	400	1:4	1.226
SAC-450	450	1:4	1.210
SAC-500	500	1:4	1.182
SAC-400	400	1:5	1.183
SAC-450	450	1:5	1.175

Table 2. The Hg⁰ adsorption capacity obtained using prepared SAC at different impregnation temperature.

Sample	Impregnation temperature (°C)	S/C Ratio	Sulfur content wt (%)	Sulfurization rate (%)	Hg adsorption capacity (mg/g)
AC	-	-	0.18	-	0.278
SAC-300	300	1:5	19.95	99.76	0.366
SAC-350	350	1:5	18.66	93.33	0.322
SAC-400	400	1:5	18.36	91.83	4.072
SAC-450	450	1:5	17.53	87.66	5.247
SAC-500	500	1:5	8.33	41.66	0.371

Table 3. The Hg⁰ adsorption capacity obtained using prepared SAC with different S/C ratio.

Sample	S/C Ratio	Impregnation temperature (°C)	Sulfur content wt (%)	Sulfurization rate (%)	Hg adsorption capacity (mg/g)
SAC-450	1:4	450	21	84	2.339
SAC-450	1:5	450	17.53	87.66	5.247
SAC-450	3:20	450	11.83	78.88	1.694
SAC-450	1:10	450	9.97	99.83	0.590
SAC-450	1:20	450	4	80	0.322

the sulfurization rate increased reaching 99.83% while at 450°C (3:20) the sulfurization rate appeared to be the lowest 78.88%.

Tested results were compared in **Table 3**. In order to check the efficient sulfur Activated Carbon, it can be found that the adsorbed amounts of sulfur at any instant increased markedly due to the increase of sulfur amounts. However, sulfurization rate was not only depending on the Sulfur amounts used. Rather, sulfurization rate of prepared sulfur impregnated activated carbon varied according to the ratio S/C used where at the percentage around 10% the sulfur impregnation decreased [19]. This effect is shown at the S/C ratio of 9.97% and 11.87% found to be in the range of the favorable ratio where most of sulfur content is

absorbed, this is proved by the sulfur content of elemental sulfur impregnated activated carbon sold on the market which is now 10% to 11% [21].

3.4. The Effect of Impregnation Time on Sulfur Loss

From **Table 4**, the effective adsorbent was prepared using sample with same S/C ratio at different impregnation temperature. Using impregnation time of 3 hours, it has been realized that the activated carbon absorbs sulfur at a faster rate, the total pore volume of activated carbon is larger and sulfur is easily adsorbed on the surface of activated carbon. As impregnation time increases the total pore volume of activated carbon gradually decreases, and the sulfurization rate slows down, where the high sulfur loss occurred at 500°C (1:10). In the previous study, the Sulfur loss increased with the increase of impregnation temperature [20].

No matter which impregnation temperature, the S/C ratio of 1:10 has remarkably shown as the best results comparing to the previous experiments done in this research. Even if at 500°C (1:10) the Sulfur loss increased till 44%, the Hg⁰ removal efficiency has been calculated considering the same obtained Hg outlet concentration 40 µg/m³.

3.5. XRD Characterization

Figure 2(a) and **Figure 2(b)** show XRD analysis before and after adsorption of sulfur doped porous activated carbon and XRD spectrum before and after adsorption of mercury by sulphur doped porous activated carbon. The spectrum of the SAC is basically the same, and the diffraction angle is 2 Theta. The two peaks at 26 degree and 43 degree indicate the indefinite form of SAC.

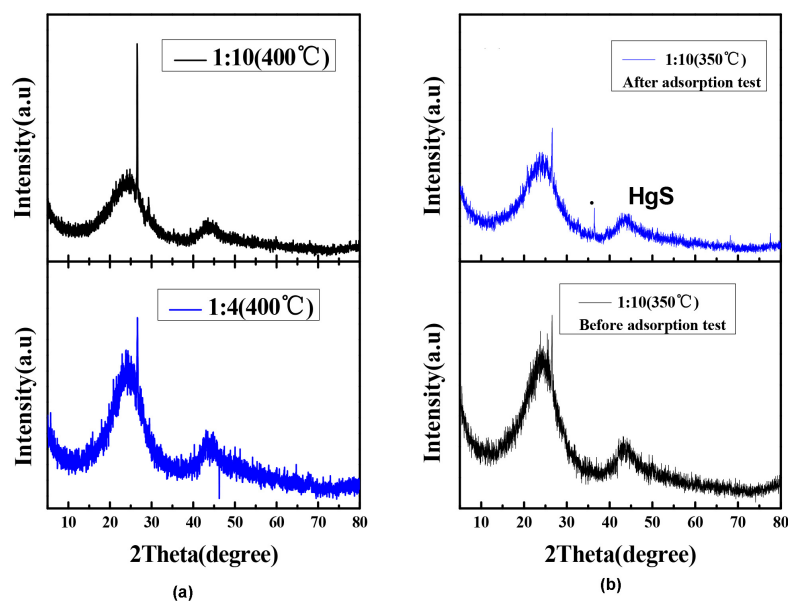
It also indicates that sulfurdoped activated carbon is not directly impregnated. The crystal morphology of Sulfur has its surface, mainly in amorphous form. Chemical load is the main factor. S-AC spectra before and after adsorption are compared. **Figure 2(a)** 1:10 (400°C) shows the peak at 26.543, 36.173 and at 42.963 while 1:4 (400°C) has the peak at 26.4 and at 43.086 **Figure 2(b)**. The difference lies in the 36.71 degree peak, which is divided by software Jade 6.5. The peak of 36.71 degree can be regarded as HgS (JCPDS75-1589). The characteristic peak shows that the adsorption of mercury in the activated carbon table is in the process. S atoms react with mercury to form HgS. **Figure 2(b)** before adsorption test 1:10 (350°C) has the peak at 26.5 other at 43.0 while after adsorption test 1:10 (350°C) shows the peak at 26.8 and at 43.0.

3.6. XPS Characterization

XPS characterization was carried out for the identification of sulfur element on Coconut husk porous activated carbon surface; obtained spectra are shown in **Figure 3(a)** and **Figure 3(b)**. Referring to the standard library spectra, the ensuing binding energy data are related to sulfur species; whereby s: free elemental sulfur has a peak around 164.05 eV; chemisorbed sulfur has a peak at 161.8 -

Table 4. The comparison of sulfur loss using samples prepared at different Sulfur impregnation temperature.

Sample	Impregnation temperature (°C)	S/C Ratio	Hg adsorption Capacity (mg/g)	Sulfur loss (%)
SAC-350	350	1:10	23.615	0.6
SAC400	400	1:10	3.279	1.3
SAC450	450	1:10	0.590	12
SAC500	500	1:10	0.349	44

**Figure 2.** (a) XRD comparing samples with different S/C ratios at 400°C (b) XRD graph comparing SAC at 350°C (1:10) before and after adsorption test.

162.6 eV; Unbound organic sulfur species, like thiophene, also show a peak between 163 - 164.1 eV; and oxidized sulfur shows a peak above 167 eV.

The sulfur on Activated Carbon surface is present mainly in free elemental form with negligible amounts of oxidized sulfur forms. **Figure 3(a)** and **Figure 3(b)** show the dominant weight loss at 1:10 (450°C) more than at 1:10 (350°C) explaining that as the temperature range increased the weight loss increased.

As shown in **Figure 3(a)** and **Figure 3(b)**, the peaks at about 161.8 and 162.6 eV correspond to chemisorbed sulfur. Meanwhile, the sulfur content increased. The peaks at about 164.08 correspond to elemental sulfur.

Unfortunately, there is an overlap between the region of elemental sulfur and that of organic sulfur. The results depicted in **Figure 3(a)** suggest that either organic sulfur or elemental sulfur was the dominant sulfur form on the AC surface. According to [12], thiophene may be the possible structure of organic sulfur products deposited on the carbon surface at high temperatures.

In **Figure 3(b)** the spectra were referenced to the Hg 4f binding energy setting to 100.7 eV for Hg⁰ and to 100.9 eV for HgS. Data acquisition and peak fitting were performed by the CASA XPS software.

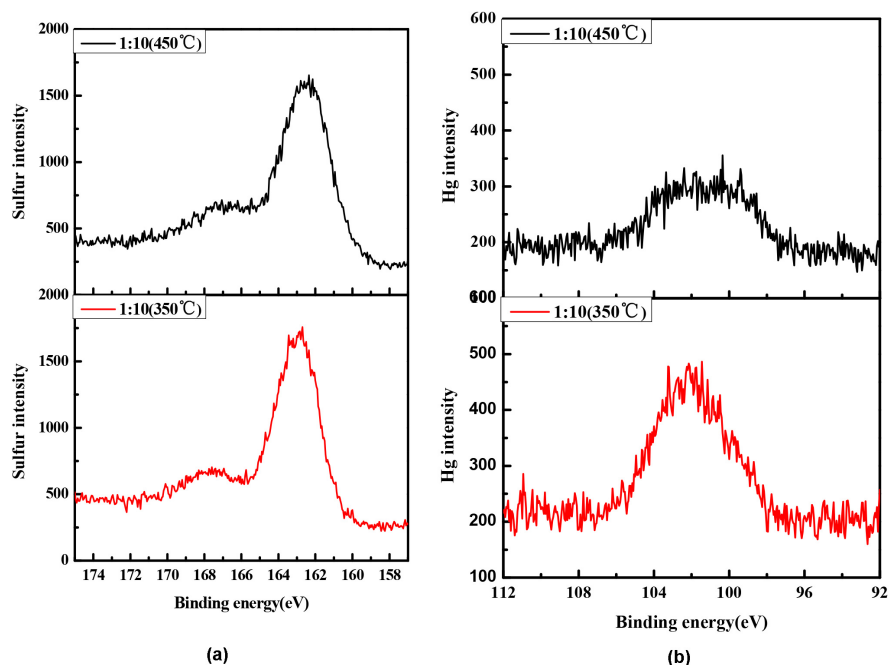


Figure 3. XPS graph comparing Sulfur intensity and Mercury intensity (a) XPS graph comparing Sulfur intensity (b) XPS graph comparing Mercury intensity.

The samples were analyzed by XPS to identify the surface characteristics of the active species. The representative photoelectron peaks of Hg 4f to the previous samples 1:10 (350 °C) and 1:10 (450 °C) are identified by an essential difference reflecting in the behaviour of the Hg 4f lines.

4. Conclusion

Mercury is naturally existing element that is mostly found in air, water and soil. A number of studies have suggested that the exposure of mercury even negligible amounts may lead to very serious health problems. This study is aimed to remove elemental mercury from natural gas by employing a bench-scale fixed-bed reactor using sulfur doped into porous activated carbon as adsorbent agent. Our findings showed that the obtained Hg⁰ adsorption capacity using sulfur doped porous activated carbon was obviously higher than that of raw activated carbon and after optimizing all conditions such as impregnation temperature and S/C ratio appreciable amount of mercury was efficiently removed with a high Hg⁰ adsorption capacity suggesting that this method with these optimized conditions may be applied in real life to remove the mercury from environment.

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Conflicts of Interest

The authors have declared no conflict of interest.

References

- [1] Faramawy, S., Zaki, T. and Sakr, A.A.E. (2016) Natural Gas Origin, Composition, and Processing: A Review. *Journal of Natural Gas Science and Engineering*, **34**, 34-54. <https://doi.org/10.1016/j.jngse.2016.06.030>
- [2] Yan, R., Liang, D.T., Tsen, L., Wong, Y.P. and Lee, Y.K. (2004) Bench-Scale Experimental Evaluation of Carbon Performance on Mercury Vapour Adsorption. *Fuel*, **83**, 2401-2409. <https://doi.org/10.1016/j.fuel.2004.06.031>
- [3] Liu, W., Vidić, R.D. and Brown, T.D. (1998) Optimization of Sulfur Impregnation Protocol for Fixed-Bed Application of Activated Carbon-Based Sorbents for Gas-Phase Mercury Removal. *Environmental Science & Technology*, **32**, 531-538. <https://doi.org/10.1021/es970630+>
- [4] Suresh Kumar Reddy, K., Al Shoaibi, A. and Srinivasakannan, C. (2014) Gas-Phase Mercury Removal through Sulfur Impregnated Porous Carbon. *Journal of Industrial and Engineering Chemistry*, **20**, 2969-2974. <https://doi.org/10.1016/j.jiec.2013.10.067>
- [5] Asasian, N. and Kaghazchi, T. (2013) Optimization of Activated Carbon Sulfurization to Reach Adsorbent with the Highest Capacity for Mercury Adsorption. *Separation Science and Technology*, **48**, 2059-2072. <https://doi.org/10.1080/01496395.2013.780833>
- [6] Pavlish, J.H., Holmes, M.J., Benson, S.A., Crocker, C.R. and Galbreath, K.C. (2004) Application of Sorbents for Mercury Control for Utilities Burning Lignite Coal. *Fuel Processing Technology*, **85**, 563-576. <https://doi.org/10.1016/j.fuproc.2003.11.022>
- [7] Liu, Y., Kelly, D.J.A., Yang, H., Lin, C.C.H., Kuznicki, S.M. and Xu, Z. (2008) Novel Regenerable Sorbent for Mercury Capture from Flue Gases of Coal-Fired Power Plant. *Environmental Science & Technology*, **42**, 6205-6210. <https://doi.org/10.1021/es800532b>
- [8] Kumar Reddy, K.S., Prabhu, A., Al Shoaibi, A. and Srinivasakannan, C. (2016) Application of Sulfonated Carbons for Mercury Removal in Gas Processing. *Energy and Fuels*, **30**, 3227-3232. <https://doi.org/10.1021/acs.energyfuels.5b02630>
- [9] Suresh Kumar Reddy, K., Al Shoaibi, A. and Srinivasakannan, C. (2014) Elemental Mercury Adsorption on Sulfur-Impregnated Porous Carbon—A Review. *Environmental Technology*, **35**, 18-26. <https://doi.org/10.1080/21622515.2013.804589>
- [10] Vidic, R.D. and Siler, D.P. (2001) Vapor-Phase Elemental Mercury Adsorption by Activated Carbon Impregnated with Chloride and Chelating Agents. *Carbon*, **39**, 3-14. [https://doi.org/10.1016/S0008-6223\(00\)00081-6](https://doi.org/10.1016/S0008-6223(00)00081-6)
- [11] El-Feky, A.A., El-Azab, W., Ebiad, M.A., Masod, M.B. and Faramawy, S. (2018) Monitoring of Elemental Mercury in Ambient Air around an Egyptian Natural Gas Processing Plant. *Journal of Natural Gas Science and Engineering*, **54**, 189-201. <https://doi.org/10.1016/j.jngse.2018.01.019>
- [12] Liu, W., Vidic, R.D. and Brown, T.D. (2000) Impact of Flue Gas Conditions on Mercury Uptake by Sulfur-Impregnated Activated Carbon. *Environmental Science & Technology*, **34**, 154-159. <https://doi.org/10.1021/es990315i>
- [13] Vidic, R.D. and McLaughlin, J.B. (1996) Uptake of Elemental Mercury Vapors by Activated Carbons. *Journal of the Air & Waste Management Association*, **46**, 241-250. <https://doi.org/10.1080/10473289.1996.10467458>
- [14] Yao, Y., Velpari, V. and Economy, J. (2014) Design of Sulfur Treated Activated Carbon Fibers for Gas Phase Elemental Mercury Removal. *Fuel*, **116**, 560-565. <https://doi.org/10.1016/j.fuel.2013.08.063>

- [15] Wdowin, M., Wiatros-Motyka, M.M., Panek, R., Stevens, L.A., Franus, W. and Snape, C.E. (2014) Experimental Study of Mercury Removal from Exhaust Gases. *Fuel*, **128**, 451-457. <https://doi.org/10.1016/j.fuel.2014.03.041>
- [16] Chen, S.Y., Hsi, H.C. and Shih, M.Y. (2018) Bioregeneration of Spent Mercury Bearing Sulfur-Impregnated Activated Carbon Adsorbent. *Environmental Science and Pollution Research*, **25**, 5095-5104. <https://doi.org/10.1007/s11356-017-9321-x>
- [17] Xu, W., Wang, H., Zhu, T., Kuang, J. and Jing, P. (2013) Mercury Removal from Coal Combustion Flue Gas by Modified Fly Ash. *Journal of Environmental Sciences (China)*, **25**, 393-398. [https://doi.org/10.1016/S1001-0742\(12\)60065-5](https://doi.org/10.1016/S1001-0742(12)60065-5)
- [18] Korpiel, J.A. and Vidic, R.D. (1997) Effect of Sulfur Impregnation Method on Activated Carbon Uptake of Gas-Phase Mercury. *Environmental Science & Technology*, **31**, 2319-2325. <https://doi.org/10.1021/es9609260>
- [19] Liu, W., Vidic, R.D. and Brown, T.D. (2000) Optimization of High Temperature Sulfur Impregnation on Activated Carbon for Permanent Sequestration of Elemental Mercury Vapors. *Environmental Science & Technology*, **34**, 483-488. <https://doi.org/10.1021/es9813008>
- [20] Feng, W., Borguet, E. and Vidic, R.D. (2006) Sulfurization of Carbon Surface for Vapor Phase Mercury Removal I: Effect of Temperature and Sulfurization Protocol. *Carbon*, **44**, 2990-2997. <https://doi.org/10.1016/j.carbon.2006.05.019>
- [21] Feng, W., Borguet, E. and Vidic, R.D. (2006) Sulfurization of a Carbon Surface for Vapor Phase Mercury Removal II: Sulfur Forms and Mercury Uptake. *Carbon*, **44**, 2998-3004. <https://doi.org/10.1016/j.carbon.2006.05.053>
- [22] Sano, A., Takaoka, M. and Shiota, K. (2017) Vapor-Phase Elemental Mercury Adsorption by Activated Carbon Co-Impregnated with Sulfur and Chlorine. *Chemical Engineering Journal*, **315**, 598-607. <https://doi.org/10.1016/j.cej.2017.01.035>
- [23] Rashid, K., Suresh Kumar Reddy, K., Al Shoaibi, A. and Srinivasakannan, C. (2013) Sulfur-Impregnated Porous Carbon for Removal of Mercuric Chloride: Optimization Using RSM. *Clean Technologies and Environmental Policy*, **15**, 1041-1048. <https://doi.org/10.1007/s10098-012-0564-4>
- [24] Zhang, Y., Duan, W., Liu, Z. and Cao, Y. (2014) Effects of Modified Fly Ash on Mercury Adsorption Ability in an Entrained-Flow Reactor. *Fuel*, **128**, 274-280. <https://doi.org/10.1016/j.fuel.2014.03.009>
- [25] Cai, J., Shen, B., Li, Z., Chen, J. and He, C. (2005) Removal of Elemental Mercury by Clays Impregnated with KI and KBr. *Chemical Engineering Journal*, **241**, 19-27. <https://doi.org/10.1016/j.cej.2013.11.072>
- [26] Johari, K., Saman, N., Tien, S.S., Chin, C.S., Kong, H. and Mat, H. (2016) Removal of Elemental Mercury by Coconut Pith Char Adsorbents. *Procedia Engineering*, **148**, 1357-1362. <https://doi.org/10.1016/j.proeng.2016.06.588>

Abbreviations:

AC: Activated carbon

CAA: Clean air act

Hg⁰: Elemental mercury

HAP: Hazardous air pollutant

N₂: Nitrogen

SAC: Sulfur doped porous activated carbon

S/C: Sulfur to activated carbon