Zhao, Haitao and Yang, Gang and Mu, Xueliang and Cao, Pengfei and Gao, Xiang and Wu, Tao (2017) Hg 0 capture over MoS2 nanosheets containing adsorbent: effects of temperature, space velocity, and other gas species. Energy Procedia, 105 . pp. 4408-4413. ISSN 1876-6102

Available online at www.sciencedirect.com

ScienceDirect

Energy Procedia 105 (2017) 4408 – 4413

Energy Procedia

The 8th International Conference on Applied Energy – ICAE2016

Hg^0 Capture over MoS_2 Nanosheets Containing Adsorbent: Effects of Temperature, Space Velocity, and Other Gas Species

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Abstract

Fossil fuel burning is the largest anthropogenic source of mercury emission, which is expected to be the first industrial sector to be addressed under Minamata Convention. In this research, the preliminary investigation has been carried out to understand the effects of temperature, space velocity, and SO_2 and O_2 on Hg^0 capture over MoS_2 nanosheets containing elemental mercury adsorbent. The adsorbent exhibited excellent performance in the removal of Hg^0 at a low temperature below 125°C (particularly at 50°C) with a space velocity below 9.0×10^4 ml/(h·g). It was found that the presence of O_2 had positive effect on Hg^0 removal whilst SO_2 had slightly negative effect on mercury capture at low temperature, such as 50°C. However, such negative effect became negligible when O_2 co-existed with SO_2 in the simulated flue gas. The research provided fundamental information for further development of the 2D graphene-like MoS_2 nanosheets containing adsorbent for mercury capture.

Peer-review under responsibility of the scientific committee of the 8th International Conference on Applied Energy.

Keywords: Hg^0 capture, MoS_2 nanosheet, transition metal dichalcogenide, effects study

1. Introduction

Mercury has become a global concern due to its potent detrimental impacts on biological system and human being's health[1]. According to the global mercury assessment 2013, fossil fuel burning related industries are the major anthropogenic sources of mercury emission, accounting for 25% of total anthropogenic mercury emissions[2], which is expected to be the first industrial sector to be addressed under Minamata Convention on Mercury emission.

In recent years, due to the ever-tightening legislations on Hg^0 emission control in western countries as well as in some developing countries such as China [3, 4], there has been considerable interests in the development of Hg^0 capture materials worldwide.

The activated carbon injection (ACI) system has been commercially deployed for mercury removal at coal-fired power plants since 2005 [5]. Due to the high affinity of sulfur to mercury, the capacity of mercury removal can be enhanced when activated carbon is impregnated with sulfur [6, 7]. However, the

Peer-review under responsibility of the scientific committee of the 8th International Conference on Applied Energy.
doi:10.1016/j.egypro.2017.03.935

activated carbon adsorbed with mercury has negative effects on the properties of fly ash, which is a by-product in power plants and can be used for concrete production [8, 9]. It is therefore necessary to develop alternative non-carbon-based sorbents for mercury emission control at coal-fired power plants [10, 11].

Recently, due to their grapheme-like structures and unique properties, two-dimensional transition-metal dichalcogenides (2D TMDCs) have attracted increasing attention [12, 13]. Based on our previous investigation on a suite of transition metal oxides and their corresponding sulfides for Hg^0 capture, MoS_2, a 2D TMDC, was identified as the active centres for mercury capture [14-16]. In order to further evaluate the performance of the MoS_2 nanosheets containing adsorbent, the effects of operating temperature, space velocity, and the existence of other gas species were studied in this research.

2. Materials & methods

2.1. Preparation of sample

The MoS_2 nanosheets containing adsorbent was prepared by the combination of incipient wetness impregnation (IWI) method and sulfur-chemical vapour reaction (S-CVR) method. $(NH_4)_6Mo_7O_{24}\cdot 4H_2O$ (analytical grade, Sinopharm Chemical Reagent Co, Ltd.) was used as the metal precursor. A commercial γ -Al_2O_3 (V-SK Co., Ltd., size range: 1.18 mm $\leqslant$ x $\leqslant$1.70 mm) was used as the support. The preparation procedures were described elsewhere in our previous study [15, 17].

2.2. Apparatus and Procedure

To evaluate the effects of different factors on Hg^0 capture performance, tests were conducted in rig specially designed for this purpose[15]. The sample was loaded into a dual-reactor system with one fixed-bed reactor used for accurate temperature detection. The simulated flue gas with different space velocities was controlled by using mass flowmeters. The mercury analysis system (Tekran 3300RS, USA) and mercury generator (Tekran 2537, USA) were used for continuously monitoring and generating of the Hg^0. The concentrations of SO_2 and O_2 at both inlet and outlet were measured using a flue gas analyser (Testo 350 Pro, Germany).

3. Results & discussions

The performance of MoS_2 nanosheets containing adsorbent for Hg^0 captured at different temperatures (in N_2 atmosphere at 4.5×10^4 ml/(h·g)) is presented in Fig. 1. The baseline concentration used in this study was approximately 30 μg/m^3. The results show that more than 25 μg/m^3 was captured for testing temperatures up to 100°C. However, with the temperature was raised from 100°C to 125°C, the outlet concentration increased significantly to be above 15 μg/m^3. During the 3 hours testing, there were noticeable increase of outlet concentration when the sample tested at 25°C and 100°C whilst it remained the same at 50°C and 75°C. The 30 μg/m^3 of Hg^0 was almost completely captured at the temperature around 50°C. The results indicated that this adsorbent exhibited excellent performance in the removal of Hg^0 at lower temperatures, especially at 50°C.

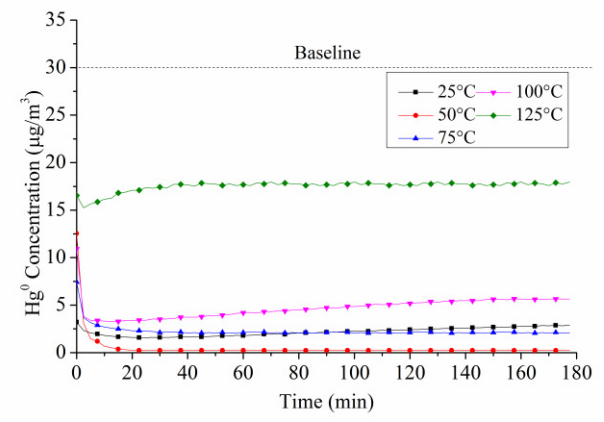

Fig. 1 The effects of different temperatures on Hg^0 capture

Generally, space velocity is one of the most important factors in chemical reactor design. Normally, weight hourly space velocity (WHSV) is commonly used to measure the space velocity, which is the quotient of the mass flow rate of the reactants divided by the mass of the sorbent used in the reactor. Fig. 2 illustrates the effects of the selected different levels of WHSV on the efficiency of Hg^0 removal.

For the highest level of WHSV, 4.5×10^5 ml/(h·g), the outlet concentration of Hg^0 reached 10 μg/m^3 within 250 min. This result indicated that the mass of the adsorbent loaded was not sufficient to capture the Hg^0 in such a high space velocity.

By contrast, there was a small increase of the outlet concentration of Hg^0 during the 1000 min test when WHSV was lowered to 9.0×10^4 ml/(h·g). The concentration remained stable between 1000 min and 2000 min in the test. This result suggested that the amount of sorbent was adequate for the capture of most of the Hg^0 but sacrificed part of the removal efficiency by approximately 7% in this case.

With the further decrease of WHSV to 4.5×10^4 ml/(h·g), the 30 μg/m^3 Hg^0 was completely captured during the 2000 min test. The results confirmed that the adsorbent could achieved high Hg^0 capture efficiency at relatively low WHSVs.

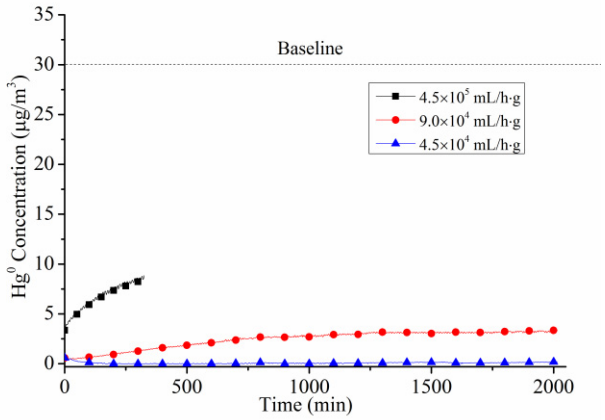

Fig. 2 The effects of different space velocities on Hg^0 capture

Oxygen is one of the components in flue gas emitted by coal-fired power plants, which might have

some influence on mercury capture. Hence, it is necessary to consider how the Hg^0 capture performance is affected by the presence of O_2 in the gas mixture.

Fig. 3 reveals the effect of oxygen on Hg^0 removal in comparison with the oxygen-free environment. It can be found that similar amount of Hg^0 was removed for both cases. However, the almost complete removal of mercury was observed when O_2 was in presence. The outlet elemental mercury concentration in the presence of oxygen was always lower than that in oxygen free environment after the 20 minutes of testing. This result demonstrated that for MoS_2 nanosheets containing adsorbent, the presence of O_2 enhanced Hg^0 removal process.

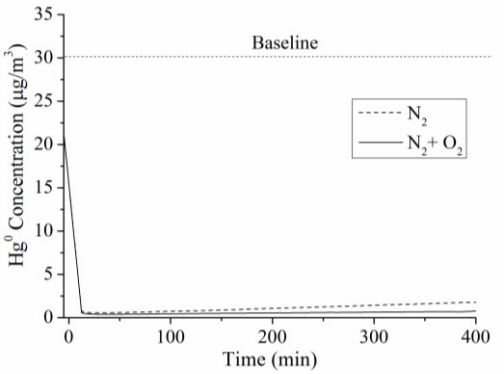

Fig. 3 The effects of O_2 on Hg^0 capture

For coal-fired power plants, SO_2 is another component presented in flue gas although in small amount. It is difficult to remove SO_2 completely from flue gas even for the power plants with existing air pollution control devices (APCDs), such as FGD. Therefore, it is important to understand the influence of SO_2 on Hg^0 removal. Fig. 4 illustrates the influence of SO_2 (600 ppm) on the efficiency of Hg^0 capture with or without the presence of O_2 (10%).

Firstly, to make it relevant to industrial applications in terms of SO_2 concentration in flue gas, 600 ppm of SO_2 was introduced into the gas mixture when the removal of Hg^0 over the studied adsorbent reached steady state after 60 minutes' test. As it can be seen from Fig. 4, the concentration of Hg^0 only increased by approximately 1 $\mu g/m^3$ when SO_2 was in presence. The result remained stable during the 120 min test. It indicates that the SO_2 alone had small but noticeable negative effect on the efficiency of Hg^0 removal.

Nevertheless, the 1 $\mu g/m^3$ of Hg^0 was reduced again when O_2 was introduced in the gas mixture. The mercury removal performance was recovered when both SO_2 and O_2 present in the gas mixture. Furthermore, when O_2 supply was cut off after another 120 minutes' test to study the performance in the presence of only SO_2, the concentration of Hg^0 started to increase again. These results suggested that the negative effect caused by SO_2 could be ignored when O_2 co-existed with SO_2 in the gas mixture, which is the case at coal-fired power plants.

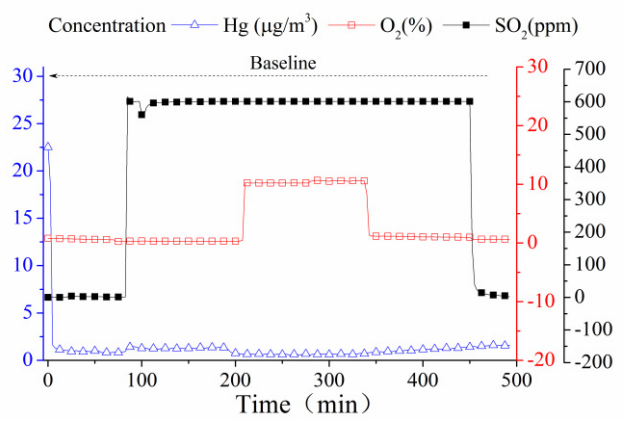

Fig. 4 The effects of SO_2 and O_2 on Hg^0 capture

4. Conclusions

In this research, the effects of temperature, space velocity, and other gas species on Hg^0 capture over the MoS_2 nanosheets containing adsorbent were studied. The adsorbent exhibited excellent performance at a temperature below 125°C and a space velocity below 9.0×10^4 ml/(h·g). The presence of O_2 enhanced Hg^0 removal whilst SO_2 showed small but noticeable negative effect on mercury removal at 50°C. Such negative effect could be ignored when O_2 co-existed with SO_2 in simulated flue gas.

Acknowledgements

Following funding bodies are acknowledged for partially sponsored this piece research: Ningbo Bureau of Science and Technology (Innovation Team Scheme, 2012B82011, and Major Research Scheme, 2012B10042), and Department of Science and Technology Zhejiang (Provincial Innovation Team on SO_x and NO_x Removal Technologies, 2011R50017).

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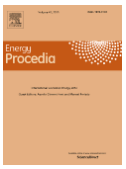

Biography

Prof. Tao Wu is currently leading the Ningbo New Materials Institute and Ningbo Municipal Key Laboratory of Clean Energy Conversion Technologies at The University of Nottingham Ningbo China (UNNC). He has over 20 years of R&D experience on the efficient conversion and utilization of fossil fuels and the related materials.