Science & Technology Asia

Vol.22 No.4 October - December 2017

Page: [ 27 – 33 ]

#### Original research article

# The Development of Coconut-Based **Activated Carbon Impregnation to Adsorb Mixture of Organic and Inorganic Gases**

# Sorn Suwannachot<sup>1,\*</sup>

<sup>1</sup>Department of Environmental Science, Faculty of Science and Technology Thammasat University, Pathum Thani 12120, Thailand

#### Warawut Suadee<sup>2</sup>

<sup>2</sup>Faculty of Public Health, Thammasat University, Pathum Thani 12120, Thailand

Received 3 August 2017; Received in revised form 10 August 2017 Accepted 11 August 2017; Available online 15 December 2017

# ABSTRACT

Activated carbons have been the most widely used adsorbent in adsorption system. This study aims to develop the coconut-based activated carbon to treat both organic and inorganic contaminants in polluted air. The proper chemical treatment was selected: nitric acid (HNO<sub>3</sub>), sodium hydroxide (NaOH) and ammonium nitrate (NH4NO3). For each chemical, varying concentrations of 0.1, 0.5, 1.0, 2.0, 3.0, 4.0 and 5.0 N were used. In a treatment, the activated carbon was soaked and stirred for 8 hours at 70 °C in the solution and heated at 105 °C for 12 hours and then cooled down in a desiccator for 12 hours. This impregnated activated carbon was observed for the surface characteristics using scanning electron microscope (SEM), analyzed for iodine number and analyzed for amount of functional groups on the surface. The suitable chemical treatment was selected by considering surface characteristics, porosity and chemical property. The adsorption capacity of the impregnated activated carbon was tested and compared with that of the original activated carbon. The treatment with 1.0 N sodium hydroxide (NaOH) appeared to be the most appropriate technique to impregnate for the coconut-based activated carbon to adsorb mixture of organic and inorganic gases. The physical adsorption ability for benzene was reduced by 55.90 percent, while, the chemical adsorption ability for hydrogen sulfide increased by 537.61 percent.

**Keywords:** Activated carbon; Impregnation; Adsorption; Organic gases; Inorganic gases; Adsorption capacity

\*Corresponding author: sosuwa@yahoo.com doi: 10.14456/scitechasia.2017.16

#### 1. Introduction

Adsorption is one of the potential options to control the air pollution because of low energy requirement, cost advantage, and ease of applicability over a relatively wide range of temperatures and pressures [1]. It is a widely used technique for volatile organic compounds (VOCs) control because it has high surface area, porous structure, and high degree of surface reactivity [2]. Activated carbons have been the most widely used adsorbent in removing, but it is less to remove inorganic substances [3] such as hydrogen sulfide (H<sub>2</sub>S).

Air pollution from some sources, such as wastewater treatment plant has low concentrations of both organic inorganic and the flow rates are varied, which make it difficult to control. One possible solution to this problem is a development of an adsorption system to be able to adsorb both organic and inorganic substances by improving adsorption ability of the adsorbent. The process to increase the adsorption ability of activated carbon is surface modification methods which could be in the form of impregnation, acid, base [4] or salt [5] treatment, heat treatment, and oxidation by gas [4]. This research has focused on the surface modification by chemicals. The chemical impregnation will result in changes of the functional groups, acidic or basic character [6], on the activated carbon surface [7]. The increase of the functional groups on activated increase the chemical will adsorption ability of the activated carbon. However, the impregnation may decrease the internal porosity of the activated carbon causing the reduction of surface area and porous volume, [4] if the concentration of the treatment chemical is too high [8].

In order to develop the adsorbent to be used for the mixtures of organic and inorganic air pollution. It is necessary to study the method to increase the adsorption ability of activated carbon by appropriate chemical treatment. The method should increase the inorganic adsorption ability while the organic adsorption ability is still maintained.

# 2. Materials and Methods 2.1 Materials

Materials in this experiment were commercial and impregnated activated carbons. The granular commercial activated carbon (AC) made from coconut shell. The properties for the commercial activated carbon are shown in Table 1. agent gases for testing The adsorption ability were benzene and H<sub>2</sub>S, which represented organic and inorganic pollutants, respectively.

**Table 1** The properties of the commercial activated carbon [9].

Properties	Detail
Size	8-16 mesh (2.36-1.18 mm)
Density	$0.48 \text{ g/cm}^3$
Ash	3.5 %w
Surface area	$1,100 \text{ m}^2/\text{g}$
Iodine number	1,050 mg/g

#### 2.2 Activated carbon preparation

The commercial activated carbon was pretreated by distilled water and dried at 105°C for 12 hours, and then cooled down in a desiccator for 12 hours. This activated carbon was referred to the original activated carbon.

#### 2.3 The impregnation selection

The original activated carbon was separately impregnated with 0.1, 0.5, 1.0, 2.0, 3.0, 4.0 and 5.0 N of nitric acid (HNO<sub>3</sub>), sodium hydroxide (NaOH) and ammonium nitrate (NH<sub>4</sub>NO<sub>3</sub>). The original activated carbon was soaked with the chemical for 8 hours at 70 °C. The sample was heated at 105 °C for 12 hours to dry, and then cooled down in a desiccator for 12 hours.

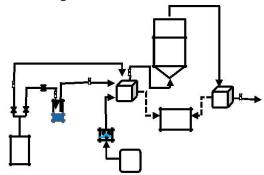
Each sample of impregnated activated carbon was tested for the surface characteristics, iodine number and the

quantity of the functional groups. Scanning electron microscope (SEM) was applied to examine the surface characteristics. ASTM D4607 was used for iodine number [10]. Acid-base titration was applied to quantify of the functional groups on the surface of the activated carbon. The sample were shaken in 0.05M NaOH and HCl for 48 hours and titrated with 0.05M HCl and NaOH for acid functional group and base acid functional group, respectively. After that, both values were combined for the total functional groups on the surface [11].

The adsorption ability of impregnated activated carbons were compared. The most suitable method was selected to adsorption capacity test.

# 2.4 Adsorption capacity test

For the adsorption capacity test, the selected impregnated activated carbon was compared with the original activated carbon in laboratory scale reactor. The reactor is a glass cylinder, 10 cm internal diameter and 1.5 liter. In the experiment, the reactor was filled with 100 g of the selected activated carbon. The agent gas was generated by introducing benzene and H2S to zero air stream at 2 liter/min. Five concentrations of benzene and 3 concentrations of H<sub>2</sub>S were introduced. The concentrations of inlet and outlet of benzene and H2S were continually analyzed by gas chromatography (Baseline® Series 8900). The schematic diagram overview for the laboratory scale adsorption system is shown in Fig. 1.



 $\begin{tabular}{ll} Fig. & 1 & Schematic & diagram & of & the \\ laboratory scale adsorption system. \end{tabular}$ 

The adsorption capacity of

impregnated and original activated carbon were calculated by using the difference between the inlet concentration and the outlet concentration, flow rate, time and mass of adsorbent by the equation (2.4).

$$Ads_{c} = \frac{\sum (C_{in} - C_{out}) \times F \times T}{M}$$
 (2.4)

where

Ads<sub>c</sub> is adsorption capacity (g/100 g AC)  $C_{in}$  is Inlet concentration (mg/m<sup>3</sup>)  $C_{out}$  is outlet concentration (mg/m<sup>3</sup>) F is air flow rate (l/min) T is time (min) M is mass of adsorbent (g)

## 3. Results and Discussion

# 3.1 Impacts to the surface character-istics of activated carbon

The surface characteristics of the impregnated activated carbon determined based on the comparison with the original activated carbon by using SEM. The surface characteristics of original activated carbon was clean and the cavity was clearly visible (Fig. 2). The effects of the chemicals treatment to the activated carbon surface were shown in Fig. 3. After treated by chemical, the covering appeared on the surface of the activated carbons, the destroyed structures of the activated carbons were observed when the concentrations of chemical treatment were higher than 0.1N for HNO<sub>3</sub>, 1.0N for NaOH and all concentrations of NH<sub>4</sub>NO<sub>3</sub>

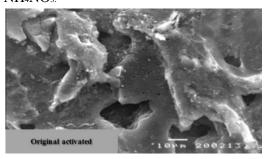


Fig. 2 Surface characteristics of the original activated carbon.

# 3.2 Impacts to the porosity

After impregnation with selected chemical, iodine number of the activated carbons were changed as shown in Table 2. The treatments effected the porosity of the activated carbon when the concentrations

were higher than 3.0N 1.0N and 0.5N for HNO<sub>3</sub> NaOH and NH<sub>4</sub>NO<sub>3</sub>, respectively. Fig. 4 shows the trends of iodine number of impregnated activated carbon along with the concentration.

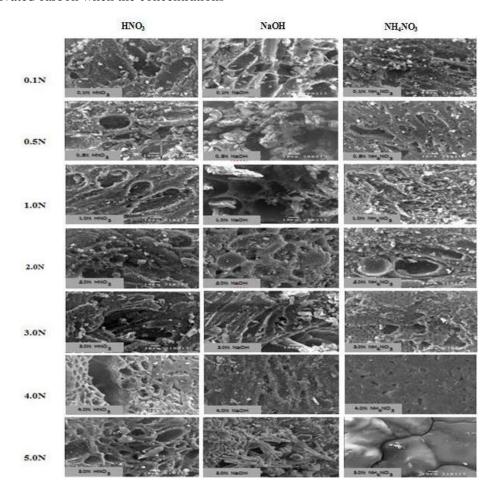


Fig. 3 Surface characteristics of the impregnated activated carbon.

**Table 2** Iodine number of original and impregnated activated carbon (mg/g).

	Original activated carbon 1,224.26				
I	Impregnated activated carbon				
	AC- HNO3	AC- NaOH	AC- NH4NO3		
0.1N	1,229.52	1,225.12	1,360.93		
0.5N	1,222.57	1,226.88	1,322.31		
1.0N	1,226.99	1,245.23	1,198.67		
2.0N	1,234.85	1,046.47	1,142.14		
3.0N	1,247.31	833.03	960.84		
4.0N	1,099.94	811.61	911.20		
5.0N	1,084.21	988.23	1,026.58		

**Table 3** The total functional groups on surface of original and impregnated activated carbon (mmol/g).

(	Original activated carbon 1.322				
Imp	Impregnated activated carbon				
	AC- HNO <sub>3</sub>	AC- NaOH	AC- NH4NO3		
0.1N	2.815	2.812	2.443		
0.5N	2.934	2.937	2.358		
1.0N	3.018	2.957	2.354		
2.0N	3.267	3.49	2.531		
3.0N	3.368	3.917	2.897		
4.0N	3.923	4.403	2.364		
5.0N	3.953	4.597	2.453		

Impacts to the functional **surface**The functional groups on groups on the surface of activated carbon were increased, for concentrations of HNO3, NaOH and NH4NO3 (Table 3). The amount of the total functional groups found on the surface of impregnated activated carbon with HNO3 and NaOH was likely increase along with the increment of concentration (Fig. 5). However, the total functional groups on the surface of impregnated NH4NO3 activated carbon was likely dropped at the concentration of 4.0N and higher since at that point its porosity were destroyed (Fig. 3).

The suitable chemical to impregnated activated carbon for adsorbs organic and inorganic contaminated air pollution is 1.0N NaOH, this is a method to maintain the physical properties, while increasing the quantity of functional groups on surface of the activated carbon.

#### 3.4 Adsorption capacity

The adsorption capacity of impregnated activated carbon for benzene decrease the comparing with original activated carbon (Fig. 6). Both adsorption isotherms were accordance with Langmuir isotherm. The Langmuir isotherm constants were calculated from the equation (3.1) and the parameters (K and qm) obtained are presented in Table 4.

$$\frac{1}{q} = \frac{1}{q_m} + \left(\frac{1}{Kq_m}\right) \frac{1}{C} \tag{3.1}$$

Where qm and K are Langmuir constants determined from the intercept and slope of the plot, and indicative of maximum adsorption capacity. C and q are equilibrium concentration and equilibrium amount of adsorbed.

Performances of the original and 1.0 N NaOH impregnated activated carbons in H2S adsorption process is shown in Table 5. The original activated carbon has a low adsorption capacity for H2S (Fig. 7), while the adsorption capacity for H2S of impregnated activated carbon was tremendously increase (Fig. 8), the adsorp-tion capacity was increased about 537.61 percent comparing with the original activated carbon.

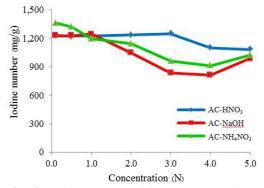
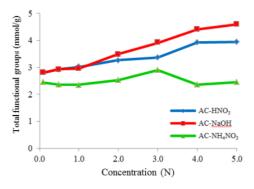
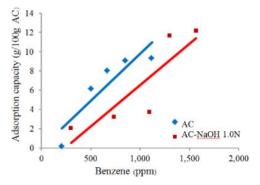


Fig. 4 Iodine number of impregnated activated carbon.



**Fig. 5** Total functional groups on surface of impregnated carbon.



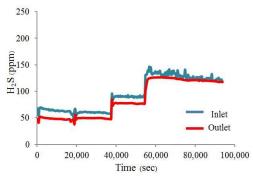
**Fig. 6** The adsorption capacity for benzene.

**Table 4** Langmuir parameters of benzene adsorption on activated carbon.

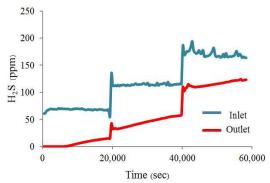
Sample	K	qm (g/100g AC)	$\mathbb{R}^2$
Original AC	0.0012	13.5870	0.9162
AC-NaOH 1.0N	0.0103	5.9952	0.7868

**Table 5** Adsorption capacities of activated carbons for H<sub>2</sub>S.

Sample	Adsorption capacity (g (H <sub>2</sub> S)/ 100 g AC)
Original AC	0.1516
AC- NaOH 1.0 N	0.8143



**Fig. 7** Adsorption curve of H<sub>2</sub>S by original activated carbon.



**Fig. 8** Adsorption curve of H<sub>2</sub>S by 1.0N impregnated activated carbon.

#### 4. Conclusion

The total functional groups on the surface of coconut-based activated carbon was increased by impregnated with nitric acid (HNO<sub>3</sub>), sodium hydroxide (NaOH) and ammonium nitrate (NH<sub>4</sub>NO<sub>3</sub>). However, the impregnation with high concentration may have the effect of damaging the physical structure of the activated carbon.

The treatment with 1.0N sodium hydroxide (NaOH) appeared to be the most appropriate technique to impregnate for the coconut-based activated carbon in this experiment. The physical adsorption ability for benzene was reduced by 55.90 percent, while, the chemical adsorption ability for hydrogen sulfide increased by 537.61 percent. The findings may be useful for treating mixed streams of inorganic and organic emissions.

# Acknowledgements

The research scholarships were supported by Ministry of Science and Technology and the PTT Research & Technology, PTT Public Company Limited. In addition, the Faculty of Science and Technology, Thammasat University supported on the laboratory in the study.

#### References

- [1] Shafeeyan MS, Wan Daud WMA, Houshmand A, Shamiri A. A review on surface modification of activated carbon for carbon dioxide Adsorption. J Analytical and Applied Pyrolysis 2010;89:143–151.
- [2] Li L, Liu S, Liu J. Surface modification of coconut shell based activated carbon for the improvement of hydrophobic VOC removal. J Hazardous Materials. 2011;192(2):683–690.
- [3] Suadee W, Pharamat T, Suwannachot S, Motina K. Final report "The development technology for control the low concentrations of the volatile organic compounds and odor Inorganic in waste gas"; 2013.
- [4] Lee S-W, Wan Daud WMA, Lee M-G. Adsorption characteristics of methyl mercaptan, dimethyl disulfide, and trimethylamine on coconut-based activated carbons modified with acid and base. J Industrial and Engineering Chemistry. 2010;16(6):973–977.
- [5] Boudou JP. An impregnated activated carbon for the separation of VOC mixtures into their individual components. [Internet]. [cited 2016 Oct 20]. Available from: http://web.anl. gov/PCS/acsfuel/preprint%20archive/Files/45\_4\_WASHI NGTON%20DC\_08-00\_0870.pdf

- [6] Fuente E, Menéndez JA, Suárez D, Montes-Morán MA. Basic Surface Oxides on Carbon Materials: A Global View. Langmuir. 2003;19:3505-3511.
- [7] Kruaysawat S. Increasing adsorption efficiency of activated carbon for H2S removal by surface oxidation and metal addition [Doctoral dissertation]. Suranaree University of Technology, Faculty of Environmental Engineering; 2006.
- [8] Yahya MA, Al-Qodah Z, Zanariah Ngah CW. Agricultural bio-waste materials as potential sustainable precursors used for activated carbon production: A Review. Renewable and Sustainable Energy Reviews. 2015;46:218-235.
- [9] Carbokarn Co., Ltd. Technical specification. (n.d.)
- [10] ASTM DESIGNATION: D 4607-94. (2006). Standard test method for determination of iodine number of activated carbon. [Internet]. [cited 2016 Jul 14]. Available from: https://www.scribd.com/document/266717635/AST M-4607-94-Determiancion-Indice-de-Yodo.
- [11] Li H. Selective catalytic oxidation of hydrogen sulfide from syngas [Master's hesis]. Pittsburgh, Pennsylvania: University of Pittsburgh; 2008.