## การเตรียมไททาเนียมไดออกไซด์ยึดเกาะบนถ่านกัมมันต์เพื่อบำบัดฟีนอลในน้ำ

## PREPARATION TITANIUM DIOXIDE SUPPORTED ACTIVATED CARBON (TiO<sub>2</sub>/AC) FOR TREATMENT OF PHENOL IN AQUEOUS SOLUTION

<u>เฉลิมพันธ์ งามโสภาสิริสกุล<sup>1</sup>,</u> สุรชัย ธชีพันธ์<sup>2</sup>, สุชาติ สุวรรณทัศน์<sup>3</sup>, อภิสิฏฐ์ ศงสะเสน<sup>1</sup>

<u>Chalermpan Ngamsopasiriskun<sup>1</sup></u>, Surachai Thachepan<sup>2</sup>, Suchart Suwanatus<sup>3</sup>, Apisit Songsasen<sup>1</sup>

<sup>1</sup>Department of Chemistry and Center of Excellence for Innovation in Chemistry, Faculty of Science, Kasetsart University, Email: ng.chalermpan@gmail.com

<sup>2</sup>Department of Chemistry, Faculty of Science, Kasatsart University, Chatuchak, Bangkok, Thailand, 10900

<sup>3</sup>Department of Physics, Faculty of Science, Kasatsart University, Chatuchak, Bangkok, Thailand, 10900

บทคัดย่อ: ไททาเนียมไดออกไซด์ยึดเกาะบนถ่านกัมมันต์ (TiO<sub>2</sub>/AC) เตรียมได้จากวิธีโซลเจลของ ไททาเนียม (IV) ไอโซโพรพอกไซด์ในสภาวะที่มีถ่านกัมมันต์ผสมอยู่ และนำไปเผาที่อุณหภูมิ ระหว่าง 350 – 500 องศาเซลเซียส เป็นเวลา 1 ชั่วโมง การตรวจสอบคุณลักษณะของ TiO<sub>2</sub>/AC โดย เครื่อง Thermal Gravimetric Analyzer (TGA) พบว่าน้ำหนักของ TiO<sub>2</sub>/AC คงที่ ที่อุณหภูมิ 300-400 องศาเซลเซียส จากเทคนิค X-ray powder diffraction (XRD) พบว่าโครงสร้างของ TiO<sub>2</sub> จะอยู่ในรูป ของ anatase เมื่อการนำ TiO<sub>2</sub>/AC ที่ผ่านการเผาที่อุณหภูมิ 400 องศาเซลเซียส ไปใช้บำบัดฟีนอล เข้มข้น 20 ppm ในน้ำภายใต้รังสี อัลตร้าไวโอเลต พบว่า TiO<sub>2</sub>/AC มีประสิทธิภาพในการบำบัดฟีนอล ดีกว่า TiO<sub>2</sub>-P25 โดย TiO<sub>2</sub>/AC สามารถบำบัดฟีนอลได้ร้อยละ 93 ภายในเวลา 1 ชั่วโมง

**Abstract:** Titanium dioxide supported activated carbon (TiO<sub>2</sub>/AC) was prepared by solgel method from titanium (IV) isopropoxide mixed with activated carbon. The TiO<sub>2</sub>/AC was calcined at temperature between  $350 - 500^{\circ}$ C for 1 hour. From Thermal Gravimetric Analysis, weight of TiO<sub>2</sub>/AC was constant at 300- 400 °C. X-ray powder diffraction (XRD) technique shows anatase phase of TiO<sub>2</sub> in the TiO<sub>2</sub>/AC calcined at 400°C. The prepared catalyst was used for the treatment of 20 ppm aqueous solution of phenol under UV irradiation. The removal efficiency was 93% within 1 hour, which is higher than the efficiency of commercial TiO<sub>2</sub>-P25.

**Introduction:** The presence of phenolic compounds in aqueous solutions has caused several environmental problems. Phenol is one of the most common phenolic compounds formed contaminated in natural aquatic resource. The main source of phenol is the discharge from chemical process industries such as coal gasification, polymeric resin production, oil refining, coking plants, paper mill, herbicides and fungicides production [1]. Phenol in environment is considered as a major aquatic pollution because of its easily contaminated in water [2]. Moreover, it has been reported that even low concentration of phenol is carcinogenic to humans. Phenol is soluble and stable in water, therefore its removal to below hazardous level ( $\leq 0.1$ -1.0 mg/L) is extremely difficult. Traditional methods such as solvent extraction, adsorption by

activated carbon and chemical oxidation, are disadvantageous due to the high cost and production of hazardous by-products. Biological degradation is an environmentally friendly and low cost alternative but it is a time-consuming method.

Photocatalytic degradation using  $TiO_2$  has become more favorable because of high efficiency, low cost and production of less toxic substance [4]. The present study reports the catalytic degradation of phenol under UV irradiation using  $TiO_2/AC$  in comparison with using either  $TiO_2$  or AC.

**Methodology:** Preparation of catalysts: The titanium dioxide supported activated carbon (TiO<sub>2</sub>/AC) was prepared from Titanium (IV) tetraisopropoxide by sol-gel method in the presence of activated carbon [2,3]. Activated carbon (1 g) was suspended in titanium (IV) isopropoxide (2 ml) under stirring condition. Isopropyl alcohol (20 ml) was subsequently added and was stirred of the mixture for 1 hour. TiO<sub>2</sub>/AC was collected by filtration and was calcined for 1 hour at temperature between 300 and 650 °C.

Characterization of catalysts: TiO<sub>2</sub> was characterized by X-ray powder diffraction (XRD) and Thermal Gravimetric Analysis (TGA) techniques.

Removal of phenol in aqueous solution: Photoactivity of the catalyst (TiO<sub>2</sub>/AC) was determined by phenol degradation in aqueous solution under UV irradiation. The catalyst (0.2 g) was added into 100 ml of solution of phenol 20 ppm. The mixture was irradiated with the UV light, and the solution was collected every 15 minutes for the determination of phenol. Concentration of phenol was determined spectrophotometrically at wavelength of 270 nm [2]. Phenol removal efficiency of the

spectrophotometrically at wavelength of 2/0 nm [2]. Phenol removal efficiency of the  $TiO_2/AC$  was compared with  $TiO_2$  and AC. Phenol removal was also carried out in the reactor without catalyst to observe the photolysis effect of the phenol degradation.

## **Results, Discussion and Conclusion:**

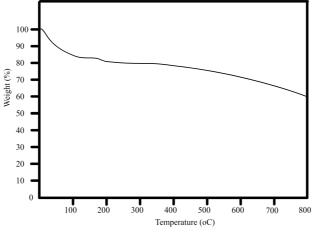
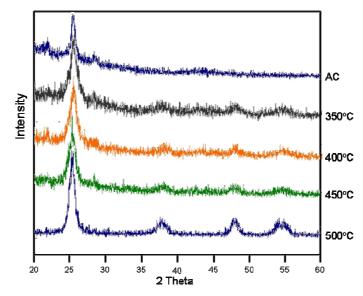


Figure.1 TGA Diagram of TiO<sub>2</sub>/AC.



**Figure 2** The X-ray Diffraction patterns of TiO<sub>2</sub>/AC at difference calcination temperatures and AC.

Figure1 shows weight lost of TiO<sub>2</sub>/AC, which is divided into three ranges. The first drastic loss between 50-100 °C is due to water evaporation. The second weight loss between 100-200 °C is due to loss of organic solvent. The weight of TiO<sub>2</sub>/AC is constant from 200 to 400 °C and above 400 °C the weight decrease slightly by oxidation of AC. The XRD patterns of TiO<sub>2</sub>/AC calcined at various temperatures from 350 – 500°C for 1 hour are shown in Fig 2. The XRD patterns indicated that TiO<sub>2</sub> in the TiO<sub>2</sub>/AC was in anatase phase when the calcinations temperature was in the range of 350 °C to 500 °C. Moreover, when the calcination temperature was higher than 400°C, the appearance of TiO<sub>2</sub>/AC sample was changed from black to gray color. Therefore, TiO<sub>2</sub>/AC calcined at 400 °C was selected to use in the removal process of phenol in the aqueous solution.

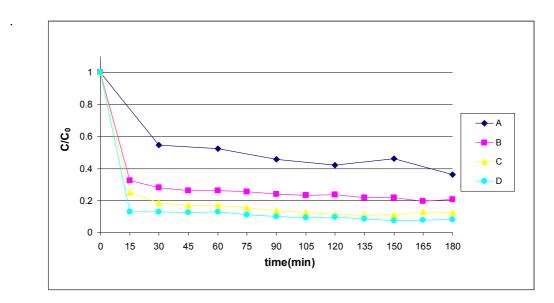
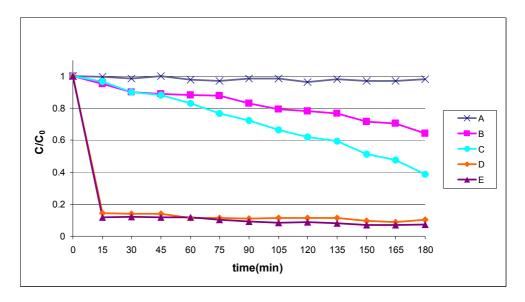


Figure 3 The phenol removal from aqueous solution under UV irradiation: Effect of amount of  $TiO_2/AC$ ; A: 0.05 g, B: 0.10 g, C: 0.15 g, D: 0.20 g.

Figure 3 shows the appropriate amount of catalysts for removal of phenol from its aqueous solution under UV irradiation. Phenol removal efficiency by 0.05, 0.10, 0.15, 0.20 g of  $TiO_2/AC$  were 64 %, 80%, 88% and 93%, respectively. The 0.20 g of  $TiO_2/AC$  gave the highest phenol removal efficiency.



**Figure 4** Phenol removal efficiency of TiO<sub>2</sub>/AC; A: photolysis, B: TiO<sub>2</sub>, C: TiO<sub>2</sub>-P25, D: activated carbon, E: TiO<sub>2</sub>/AC.

Percentage of phenol removal of TiO<sub>2</sub>/AC was compared with TiO<sub>2</sub>, TiO<sub>2</sub>-Degussa-P25, AC and photolysis of phenol are 93%, 36%, 62%, 90% and 3%, respectively. Which shown in Fig. 4, TiO<sub>2</sub>/AC has the highest phenol removal efficiency. However, the removal of phenol by AC may be due to the adsorption of phenol unlike TiO<sub>2</sub>/AC, which has both adsorption and photoactivity. The degree of adsorption and degradation of phenol by TiO<sub>2</sub>/AC will be further investigated.

In conclusion, we have prepared  $TiO_2/AC$  as a catalyst for phenol removal from its aqueous solution, the catalyst is more efficiency than only  $TiO_2$  and AC because it included photoactivity of the  $TiO_2$  and adsorption activity of AC.

## **References:**

- 1. A. Dabrowski, P. Podkoscielny, Z. Hubicki and M. Barczak, *Chemosphere.*, 2005; **58**: 1049–1070.
- 2. B. Tryba, A.W. Morawski and M. Inagaki., *Applied Catalysis B: Environmental*, 2003, **41**: 427–433.
- 3. Tryba, A.W. Morawski and M. Inagaki., *Applied Catalysis B: Environmental*, 2003, **46**, 203–208.
- 4. L. P. Gianluca, A. Bono, D. Krishnaiah and J. G. Collin, *Journal of Hazardous Materials*, 2008, **157**, 209–219.

Keywords: TiO<sub>2</sub>, Activated carbon, Phenol

**Acknowledgements:** We would like to thanks the Department of Chemistry, Faculty of Science, Kasetsart University. Financial support from the Center of Excellence for Innovation in Chemistry (PERCH-CIC) Commission on the Higher Education, Ministry of Education is gratefully acknowledgement.