Activated Carbon Production from Coconut Shell Charcoal Employing Steam and Chemical Activation for Ammonia Adsorption Application

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***Abstract*—Coconut shell charcoal was used to produce activated carbon (AC) for ammonia adsorption application. Activation process was done by steam and chemical activation. Activation temperatures were 700, 800, and 900 °C. Steam activation was done by injecting low temperature steam while chemical activation was done by H_2SO_4 impregnation. Results showed that the activation temperature at 700 °C gave the highest yield followed by 800 and 900 °C, respectively. For the BET surface area, the activation at 900 °C has the highest value (steam AC – 343.9 m^2/g and impregnated AC – 299.3 m^2/g). Pore size distribution analysis showed that the produced ACs was mesopore-type with micropore volume. The impregnated AC from 900 °C also showed the highest iodine number. For the characteristic of ammonia adsorption, the steam AC and impregnated AC from 800 and 900 °C showed better performance when compare to the others.**

***Index Terms*—Biomass, Carbonization, Activated carbon, Ammonia.**

I. INTRODUCTION

THIS work focuses on activated carbon (AC) production from biomass material using conventional carbonization and steam/chemical activation process. Ammonia gas is produced in poultry farms. It is irritating and toxic to a respiratory of chicken [1]. Ammonia is derived from the bacteria activity with uric acid in chicken manure at an appropriate condition. The nitrogen in chicken manure can be devolatilized resulted in an ammonia gas. This poses threats to the environment and the animal. More importantly, the productivity of the farm can be decreased. Humidity, moisture, and ventilation are the important factors to control ammonia in the chicken farm [2]. Supportive bed material such as wood shaving, rice straw, rice hull, etc., is normally provided on the floor of the poultry farm [3,4]. However, their adsorptive capabilities are limited. In addition, they can be the source of ammonia when agglomerated on the floor. Therefore, an alternative ammonia adsorptive bed material shall be investigated.

Coconut shell is a common biomass found in Southeast Asian country. It is generally used for charcoal production that widely used as fuel for cooking. On the other hand, coconut shell charcoal can be used for AC production. The value of the charcoal would be increased. A low-cost AC can be used for ammonia adsorption in poultry farm. Therefore, this research work presents a study on AC production from coconut shell charcoal from steam and chemical activation. Chemical composition of the coconut shell charcoal and AC products was analyzed. Surface area and pore characteristic of the AC were investigated. Finally, ammonia adsorption test was performed.

II. MATERIALS AND METHODS

A. Raw material

Coconut shell charcoal was derived from local community in Samut Sakhon province, Thailand. The charcoal was prepared by a traditional carbonization process. The chemical composition is presented in Table I.

TABLE I
PROPERTY OF COCONUT SHELL CHARCOAL

Item	Unit	Value
Proximate analysis		
Fixed carbon	wt.% db.	53
Ash	wt.% db.	9
Volatile matter	wt.% db.	30
Moisture	wt.% db.	8
Elemental analysis		
Carbon	wt.% db.	60.3
Hydrogen	wt.% db.	3.5
Nitrogen	wt.% db.	0.3
Oxygen	wt.% db.	26.9

B. AC production

AC from coconut shell charcoal was produced from two methods: (1) steam activation and (2) H_2SO_4 impregnation. The schematic of the activator used for activation process is presented in Fig. 1. In each batch, 25 g of coconut shell charcoal was placed in the cylindrical-type reactor made from SUS316 equipped with an electric heater (6.4 kW with PID controller). Nitrogen gas was continuously supplied to prevent oxidative condition. Low temperature steam (100-120 °C) produced from autoclave (All American Sterilizer's Model 25X-120) was provided for steam activation process. The target activation temperature was 700, 800, and 900 °C. Heating rate was set at 10 °C/min.

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C. Impregnation process

A solution of sulfuric acid was prepared for impregnation process. The coconut shell charcoal was impregnated with 8% (w/v) H_2SO_4 solution for 24 h and stored in the oven at 60 ºC. The acidic solution to AC ratio was 0.73 ml/g. After that the impregnated AC was dried in an oven at 105 ºC for 24 h. Then, calcination process of dried product was performed at 300 ºC for 3 h. Finally, the impregnated AC was cooled down to the room temperature and stored in a desiccator for future use.

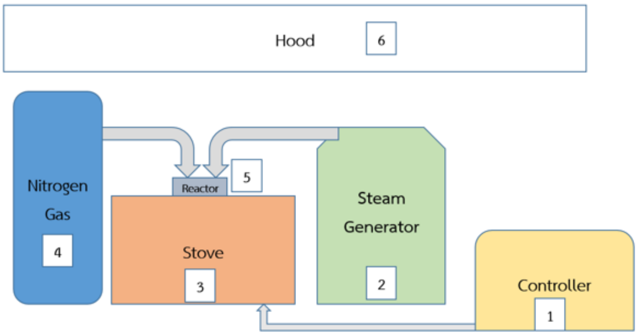

Fig. 1 Steam activation apparatus

D. Material analysis

The raw material and the AC products were dried at 105 ºC in the electric oven for 24 h and grinded into powder. The samples were characterized by proximate analysis, elemental analysis, surface area analysis. Proximate analysis (ASTM D7582) was performed by Simultaneous Thermal Analyzer (model STA 449 F3 Jupiter). Elemental analysis (ASTM D5373) was done by Elemental Analyzer – CHNS/O (model Thermo Flash 2000). Surface area (ASTM D4222) and pore distribution (ASTM D1993-03) were characterized by Surface Area Analyzer (model Quantachrome/Autosorb – 1). Iodine adsorption test was performed according to the ASTM D4607.

E. Ammonia adsorption test

In-house designed ammonia adsorption test kit was fabricated using SUS304 material equipped with ammonia detector instrument. Ammonia solution was mixed with distilled water at the ratio of 1:1.5 (200 ml of ammonia solution and 300 ml of distilled water). Then, the solution was filled into the tank and closed tightly. Connecting rods were connected with the nitrogen gas tank with the ammonia resource tank. Nitrogen gas was used as a medium for blowing the ammonia gas to the adsorption cylinder. The prepared AC was packed in the adsorption cylinder. The feeding rate was in the range of 6-8 liters per min. The ammonia concentration was collected during the experiment by the ammonia detector and recorded.

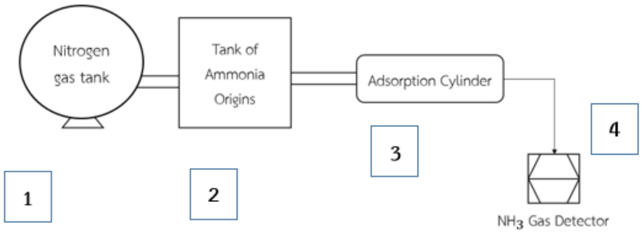

Fig. 2 Ammonia adsorption testing apparatus.

III. RESULTS AND DISCUSSION

A. Yield of AC

After finishing the production of activated carbon, AC yield was calculated relative to the coconut charcoal raw material. It can be seen that the AC yield from activation temperature at 700 °C was the highest (71.5% in average) whereas the AC yield from 900 °C was the lowest (62.3% in average). The activation temperature of 800 °C showed average yield of 66.2%. A clear trend of AC yield was observed as higher activation temperature reduced the AC yield. Because the higher activation temperatures helped eliminating volatile matter from the structure of the charcoal. For impregnated AC, activation temperature did not clearly affect the yield of AC as it was around 48% in average. However, the effect of adding chemical to charcoal structure was obvious. The AC yield from chemical activation was significantly lower than that of the steam activation showing 23% lower compared to the lowest AC yield from steam activation.

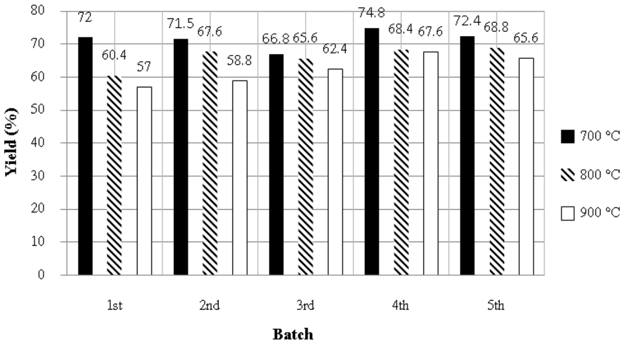

Fig. 3 Yield of AC from different activation temperature.

B. Characterization of AC products

Proximate analysis shows fixed carbon, volatile matter, and ash content of the material. Table II presents proximate analysis of AC products. For the coconut shell charcoal, the amount of fixed carbon and ash was lower than that of the AC but it has higher volatile matter. After activation process, volatile matter was released resulted in a higher content of fixed carbon and ash. When volatile matter was removed from the structure of the charcoal, porosity was generated. The activation temperature affected the amount of fixed carbon and volatile matter significantly. For steam activation, fixed carbon was increased from 63.9% to 70.2% accounting 9.9% increase while the volatile matter was reduced from 15.8% to 9.7% showing 38.5% reduction. Ash content of AC was two-time higher than that of the coconut shell charcoal. Impregnated AC showed different characteristic. Leftover volatile matter was higher than that of the AC produced from steam activation. Increase of fixed carbon was less affected by the temperature in impregnated AC. However, ash content was originally low in this type of AC because the acidic condition of the impregnation process that leached the inorganic material from the charcoal structure.

From elemental analysis presented in Table II, carbon content of the AC was increased compared to the original charcoal. However, the effect of activation temperature on carbon content was not observed. For all ACs, the highest carbon content was from 800 °C activation condition. Also, hydrogen was reduced after activation whereas nitrogen was not changed much. Oxygen content of impregnated AC was significantly higher than that of the steam AC. Sulfur was detected in the case of impregnated AC because of H_2SO_4 impregnation process. Elemental analysis showed that the major composition of the ACs was carbon and the impregnated AC contained sulfur and significant amount of oxygen.

TABLE II
PROPERTY OF ACTIVATED CARBON

Item	A	B	C	D	E	F
Proximate analysis (wt.% db.)						
Fixed carbon	63.9	69.6	70.2	63.9	66.1	67.3
Ash	20.3	15.4	20.0	7.3	7.4	7.5
Volatile matter	15.8	15.0	9.7	28.8	26.6	25.2
Elemental analysis (wt.% db.)						
Carbon	77.6	83.2	79.7	66.3	67.2	66.7
Hydrogen	2.8	2.7	2.2	2.6	2.4	2.2
Nitrogen	0.3	0.3	0.2	0.4	0.3	0.3
Sulfur	-	-	-	1.6	1.2	1.3
Oxygen	19.3	13.7	17.9	29.1	29.0	29.6
Surface area and pore characteristic						
BET surface area (m^2/g)	159	244.2	343.9	210.7	277.4	299.3
Total pore volume (cc/g)	0.10	0.14	0.21	0.13	0.18	0.18
Ave. pore dia. (Å)	26.7	23.4	24.5	24.3	25.4	24.0
Pore size distribution						
Micropore (cc/g)	0.04	0.05	0.07	0.06	0.08	0.08
% Micropore	22.2	28.5	14.8	24.0	19.6	20.6
Mesopore (cc/g)	0.12	0.14	0.37	0.20	0.34	0.32
% Mesopore	74.1	71.0	83.5	75.4	78.1	78.5
Macropore (cc/g)	0.01	0.00	0.01	0.00	0.01	0.00
% Macropore	3.8	0.5	1.8	0.6	2.3	0.9

A: AC from 700 ˚C; B: AC from 800 ˚C; C: AC from 900 ˚C; D: Impregnated AC from 700 ˚C; E: Impregnated AC from 800 ˚C; F: Impregnated AC from 900 ˚C.

C. Surface area and pore characteristic of AC products

The original BET surface area, total pore volume, and average pore diameter of coconut charcoal was 3.37 m^2/g, 0.007 cc/g, and 0.881 Å, respectively. The surface area and pore characteristic have been improved significantly after the activation process. The BET surface area and pore characteristic results are presented in Table II. BET surface area was in range of 159-343.9 m^2/g. The effective activation temperature that produced highest surface ACs area was 900 °C. At this temperature, the steam AC showed the highest surface area of 343.9 m^2/g and the impregnated AC showed 299.3 m^2/g. The total pore volume of the ACs was developed as the activation temperature increased similar to the BET surface area. The total pore volume was about 0.1-0.21 and 0.13-0.18 cc/g for steam AC and impregnated AC, respectively. Average pore diameter of the ACs was in range of 23.4-26.7 Å and 24.0-25.4 Å for steam AC and impregnated AC, respectively. The trend of average pore diameter was not clearly observed when changing the activated temperature in both steam AC and impregnated AC cases. The surface area and pore characterization results showed that the activation process was successful and the ACs can be used as adsorptive material.

Pore size distribution including micropore, mesopore, and macropore of all ACs is presented in Table II. Low contribution of macropore was observed in the pore structure of the all ACs (0.5-3.8%) showing the effectiveness of activation process. Major pore component was mesopore-type. It represented 71.0-83.5% and 75.4-78.5% for steam AC and impregnated AC, respectively. Significant amount of micropore was also observed from all ACs. The micropore was in range of 14.8-28.5% and 19.6-24.0% for steam AC and impregnated AC, respectively. High activation temperature seemed to produce more mesopore and reduce the micropore. In sum, the steam AC and impregnated AC from coconut shell charcoal in this study were the mesopore-type AC with large portion of micropore.

D. Iodine adsorption test

Iodine adsorption test implied the effectiveness of AC adsorption for the substance that has a molecular size of iodine (10 Å). Iodine number of raw material and all ACs is presented in Fig. 4. Coconut shell charcoal showed the lowest iodine number whereas the iodine number of all ACs was significantly improved. Activation temperature strongly affected the iodine number of AC products. The effect of activation temperature on iodine number from steam AC was more significant than that of the impregnated AC. The highest iodine number was from 900 °C steam AC (230.3 mg/g) and 900 °C impregnated AC (237.2 mg/g). The iodine number results are consistent with the BET surface area from the aspect of highest adsorption capability as well as the trend.

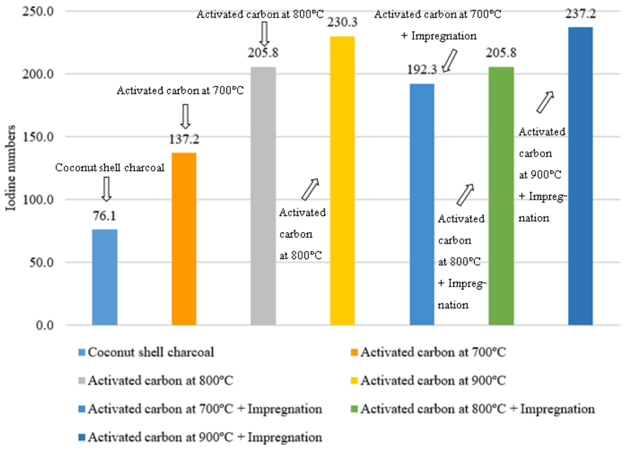

Fig. 4. Iodine adsorption test

E. Ammonia gas adsorption

The ammonia gas adsorption test was done using in-house designed equipment. The data collecting process was done during the whole experiment and it was terminated after the concentration rate of the ammonia in the gas was stabilized.

Ammonia concentration during the experiment is presented in Fig. 5. The blank test with no AC in the adsorption cylinder showed that the ammonia concentration was not constant during the test. This was because the source of ammonia was limited in each test and the ammonia was gradually depleted during the experiment. However, at the beginning of the test, the ammonia source was controlled at the same concentration in all tests. With the AC adsorption material, the ammonia concentration was significantly reduced regardless of the types of AC. From the figure, it can be observed that the steam AC from 700 °C showed the lowest ammonia adsorption performance. This was due to the low BET surface area.

Ammonia removal percentage is presented in Fig. 6. The impregnated AC showed better ammonia adsorption results compared to the original steam AC. Also, low activation temperature steam AC had relatively poor ammonia adsorption performance compared to other ACs. This was clearly observed in the case of 700 °C steam AC. At the beginning of the test, the removal rate of 800 °C steam AC showed the highest peak. This was attributed to its high micropore component consistent with the result shown in Table II. From the ammonia adsorption test, the effective ammonia removal ACs were high temperature (800-900 °C) steam AC or impregnated AC.

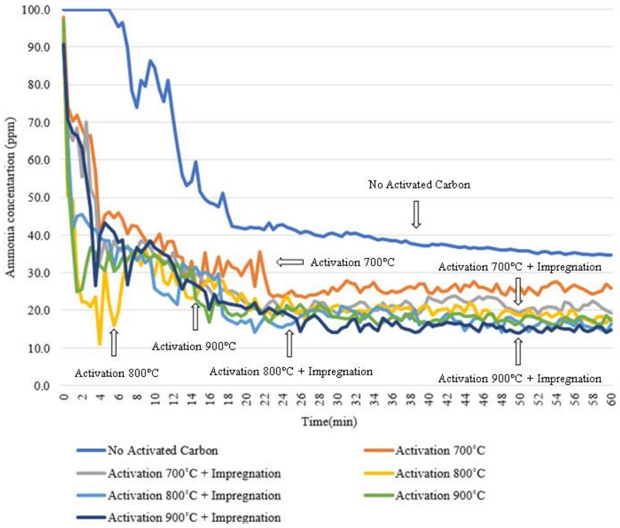

Fig. 5. Ammonia concentration during the test.

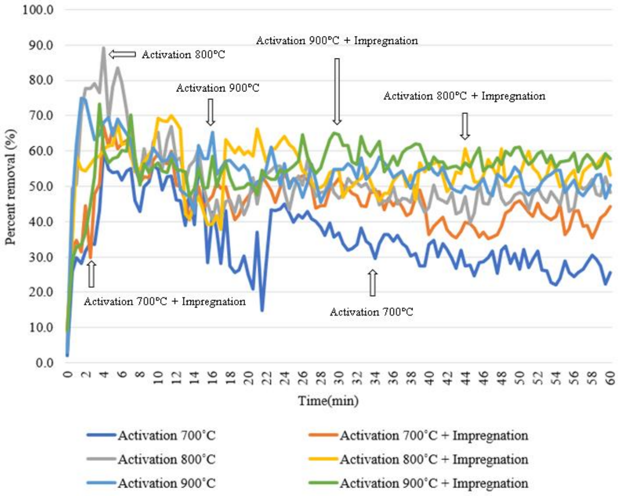

Fig. 6. Ammonia removal rate

IV. CONCLUSION

This paper presents the AC production from steam and chemical activation using coconut shell charcoal as the raw material. The produced ACs were characterized and they were subjected to the ammonia adsorption test. Conclusion can be summarized as follows:

Low activation temperature showed higher AC yield. Steam AC and impregnated AC yield were 62.3-71.5% and 48%, respectively. Carbon content and fixed carbon were increased after activation. Impregnated AC had low ash content. BET surface area of the produced ACs was in range of 159-343.9 m^2/g and the 900 °C activation temperature gave the highest surface area from both steam AC (343.9 m^2/g) and the impregnated AC (299.3 m^2/g). The ACs produced in this study was mesopore-type with significant portion of micropore. Iodine adsorption test showed that high activation temperature can produce high iodine number ACs. Finally, from the ammonia adsorption test, the ACs produced from high temperature activation and impregnated showed high ammonia removal rate. Therefore, the coconut-shell derived AC could be used as ammonia adsorptive bed material for poultry farm.

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