

## Removal of Iron (Fe) by Adsorption using Activated Carbon *Moringa oleifera* (ACMO) in Aqueous Solution

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### Abstract:

Activated carbon has been widely developed as an adsorbent to adsorb heavy metal in wastewater due to its high efficiency and low cost. Activated carbon of *Moringa oleifera* (ACMO) was prepared from the woods for the removal of iron in aqueous solution. The ACMO was produced using chemical activation process impregnation with phosphoric acid as the activating agent. The surface morphology of ACMO was characterized using SEM, and the functional group present was determined using FTIR. Effects of various operating variables namely the carbon dose, adsorbate concentration and contact time on the removal of iron ions have been studied. The adsorption capacity was determined as a function of carbon doses (0.1, 0.2, 0.3, 0.4 and 0.5 g), adsorbate concentration (2, 4, 6, 8 and 10 mg/L) and the contact time between ACMO and aqueous solution (30, 60, 90 and 120 min) with constant parameters of temperature 303 K, agitation at 180 rpm and pH 8. From the data obtained, percent of moisture present in ACMO and raw *M. oleifera* (RMO) were 6.752% and 9.512% respectively. Meanwhile, amount of ash in ACMO was 1.451% and in RMO was 7.592%. In addition, the volatile matter present in ACMO was replaced from RMO about 28.431% and 23.715% respectively. The percentage of fixed carbon of ACMO and RMO were 63.366% and 59.181% respectively. The highest adsorption capacity obtained was at 8.043 mg/g with the amount of carbon dosage of 0.1 g, concentration of iron solution of 10 mg/L and contact time of 120 min.

Keywords: *Moringa oleifera*, ACMO, AAS and SEM.

### Introduction

Industrialization and urbanization has posed a great problem worldwide due to excessive release of heavy metals to the environment. Recently, the removal of heavy metals from wastewater has become the subject of considerable interest. This is due to more strict legislations introduced to control water pollution. In developing countries, water born disease has become the main problem with about 1.6 million people are compelled to use contaminated water (Raj et al., 2010; Schwarz, 2000). Unlike organic pollutant which susceptible to biological degradation, heavy metals are non-biodegradable and are persistence. The presence of heavy metal ions is a major concern due to their toxicity and caused a variety of illness to human (Gupta et al., 2001; Kardivelu et al., 2001). Arsenic, copper, cadmium, lead, mercury, nickel and zinc are among heavy metals that have most severely affected the environment (Kalavathy & Miranda, 2010).

Various treatment technologies have been developed for heavy metal removal from wastewater

including precipitation, membrane filtration, ion exchange, adsorption and co-precipitation/adsorption. Chand et al. (1996) revealed that adsorption is a highly effective technique for the removal of heavy metal from waste stream and activated carbon, and has been widely used as an adsorbent. Activated carbon is a unique and effective agent for purification, isolation and recovery of trace metal. Activated carbon derived from agricultural waste and natural materials such as rice husk, coconut shell and bamboo have been developed due to their low cost and efficiency (Kadirvelu et al 2001). Activated carbon of *Moringa oleifera* (ACMO) has been developed as an agent to remove heavy metals in wastewater. *M. oleifera* belongs to the Moringaceae family that is native to the sub-Himalayan tracts of India, Pakistan, Bangladesh and Afghanistan. *M. oleifera* (MO) is also known as the horseradish tree, a pan-tropical species that is known by such regional names as benzolive, drumstick tree, kelor, marango, mlonge, mulangay, nébéday, saijhan, and sajna. Over the past two decades, many reports have appeared in mainstream scientific journals describing its nutritional

and medicinal properties as well as on its potential as coagulant, flocculants and adsorbent (Fahey, 2005).

It was reported that, the seeds of MO was identified as a very efficient cogulant whereas the powdered woods of MO were used to flocculate contaminants and purify drinking water (Jahn, 1999; Gassenschmidt et al., 1995; Berger et al., 1984; Olsen et al., 1987). Other study have already reported the use of activated carbon of MO woods in removing copper, nickel and zinc in aqueous solution (Kalavathy & Miranda, 2010). Therefore, our study focused on the removal of iron in wastewater. Although, iron is needed in the diet to maintain good health, particularly for making red blood cells that carry oxygen around the body, iron overload may cause liver disease, heart disease and pancreatic disease (Maddrey et. al., 2007).

MO wood has no utility and therefore a cause of environmental degradation. Moreover, the tree is normally cut back annually to less than 1m and allowed to grow, so that pods and leaves remains within arm's reach (Kalavathy & Miranda, 2010). ACMO woods have extensive surface area, which are suitable attribute for metal adsorption. Removing heavy metals by adsorption method from aqueous solution using MO woods are important in order to reduce the amount of heavy metals such as iron presence in the wastewater. Thus, activated carbon from MO can be one of the alternatives to recover the pollution as the activated carbon produced is a natural adsorbent. This study will investigate the efficiency of ACMO as natural adsorbent in varies concentration of metal ions which are in the range of 2 to 10 mg/L. Other than that, this study also will determine the minimum amount of ACMO and the contact time between ACMO and the aqueous iron solution required on increasing the adsorption efficiency.

## Materials and Methods

### *Reagents and apparatus*

The chemicals used were phosphoric acid, hydrochloric acid, sodium carbonate, and standard iron solution, 1000 ppm. Spectroscopic techniques used were fourier transform infrared spectroscopy (FTIR), atomic absorption spectroscopy (AAS), scanning electron microscope (SEM).

### *Preparation of activated carbon of Moringa oleifera (ACMO)*

*Moringa oleifera* wood was collected from Penang, the outer pericarp was removed and they were cut into small pieces. Soluble and coloured components were removed from the seeds by washing with boiling water.

This was repeated until the water virtually colourless. The wood was then washed with distilled water and dried in a hot air oven at 383 K till it reaches constant weight. After this they are ground and sieved to particle size of 1 mm. The sample is then mixed with the phosphoric acid at an impregnation ratio of 1:2 (weight of MO: weight of phosphoric acid) and soaked for 24 h so that the reagents were fully adsorbed onto the raw material. The mixture was dried at 383 K for 1.5 h and then transferred to a sealed ceramic container. The dried mixture was activated in a muffle furnace at 673K for a period of 1 h. The carbons (ACMO) thus produced are then repetitively washed with distilled water to recover all the acid and then with 1% NaHCO<sub>3</sub> solution to remove the residual acid (Kalavathy et al., 2005). The activated carbon was powdered, sieved and stored in sealed containers. The fraction which passed through 200 mesh screen was used for all the experiments.

### *Characterization of adsorbents*

The ACMO was characterized by finding out its moisture (%), ash (%) and volatile matter (%). Moisture content of adsorbent can be measured by weighing 2 g of ACMO and oven dried for half day at 103 °C before being cooled in the desiccators for 30 minutes. After that, the sample was weighed. Meanwhile, to determine the ash content of ACMO, 2 g of the sample was put in the furnace for half day at 550°C and then cooled in the desiccators for 30 minutes. For determination of volatile matter, ACMO was kept in the furnace at 950°C for about 7 minutes. The sample weight was calculated and the difference in weight was recorded (Subhasree, 2007).

### *Scanning electron microscope (SEM)*

SEM images were recorded using JEOL JSM-6360 field emission SEM. The surface functional groups of adsorbents were identified using fourier transform infrared spectrophotometer (FTIR). Fourier transform infrared spectra were obtained using a spectroscope (Tensor 29, Bruker) at resolution 1 cm<sup>-1</sup>. Pressed potassium bromide (KBr) pellets at a sample/KBr weight ratio of 1:100 were scanned and recorded between 4000 cm<sup>-1</sup> and 400 cm<sup>-1</sup> (Kalavathy et al., 2005).

### *Preparation of iron standard solution*

1, 4, 7, 10 and 13 ppm of standard iron solution were prepared from 1000 ppm of iron solution by diluting with deionised water in 100 mL of volumetric flask.

### Preparation of sample solutions

Initial iron concentration of 2, 4, 6, 8 and 10 ppm were prepared by diluting the 1000 ppm iron standard solution with deionised water in 100 mL of volumetric flask. Then, the variables were set up in order to identify the effect of carbon dosage, initial metal ion concentration as well as the effect on contact time of ACMO in the aqueous solution.

### Batch adsorption studies

A series of Erlenmeyer flasks of 250mL capacity containing desired amount of metal ion solutions of known concentrations and mass of ACMO were agitated in a temperature controlled shaker (Orbital, Scigenics) at 180 rpm. Different mass of ACMO (0.1, 0.2, 0.3, 0.4 and 0.5 g and different iron concentration (2, 4, 6 and 10 ppm) were mixed with contact time varies from 30 to 120 minutes. After that, the solutions were filtered before being analyzed in AAS. The amount of metal ions adsorbed at any time  $t$ , was calculated using the equation:

$$q_t = \frac{(C_o - C_t)}{M} \times V$$

where  $V$  is the adsorbate volume (L),  $M$  the amount of ACMO (g), and  $C_o$  and  $C_t$  (mg/L) are the initial concentration of metal ion and concentration of metal ion at any time  $t$ , respectively.

## Results and Discussion

### Scanning electron microscope (SEM) results

ACMO was prepared on chemical activation with phosphoric acid as the activating agent. Phosphoric acid is mostly used in developing activated carbon in various raw materials since it can be washed from the activated carbon by rinsing with boiled pure water. The SEM images clearly reveal the surface texture and morphology of the adsorbent. The surface of raw *M. oleifera* (RMO) as shown in Figure 1(a, b) are more irregular and no pores are seen, but after carbonizing and activating, ACMO has more porous surface and are homogenous as shown in Figure 1(c, d). Therefore ACMO has a larger specific surface area.

An important factor in chemical activation is the degree (coefficient) of impregnation. This coefficient is the dry materials to the weight ratio of the anhydrous activation agent which is 1:2 (weight of MO: weight of phosphoric acid). The effect of the degree of

impregnation on the porosity of the resulting product is apparent from the fact that the volume of salt in the carbonized material is equal to the volume of pores which are freed by its extraction. ACMO undergo small degrees of impregnation which means that the small increase in impregnation amount causes an increase in the total pore volume of the product showing an increase in the volume of smaller pores. When the degree of impregnation is further raised, the number of larger diameter pores increases and the volume of the smallest pores decreases (Balci, 1994).

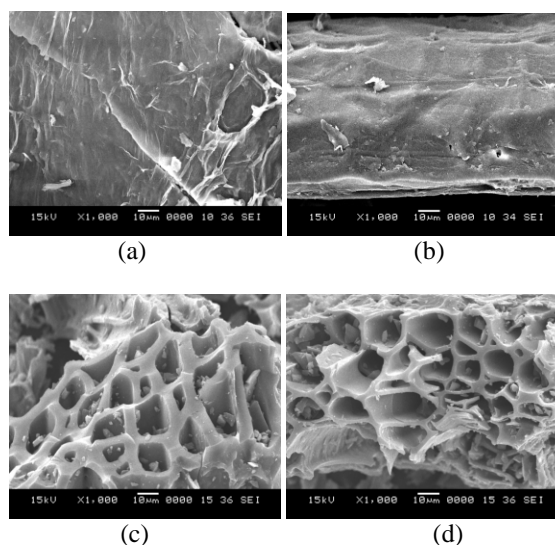


Figure 1: Scanning electron microscope (SEM) images of (a, b) RMO, (c, d) ACMO.

The distribution of pores in activated carbons can vary significantly depending upon the raw material. Figure 2 shows the different shape of pore with different raw material. The pore size distribution also affects the efficiency and selectivity of adsorption. The shapes of pores can have important effects on some properties of carbons and graphite, for example mechanical strength and kinetics reaction. The distribution of pores in ACMO is quite similar as in Figure 2(d) which is slightly round in shape. A consideration of the dimensions of some pollutants showed that activated carbon can feasibly be used to remove many of the impurities occurring in water (Radovic, 2001).

### Fourier transform infrared spectroscopy (FTIR) results

IR spectrum of RMO was compared with IR spectrum of ACMO. Figure 3 showed that a broad band around  $3657 \text{ cm}^{-1}$  -  $3397 \text{ cm}^{-1}$  were attributes to the surface hydroxyl group and chemisorbed water.

The peak appeared at  $2921 \text{ cm}^{-1}$  indicates the stretching vibration of C-H alkane group. Meanwhile,

the bands between  $1641\text{ cm}^{-1}$  and  $1734\text{ cm}^{-1}$  were assigned to strong C=O ketonic and aldehyde group respectively. Small peaks observed at  $1508\text{ cm}^{-1}$  and  $1424\text{ cm}^{-1}$  -  $1327\text{ cm}^{-1}$  s were attribute to C=C alkene aromatic ring and COO<sup>-</sup> carboxylate groups respectively. At around  $1243\text{ cm}^{-1}$  the band can be assigned to strong C-O bond. Fewer peaks are observed in Figure 4 for the functional group of ACO. Less peak are observed for ACO as shown in Figure 4. The disappearance of the above mentioned groups may be due to hydrolysis effect of  $\text{H}_3\text{PO}_4$ , resulting in the decomposition of these groups and subsequent release of their by-products as volatile matter (Kalavathy & Miranda, 2010). This proves that there are only elemental carbons on the surface of ACO. Therefore, the removal of Fe from aqueous solution is not due to complex formation phenomenon with surface functional group but it is only adsorption which is surface phenomena.

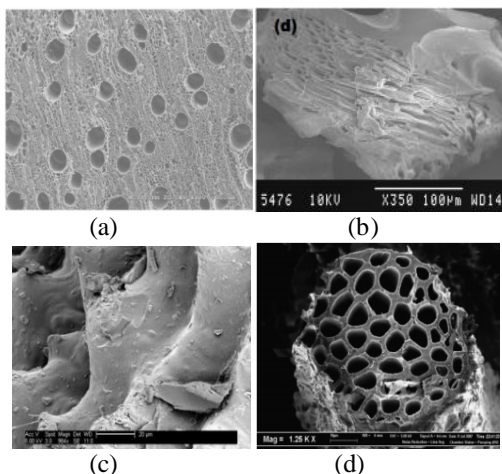


Figure 2: SEM shows different shape of pores depending on types of raw materials: (a) coal (Aso et. al., 2004); (b) wood (Kumar and Chinnaiya, 2009); (c) rice husk (Kearns and Knappe, 2008); (d) coconut (Tan et. al., 2007)

*Moisture and ash content results*

Moisture content is the amount of water physically bound on the activated carbon under normal condition. Activated carbon is generally towards moisture free basis, although occasionally reported in some studies with some moisture content of 3, 8 or 10 % (Nurul ‘Ain, 2007). This may due to the humid conditions that the activated carbon adsorbs considerable moisture over period of time. They may adsorb as much as 25 % to 30 % of moisture and still appear dry (Nurul ‘Ain, 2007). From Figure 5, ACO showed that reduction of moisture available in the RMO is from 9.512 % to 6.752 %. Moisture content of activated carbon does not

affect the adsorptive power, but obviously it dilutes the carbon.

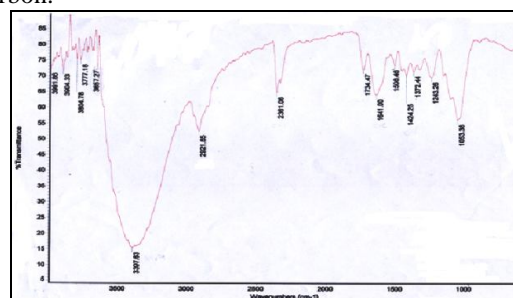


Figure 3: FTIR spectrum of RMO

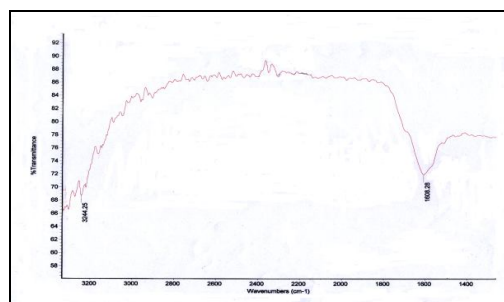


Figure 4: FTIR spectrum of ACO

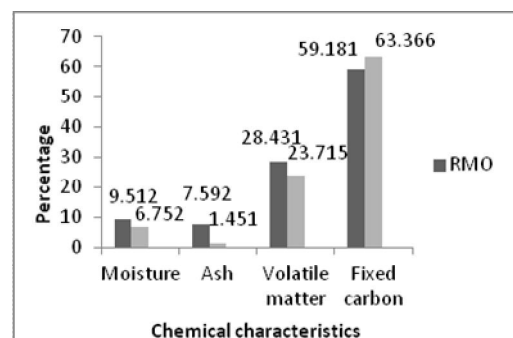


Figure 5: Chemical characteristics of RMO and ACO (%)

Meanwhile, ash content of a carbon is the residue that remains when the carbonaceous materials is burned off. The inorganic material contained in activated carbon is measured as ash content, generally in the range between 2 % and 10 %. Ash content in ACO is reported to be much lower than RMO which are 1.451 % and 7.592 % respectively. The lower the ash content, the better the activated carbon because ash content can lead to increase hydrophilicity and can have catalytic effects, causing restructuring process during regeneration of used activated carbon (Nurul ‘Ain, 2007).

In addition, volatile matter present in the ACO is much lower than RMO. The decrease in volatile matter was due to its removal during carbonization process.

Increase in carbonization temperature promoted the release of volatile matter from breaking of weaker bridges and bonds in organic matrices as well as enhancing the condensation and collapsing reactions of organic matters in the sample to become char with predominantly fixed carbon. Further removal of volatile matter also occurred during chemical activation of EFB by  $H_3PO_4$ . The low volatile matter content implied the high porosity of the adsorbent since less volatile matter remained clog in the pores (Qian et al., 2007).

#### Effect of carbon dosage

Figure 6 illustrates that adsorption capacity of ACMO is decreased when the amount of ACMO is increased. The highest adsorption capacity is for 0.1 g of ACMO with  $5.988 \text{ mg g}^{-1}$ , followed by 0.2, 0.3 and 0.4 g with value of 3.035, 2.045 and  $1.5018 \text{ mg g}^{-1}$  respectively.

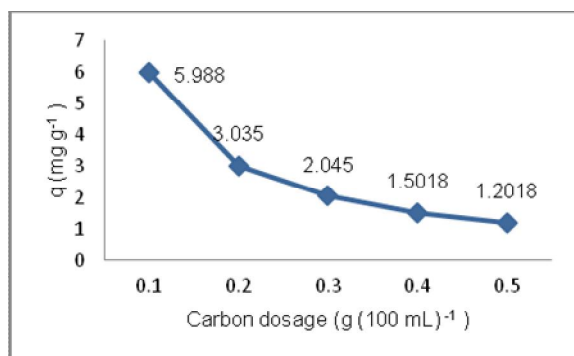


Figure 6: Effect of carbon dosage on adsorption capacity of ACMO, initial iron ion concentration 6 mg/L, temperature 303 K, contact time 120 min, pH 8.

It is found that 0.5 g of ACMO give the lowest adsorption capacity of ACMO which is  $1.2018 \text{ mg g}^{-1}$ . This is due to the fact that at higher carbon dosage, the solution metal ion concentration drops to a lower value and the system reaches equilibrium at lower values indicating the adsorption sites remain unsaturated. This is due to the fact that the actual number of active sites per gram of adsorbent does not increase proportionately and hence there is a regular decrease in adsorption capacity (Kalavathy et al., 2010). According to Abdulrasaq and Basiru (2010), further increase in the mass will not bring about appreciable adsorption because almost all the metal ions might have been removed. In addition, Kumar et al., (2010) also concluded that this reducing trend is because as the adsorbent dose increases, the number of adsorbent particles increases and thus more iron is attached to their surfaces and the adsorption sites remain unsaturated.

#### Effect of initial metal ion concentration

As shown in Figure 7, the amount of metal uptake per unit weight of the adsorbent increases with the increase in initial iron concentration showing the maximum adsorption capacity of  $8.043 \text{ mg g}^{-1}$  for 10 ppm solution and the minimum adsorption capacity of  $1.988 \text{ mg g}^{-1}$  for 2 ppm solution.

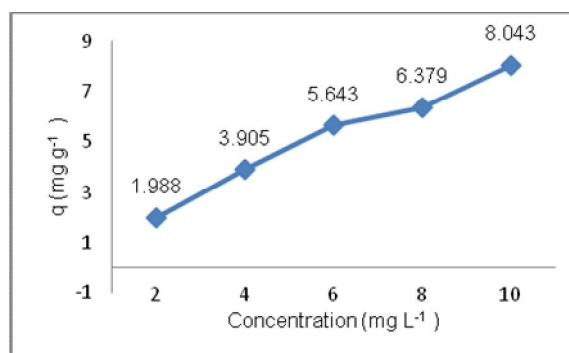


Figure 7: Effect of initial iron concentration on adsorption capacity of ACMO, carbon dosage 0.1 g (100 mL)<sup>-1</sup>, temperature 303 K, contact time 120 min, pH 8.

This is because at higher initial concentrations, the ratio of initial number of moles of metal ions to the available adsorption surface area is high. This may be attributed to an increase in the driving force of the concentration gradient with the increase in the initial iron concentration solution (Kalavathy et al., 2010). Previous study also reported the same trend using coconut husk, chitin and Bengal Gram husk powder as the adsorbent (Abdulrasaq & Basiru, 2010; Karthikeyan et al., 2005; Kumar et al., 2010).

#### Effect of contact time

Figure 8 shows that the removal of iron was found to increase with increasing contact time and attained maximum value for 120 minutes with adsorption capacity of  $9.128 \text{ mg g}^{-1}$ . Meanwhile, for contact time ranging from 30, 60 and 90 min show an increase in adsorption capacity from 7.876, 8.300 and  $8.554 \text{ mg g}^{-1}$  respectively. This is due to the larger surface area of the ACMO available initially for the adsorption of iron ions. As the surface adsorption sites become exhausted, the uptake rate is controlled by the rate at which the adsorbate is transported from the exterior to the interior sites of the adsorbent particles.

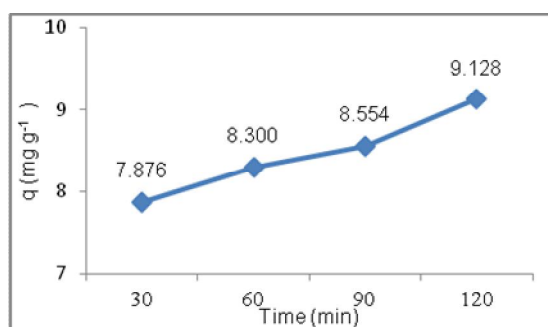


Figure 8: Effect of contact time on adsorption capacity of ACMO, initial iron ion concentration 10 mg/L, carbon dosage 0.1 g in 100 mL, temperature 303 K, pH 8.

According to Karthikeyan et. al., (2005), the amount of iron removal by using chitin as the adsorbent was also increases as the time increased. Maximum adsorption of iron occurred at 8<sup>th</sup> minutes while minimum adsorption occurred at 2<sup>th</sup> minutes with amount adsorbed was 6.5 mg/L and 4.0 mg/L, respectively. In addition, Kumar et. al., (2010) showed that the percentage removal of iron ions by using Bengal Gram husk powder as the adsorbent increased from 22.31% to 78.95% for 5 minutes and 30 minutes, respectively.

## Conclusions

The adsorption capacity was determined as a function of carbon doses (0.1, 0.2, 0.3, 0.4 and 0.5 g), adsorbate concentration (2, 4, 6, 8 and 10 mg/L) and the contact time between ACMO and aqueous solution (30, 60, 90 and 120 min) with constant parameters of temperature 303 K, agitation at 180 rpm and pH 8. The highest adsorption capacity obtained was at 8.043 mg/g with the amount of carbon dosage of 0.1 g, concentration of iron solution of 10 mg/L and contact time of 120 min.

## Acknowledgment

The authors are grateful to Universiti Teknologi MARA (Perlis) for research facilities to accomplish this study.

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