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Characterization of Activated Carbon Produced from *Leucaena leucocephala* Biomass as Heavy Metal Adsorbent: Adsorption of Cadmium from Aqueous Solution

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ABSTRACT. Activated carbon (ACs) of *Leucaena leucocephala* biomass was produced by NaOH activation at impregnation ratios NaOH:char (w/w) of 1:1 (AC-1), 2:1 (AC-2) and 3:1 (AC-3). ACs was prepared under activation temperature 750 ºC. The properties of these activated carbons which are BET surface area was analyzed. It was found that the ACs BET surface area was in order of 140 m^2g^{-1} for AC-1, 531 m^2g^{-1} for AC-2 and 735 m^2g^{-1} for AC-3. Batch adsorption experiment was conducted for the adsorption of cadmium (Cd) from aqueous solution. The ACs samples were tested for adsorption capacity. Batch adsorption was studied in different parameters which were the contact time, effect of pH, initial concentration of cadmium (Cd) and temperature. The results show that the adsorption of Cd was reaching equilibrium within 40 min of contact time for metal ions at initial concentrations of 30 mg/l. Higher pH (>7.0) was found to be favorable for the adsorption of cadmium removal.

Keywords: *Activated carbon, Leucaena leucocephala, Petai belalang, Cadmium, Adsorption;*

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1. INTRODUCTION

For the last few decades, phenomenal economic development has been observed throughout Malaysia. Urbanization, agricultural expansion and industrialization have resulted in increased usage of water and the use of different toxic chemicals. This rapid development has resulted in widespread contamination of soil, vegetables and above all fouling of water bodies up to a greater extent [1]. Water pollution is a very persistent problem. This situation has evolved gradually over time. Recognition of these sorts of problem usually takes a long time and application of necessary preventive measures takes even longer time [1]. Different reports and complaints about industrial waste disposal, stinking water courses within overcrowded cities were an early manifestation of water pollution.

Momodu and Anyakora [2] reported that industries such as plating, ceramics, glass, mining and battery manufacturing are considered as the main sources of heavy metals in local water streams, which can cause the contamination of water with heavy metals. Excessive accumulation of these metals in water, soil, and plants may threaten the safety of ecosystems and also pose serious health risks to human. For example, long-term exposure to as from drinking water and food could cause various human health problems like skin lesions (e.g., hyperkeratosis and pigmentation changes), hypertension, cardiovascular disease, and cancers [3].

Among the techniques that had been developed in cleaning waste water from those dangerous materials are dialysis or electro dialysis, electrolyte extraction, chemical precipitation, solvent extraction, oxidation-reduction, reverse osmosis, ion exchange, evaporation, cementation, dilution, adsorption and filtration [4]. Adsorption is the most attractive option for heavy metal removal [5]. Adsorption is normally used in water treatment to remove dissolved contaminants by physical phase separation in either a steady-state (PAC) or a nonsteady-state (GAC) process [6]. Activated carbon is one of the materials used to remove impurities from solutions. It had been widely used to treat industrial and household water because of its excellent adsorption properties, characterized by a high specific surface area [7].

Generally, activated carbons derived from natural sources, such as coconut shells, wood, coal, and carbonization of synthetic polymers, have large pore volume and high surface area [8]. A number of researches had been reported using biomass as a raw material for producing activated carbon [9]. Most researchers are focusing on the best way on removing metal and dyes for their experiment. In Malaysia, there are many potential of raw material resources for the production of activated carbon. The concept of waste-to-wealth had been promoted and became popular widely in Malaysia where the unwanted wastes are converted into valuable materials [10]. *Leucaena leucocephala* which is *petai belalang* biomass is a suitable candidate to be used for producing activated carbon due to the availability and abundance of the species.

Therefore, this research utilizes *Leucaene leucocephala* biomass for the main material for producing activated carbon. Besides that, evaluation of heavy metal uptake from contaminated water using activated carbon made from *Leucaena leucocephala* biomass were also done to see their efficiency in service.

2. MATERIALS AND METHODS

2.1 Material preparation. Samples from wild *Leucaena leucocephala* were collected around Selangor, Malaysia. Stem of the *Leucaena leucocephala* were oven dried to a moisture content of around 10%. Samples were cut into smaller pieces and further grind to turn them into particles. Produced wood particles were sieved into 1 µm to 2 µm of size. Actual moisture content of the wood particles was determined using oven-dry method and the particles were kept in a closed container for further use.

The dried sample was prepared for pyrolysis to produce charcoal. Pyrolyzer is an old method used for producing charcoal. Pyrolizer was chosen as the carbonization method as it can produce large amount of charcoal at faster rate. About 6 kg was placed inside the pyrolyzer and only 1.5 kg left after carbonization process.

2.2 Activated carbon preparation. *Leucaena leucocephala* biomass particles were activated using chemical means. *Leucaena leucocephala* char was impregnated with NaOH which is sodium hydroxide. This step was performed using impregnation ratio of NaOH:char at 3:1, 2:1 and 1:1. Next, 10 g of char were mixed with 30 g of NaOH and 100 mL distilled water in a vertical stainless steel reactor under magnetic stirring for 2 hours before placed in the oven at 105 ºC for 24 hours.

After 24 hours drying in the oven, the sample was place into the muffle furnace for activation process. The furnace was heated from the room temperature to 700 ºC, 750 ºC and 800 ºC for 90 min. The nitrogen gas will be steam into the furnace at constant 150 cm^3/min flow rate. At the end of the activation time, heated sample will be left to cool down with nitrogen gas flow until room temperature to avoid sample to become ash.

2.3 Activated carbon surface area characterization. Surface analyser, Tri Star 3000 (Micromeritics, USA) was used to measure the surface area, determined by N_2 isotherms using the Brunauer-Emmett-Teller equation (BET). The sample was first degassed at 300 °C for 2 hours before analysis by nitrogen adsorption isotherm at 77 K in the range of relative pressure 10^{-6} to 1.

2.4 Activated carbon adsorption capacity. The adsorption capacity of activated carbon was measured under different conditions such as contact time, pH of solution, initial concentration of adsorbate and adsorption temperature. Approximately 0.5 g of the *Leucaena leucocephala* activted carbon was mixed with 50 ml of the aqueous solutions of cadmium ion in different conical flasks using a temperature-controlled water bath shaker. The mixtures were stirred for pre-determined contact time before filtered using filter paper to get the treated solution. Final solution was analysed using Inductively coupled plasma mass spectrometry, ICP-MS in triplicate to reduce error in experimental results [11].

3. RESULTS AND DISCUSSION

3.1 Properties of activated carbon

The pore size distributions of the prepared AC from *Leucaena leucocephala* biomass is 140.53 m^2g^{-1}, 531.19 m^2g^{-1} and 735.68 m^2g^{-1} for AC from chemical activation with NaOH:char ratio 1:1, 2:1 and 3:1, respectively. The data show that the BET of the AC increased as the NaOH:char ratio increased from 1 to 3. The differences between the BET values of AC-1 and AC-2, AC-2 and AC-3 were of approximately 600 m^2g^{-1}, showing a surface area increment with increasing of the NaOH:char ratio. As observed, the increase in the NaOH:char promotes an increase of the BET but showed a decrease in the AC yields.

The surface areas of activated carbon can be up to 3000 m^2g^{-1}. The surface area of commonly available activated carbon is about 1000 m^2g^{-1}. These high surface area results from development of mainly micro- and meso pores of different size and shape. Macro pores have little contribution to the development of surface area [12]. The adsorption capacity of an activated carbon for a specific adsorbate is dependent on its physico-chemical properties such as effective surface area, pore volume and pore size distribution and surface functional groups. The development of micropores and mesopores is vital as they entrap and retain various types of adsorbate either from gas or liquid phases [13].

3.2 Batch adsorption study

Effect of contact time on the cadmium adsorption. The adsorption contact time is an important parameter to an effective interaction between adsorbate-adsorbent. This data is important because equilibrium time is one of the parameters for the economic wastewater treatment plant application [14]. Experimental studies were carried out with contact time 10 min until 160 min with initial metal ion concentrations used were 30 mg/l of Cd (II) using 0.5 g/l of adsorbent dosage at pH 7.0. Fig. 1 shows the contact time effect on the percentage removal of Cd (II). Equilibrium adsorption was established within 40 min for metal ions at initial concentrations of 30 mg/l.

The highest ratio of NaOH:char resulted in higher adsorption for removal Cd (II) due to higher surface area for adsorption. It is very clear from the results that the agitation time required for maximum uptake of metal ions by activated carbon was dependent on the initial metal ion concentration. Obviously, the initial high adsorption rate is due to the abundance of free binding sites. The meso-pores become almost saturated with Cd (II) ions during the initial stage of the adsorption process. Thereafter, the Cd (II) ions have to traverse farther and deeper into the micro-pores encountering much larger resistance, thus leading to decreased driving force and adsorption rate. Additionally, with the process of adsorption, desorption rate increased, adsorption-desorption equilibrium is also affected around equilibrium [15].

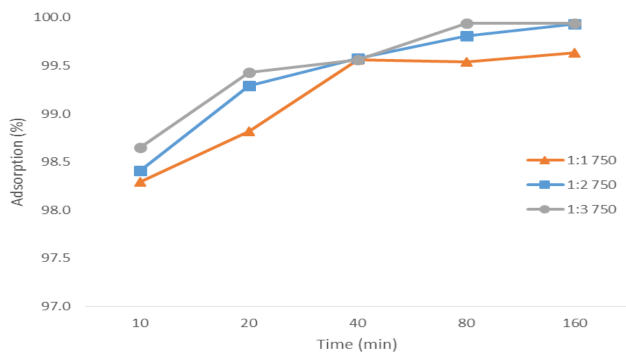

Fig. 1 Effect of contact time on removal cadmium

***3.3 Effect of pH on the cadmium adsorption*.** Fig. 2 shows that with an increase in pH, the residual of Cd (II) decreased and adsorbents are effective for the quantitative removal of Cd (II) ions at pH = 8.0. A sudden change occurred at pH ranging from 7.0 to 8.0. At higher pH (> 7.0), the adsorption of Cd (II) ions is nearly constant. The maximum removal and adsorption capacity of *Leucaena leucocephala* AC occurred at the pH 8.0. Almost 100% cadmium were removed at the pH 8.0.

Similar results with pH have been reported by many studies that adsorption capacity of adsorbents is low at acidic pH while increases at higher pH values. When the pH is < 6.0, the surface charge on the surface of *Leucaena leucocephala* AC is positive. A significant electrostatic repulsion exists between the positively charged surface and the cationic Cd (II) ions, which inhibits the adsorption of Cd (II). Besides, a higher concentration of H+ in the solution competes with Cd (II) ions for the adsorption sites, resulting in the reduced uptake. The increase in adsorption with the increase in pH can be attributed to the fact that the positively charged metal cations are repulsed less by the oxide surfaces at higher pH values. As the pH of the system increases, the number of positively charged sites decreases and the number of negatively charged sites increases on the surface of adsorbents. Obviously, a negatively charged surface site favors the adsorption of cationic Cd (II) ions due to electrostatic attraction [15].

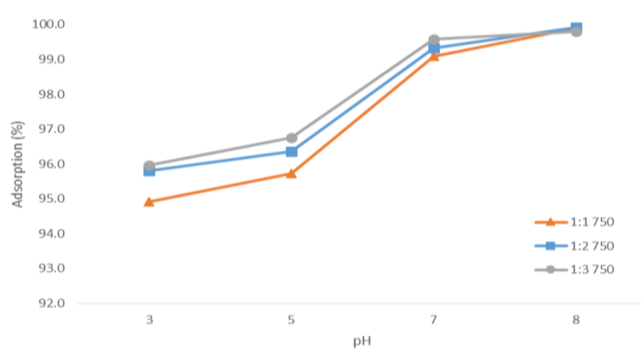

Fig. 2 Effect of pH on removal cadmium

***3.4 Effect of concentration on the cadmium adsorption*.** The effect of initial concentration on the adsorption of cadmium by *Leucaena leucocephala* activated carbon was investigated with varying solution concentrations (10, 20, 30, 40 and 50 mg/L) using 0.5 g adsorbent dose and pH 7.0. Lower initial concentration resulted in highly adsorption percentage for cadmium removal. However, for highly concentrated initial solution resulted in lower adsorption percentage. This can be conclude that, at low initial solution concentration, the surface area and the availability of adsorption sites were relatively high, and the cadmium were easily adsorbed and removed [16]. At higher initial solution concentration, the total available adsorption sites are limited, thus resulting in a decrease in percentage removal of cadmium. Fig. 3 shows the different adsorption curve for the ratio 1:1, 2:1 and 3:1. The ratio 1:1 is the lowest adsorption percentage between 2:1 and 3:1. It is because of the diferent surface area of activated carbon.

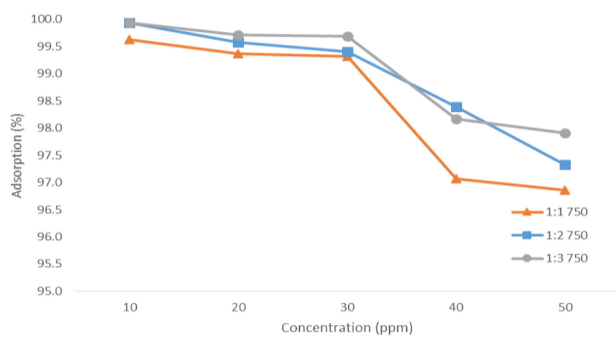

Fig. 3 Effect of concentration on removal cadmium

3.5 Effect of temperature on the cadmium removal. The effect of temperature on the adsorption of cadmium by *Leucaena leucocephala* activated carbon examine with different temperature from 30 °C to 70 °C using 0.5 g adsorbent dose, pH 7.0 and with initial concentration 40 mg/l. Based on the results in Fig. 4, adsorption percentage were incrase on temperature 30 °C and started to decrease on temperature 40 to 70 °C for both ratio. However, the ratio for 3:1 resulted in higher adsorption rate. It can be conclude that the percentage of adsorption for cadmium is decrease when the temperature is increase. So, temperature 30 °C is equilibrium point for the temperature parameter. This result has a similarity from the previous studies that has been reported from Kumar, Ramakrishnan, Kirupha and Sivanesan [17], the decreasing of adsorption rate from 86.87 to 76.25% for 50 mg/L concentration of the cadmium with an increase in temperature from 25 to 55 °C.

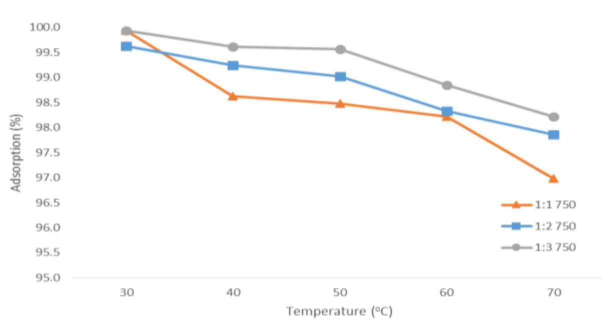

Fig. 4 Effect of temperature on removal cadmium

4. SUMMARY

In the present study, batch adsorption of cadmium removal from aqueous solution had been carried out using *Leucaena leucocephala* biomass as an adsorbent. The adsorption characteristic had been examined at a different contact time and pH values. For the contact time, the equilibrium adsorption was established within 40 min for metal ions at initial concentrations of 30 mg/l. The highest adsorption percentage for contact time is by ratio 3:1 which is NaOH:char. It is very clear from the result that, longer time required for maximum uptake of metal ions by activated carbon was dependent on the initial metal ion concentration. Generally, adsorption capacity of cadmium ions by *Leucaena leucocephala* AC increased with an increase in the pH of the adsorbate solution. The optimum pH for cadmium removal is 8.0. Based on the obtained results, activated carbon from *Leucaene leucocephala* biomass would be useful for the economic water treatment that containing cadmium metals.

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