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BIOSORPTION OF PHENOL FROM AQUEOUS SOLUTION USING WATER MELON RIND (*CITRULLUS C. LANATUS*)

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ABSTRACT

This study focuses on investigating the effectiveness of water melon rind in phenol removal from aqueous solution. The effects of various parameters (pH, initial phenol concentration, biosorbent dosage and contact time) on phenol adsorption were investigated. The pH of 2, initial phenol concentration of 40 mg/L, biosorbent dosage of 0.6 g and contact time of 6 h also deduced to be the optimum conditions for the adsorption process. The maximum phenol removal under optimized conditions was 85%. The sorption data fitted to the Freundlich isotherm with a regression coefficient of 0.9824. The kinetics was best described by the intraparticle diffusion model and Elovich equation with regression coefficients of 1 and 0.8461 respectively showing that the reaction is chemisorption on a heterogeneous surface and the intraparticle diffusion rate only is the rate determining step. The study revealed that water melon rind have a potential of removing phenol from industrial wastewaters.

Keywords*:* Biosorption, phenol, water melon rind

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1. INTRODUCTION

Phenol is produced by various industrial processes which include paint and paper making, coal gasification, crude tar refinery, plastics, insecticides and dye synthesis (Singh and Balomajumder, 2016). Phenols are among the most poisonous contaminants in industrial effluent and are toxic to humans, animals and aquatic life even at low concentrations (Dotto*et al*, 2013). Concentration levels of about 0.02 µg/L and 60 µg/kg body weight or less are considered nontoxic for aquatic life and humans respectively (World Health Organisation, 1994). Phenols poisoning may result in mutagenesis and carcinogenesis in humans and other living organisms and hence it should be removed from waste waters before discharge (Michalowicz and Duda, 2007).

Several conventional methods such as chlorination, ion exchange, electrochemical oxidation, ozonation, ultra-filtration, precipitation, chemical coagulation, solvent extraction, membrane separation, photocatalytic degradation, distillation and biological degradation have been used for the removal of phenolic compounds and their derivatives from waste water(Comninellis and Pulgarin, 1993; Mvula and von Sonntag, 2003; Iranpour *et al*, 2005; Ince *et al*, 2013;Agrawal *et al*, 2013; Dash *et al*, 2009). These methods suffer shortcomings of high costs, inability to remove trace pollutant concentrations, generation of toxic secondary pollutants and large amounts of sludge (Prasad and Santhi, 2012). The application of activated carbon (Mukherjee *et*

al, 2007; Rengaraj *et al*, 2002; Nwufo *et al*, 2014) is considered to be an efficient method for the removal of phenol from waste water due to its large surface area, micro-porous nature, high sorption capacity, high purity and easy availability. However, the employment of activated carbon as an adsorbent in developing countries had been hampered
by challenges such as high capital, regeneration and operational costs and problems associated with residue or sludge disposal. This has directed research focus to the search for low cost and readily available adsorbents which are of agricultural origin (Djebar *et al*, 2012).

The advantages of biosorption are high efficiency, ease of operation, low capital and operating costs, ease of biosorbent regeneration, eco-friendly process, minimization of chemical sludge and the use of natural low-cost and renewable biosorbent (Nadavala *et al*, 2009). Various biosorbents such as immobilized activated sludge (Aksu and Gonen, 2016; Otero *et al*, 2003),*Pleurotus sajor-cajufungus* (Denizli *et al*, 2005), *Phaneroclaute chrysosporium* (Denizli *et al*, 2004), coal fly ash (Mahadevawamy *et al*, 1997), *Trametes versicolor polyporous* (Kumar and Bandyopadhyay, 2006), beutonite (Banat *et al*, 2006), *Saccharomyces cerevisiae* (Moyo *et al*, 2012), brown alga (Rubin *et al*, 2010), *Aspergillus niger* Rao and Viraraghavan, 2002), *Lessonia nigrescens bory* and *Macrocystis integrifolia* (Navarro *et al*, 2008), *Phanerochaete chrysosporium* (Farkas *et al*, 2013) and *Pinus densiflora Sieb* bar powder (Nadavala *et al*, 2014)have been used in the removal of phenol from aqueous solutions. To the best of our knowledge, literature does not indicate any studies on the biosorption of phenol using dried water melon rind. This study therefore assesses the potential of water melon rind as effective sorbent for the removal of phenol from aqueous solutions.

2. MATERIALS AND METHODS

2.1. Reagents and instrumentation

All reagents were of analytical grade and were purchased from Sigma-Aldrich. Phenol stock solution (1000 mg/L) was prepared by dissolving 1 g phenol in 1 L deionised water. The calibration standards and test solutions were prepared by serial dilutions of the stock solution to desired concentrations. The pH value of the biosorption mixtures was adjusted to the required value using 0.1 M NaOH or 0.1 M HCl. The concentration of phenol was determined using an Ultraviolet Visible light (UV-Vis) Spectrometer (Shimadzu Model UV-1601) set at 270 nm wavelength. The adsorbent was characterized using the Thermo Scientific Nicolet 6700 Fourier Trasform Infrared (FTIR) spectrometer before and after sorption to identify functional groups.

2.2. Adsorbent preparation

The water melon rinds were collected from a local vegetable market and washed thoroughly with tape water followed by distilled water to remove impurities. The rinds were cut into small pieces and sun dried for 10 days to remove all the moisture present. The sun dried pieces were then washed with hot distilled water at 75 °C to remove any soluble mater followed by drying in an oven to constant mass at 85 ^{o}C (Reddy *et al*, 2014). The dried biomass was pulverized into a powder and sieved with a 300 µm sieve. The biosorbent was then stored in a desiccator for further use.

2.3 Batch adsorption experiments

Batch adsorption studies were performed in stoppered 250 mL flasks containing 50 mL of phenol solutions at room temperature of 25 °C at different pH (1-9), sorbent dose (0.2-2g), initial phenol concentration (5-100 mg/L) and contact time (0.5-24 h) to obtain the equilibrium data. The mixture was agitated on a shaker at a speed of 100 rpm. After attainment of equilibrium, the samples were centrifuged, and the residual phenol in the supernatant was analysed on a UV-Vis

spectrometer. All experiments were performed in replicates with reproducibility within at most 5% error and the results average was reported.

The amount of phenol adsorbed per gram of biomass and the sorption efficiency (%) were calculated according to the following equations;

$$Q_e = \frac{(C_0 - C_e)\,V}{M} \quad [1]$$

$$Sorption\ efficiency\ (\%) = \frac{(C_0 - C_e)}{C_0} \times 100 \quad [2]$$

Where, Q_e is the amount of phenol biosorbed per gram of biomass, mg/g, C_o and C_e are the initial and equilibrium concentrations of phenol (mg/L) respectively, V is the volume of solution (L), and M is the mass of biosorbent (g).

3. RESULTS AND DISCUSSION

3.1 FTIR analysis before and after Sorption

The FTIR spectra of the adsorbent before and after adsorption are shown in Figure 1. A broad peak at 3442 cm^{-1} before adsorption indicates stretching vibrations of –OH groups of cellulose, lignin and pectin. After adsorption the peak shifted to 3421 cm^{-1} indicating the binding of the hydroxyl groups to phenol by physical adsorption. The peak at 2919 cm^{-1} represents the -CH stretching vibrations of methyl and methoxy groups. The absorption band around 2850 cm^{-1} was assigned to the C–H stretching of methylene groups which disappeared after sorption indicating possible chemisorption. The bands from 1300-1000 cm^{-1} can be assigned to the C-O stretching vibrations of carboxylic acids and alcohols. The peak at 1630 cm^{-1} confirms the presence ionic asymmetric $–COO^-$ groups which lost intensity after sorption indicating its adsorbent dosage of 1 g and initial concentration of 30 mg/L. Figure 2a shows
participation in the chemical process. A peak at 1384 cm^{-1} may be assigned to the symmetric vibrations of $–COO^-$ bands of pectin and the peak lost intensity after the sorption of phenol indicating the its participation in the process.

3.2 Effect of pH

The effect of pH on the adsorption of phenol was carried out by varying the pH of the test solutions from 1-9 and keeping other parameters constant at contact time of 24 h, the effect of pH on the adsorption of phenol.

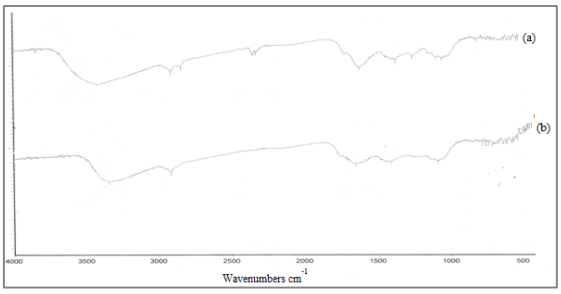

Figure 1. FTIR spectra of adsorbent before (a) and after sorption (b)

There was an increase in phenol adsorption as the pH increased up to a maximum sorption of 62.5% at pH 2. At low pH phenol is in its neutral form, hence it forms hydrogen bonds with most of the functional groups of the biosorbent. Above pH 2 there is a decrease in adsorption due to increase in competitive adsorption between the OH^- and the phenolate ions (Anandkumar and Mandal, 2009). The decrease in sorption efficiency at higher pH values could also be attributed to electrostatic repulsions between the negatively charged adsorbent surface and the phenolate ions in solution. Related trends were reported for the biosorption of phenol using yeast (Antizar-Ladislao and Galil, 2004).

3.3Effect of contact time

The effect of contact time on the adsorption of phenol was investigated by varying the contact time from 0.5 to 24 h using adsorbent dosage of 1 g, initial phenol concentration of 30 mg/L and optimized pH of 2 from above. The effect of contact time on phenol adsorption is shown in Figure 2b. The adsorption of phenol increased with time until

it reached a constant value (81%) at 6 h. An increase in adsorption with time can be attributed to a large number of surface active sites available (Agarry and Aremu, 2012). The leveling off of phenol adsorption capacity is attributed to saturation of adsorption sites. Comparable results were reported for the adsorption of phenol onto powdered and granular activated carbon prepared from *Eucalyptus* wood (Trancredi *et al*, 2004).

3.4. Effect of adsorbent dosage

The effect of adsorbent dosage on the sorption of phenol was studied by varying the dosage from 0.2 to 2 g at initial phenol concentration of 30 mg/L, optimized pH of 2 and optimized contact time of 6 h from above. As indicated in Figure 2c, there was a rapid increase in the sorption of phenol from 57.4 to 85.4% when the biosorbent dose was increased from 0.2 to 0.6g. Increasing the adsorbent dosage increases the number of sorption sites available for the sorbent-sorbate interaction resulting in increase in the quantity of phenol removed from the solution (Alzaydien, and Manasreh, 2009). Above a dosage of 0.6g phenol adsorption becomes almost constant. This is due to a high adsorbent concentration which results in a very fast superficial adsorption onto the sorbent surface producing a lower solute concentration than when the dosage is low (Banerjee*et al*, 2012). A similar trend was reported for the adsorption of phenol using agro waste (*Hemidesmus Indicus*) based activated carbon (Srihari and Das, 2009).

3.5Effect of initial phenol concentration

The effect of initial phenol concentration on its sorption on water melon rind adsorbent was investigated by varying the concentration from 5-100 mg/L, at optimized pH of 2, optimized contact time of 6 h and optimized adsorbent dosage of 0.6 g from above. Figure 2d shows the effect of initial phenol concentration on its adsorption. Phenol sorption efficiency increases sharply from 4% to 75.4% at low initial concentrations (5 to 40 mg/L) followed by gradual increase and levelling off at higher initial concentrations (above 40 mg/L). The increase in phenol adsorption with increasing initial concentration may be attributed to increased interaction between phenol and the biosorbent sites. The increase can also be attributed to increase in mass transfer driving force as the phenol concentration is increased. A similar trend was reported for the biosorption of phenol in fluidized bed using rice husk (Al-Sultani Kadhim and Al-Seroury, 2012).

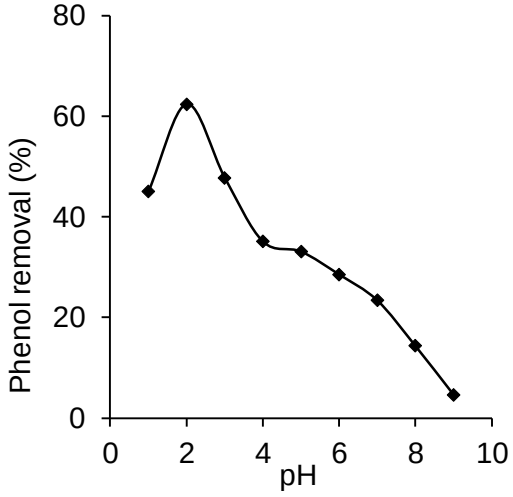

Figure 2a. Effect of pH on phenol removal

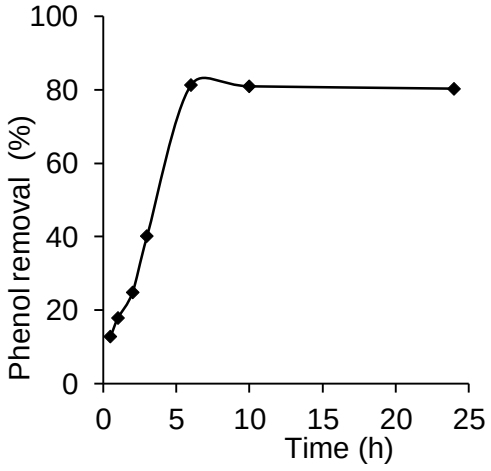

Figure 2b. Effect of contact time on phenol removal

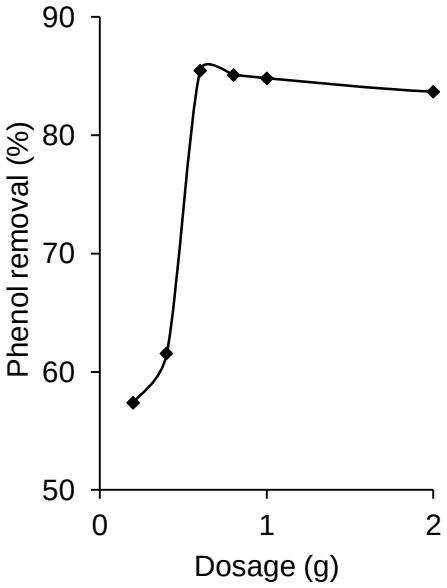

Figure 2c. Effect of dosage on phenol removal

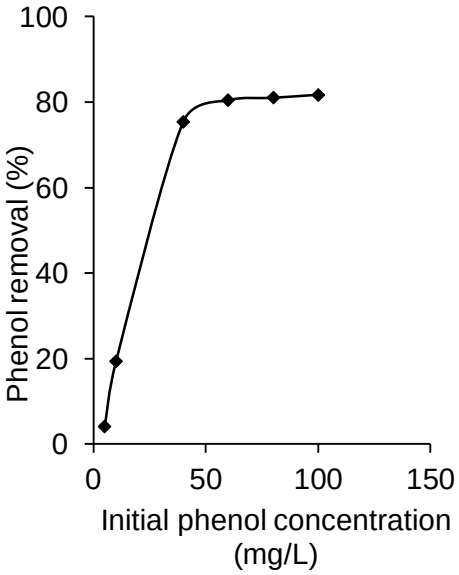

Figure 2d: Effect of initial concentration on phenol removal

3.6 Adsorption Isotherms

The data obtained from the effect of initial phenol concentration at optimized conditions of pH, contact time and adsorbent dosage was fit to adsorption isotherms. The equilibrium data on the adsorption of phenol on water melon rind was found to fit best to the Freundlich isotherm with a correlation coefficients (R^2) of 0.9824. The Freundlich isotherm assumes that adsorption takes place on heterogeneous surfaces with non-uniform distribution of energy level and its linear form is given as:

$$\ln q_e = \ln K_f + \tfrac{1}{n} \ln C_e [3]$$

Where, K*f* and *n* are the Freundlich constants indicating the adsorption capacity and intensity respectively. These constants were determined from a plot of ln q_e versus ln C_e (Figure 3). The value of *n* (0.23 g/L) for the Freundlich isotherm was less than 1indicating that the adsorption of phenol onto water melon rind is a chemical process. A high value of 1/n showed that the surface of the adsorbent is heterogeneous (Farhan *et al*, 2012) and possesses high adsorption intensity (Srihari and Das, 2008).

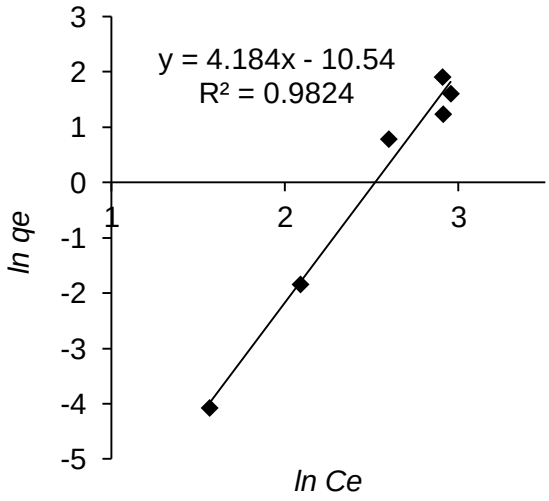

Figure 3. Linearized Freundlich isotherm for phenol adsorption

3.7 Adsorption kinetics

The data obtained from the effect of varied contact time on the sorption of phenol on water melon rind under optimized conditions was used fit to kinetic models. Adsorption

kinetic data was analysed using the intraparticle diffusion model (McKay and Poots, 1980)and the Elovich equation (Chien and Clayton, 1980).The intraparticle diffusion equation can be written as;$q_t = k_{id} t^{1/2} + C$ [4]

Where, $q_t$is the quantity of adsorbate adsorbed, k_{id} is the intraparticle diffusion rate constant and *C* is related to the thickness of the boundary layer. Figure 4a shows a linear plot of $q_t$as a function of $t^{1/2}$. The straight line observed with zero intercept shows that phenol sorption process was controlled by the intraparticle diffusion only (Ekpete *et al*, 2012).

The linear form of Elovich equation is represented by the following equation;$q_t = \frac{1}{\beta}\ln(\alpha\beta) + \frac{1}{\beta}\ln t$ [5]

Where, α is a constant representing the initial adsorption rate (mg/g/min) and β is the constant showing the extent of surface coverage and activation energy for chemisorption (mg/g). The Elovich plot is shown in Figure 4b. A fairly high value of R^2 (0.8461) shows a good fit of the equation to the experimental data indicating that the sorption process can be best described as a chemisorption on a heterogeneous surface.

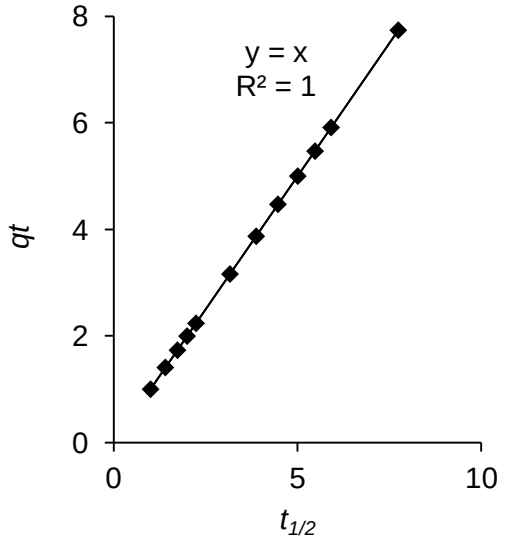

Figure 4a. Intra particle diffusion rate plot for phenol adsorption

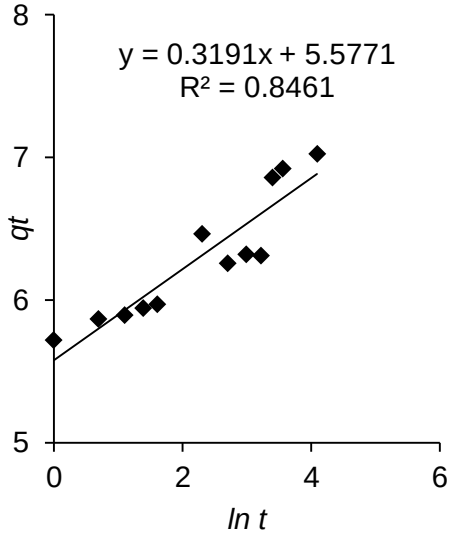

Figure 4b. Elovich plot for phenol adsorption

4. CONCLUSION

Water melon rinds have shown a great potential for the removal of phenol from waste industrial water. The optimum conditions for phenol removal were pH 2, initial concentration of 40 mg/L, biosorbent dosage of 0.6 g and 6 h contact time. The data fitted well to the Freundlich isotherm. Adsorption kinetics fitted the intraparticle diffusion model. The Elovich equation revealed that the adsorption of phenol is chemisorption on a heterogeneous surface. The presence of functional groups on the surface of metal melon rind plays a role in the process of phenol adsorption.

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