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UTILIZATION OF DURIAN PEEL AS POTENTIAL ADSORBENT FOR BISPHENOL A REMOVAL IN AQUOEUS SOLUTION

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Graphical abstract

Agricultural waste – Durian Peels

Abstract

This study explored the low-cost adsorbent of durian peel for BPA removal from aqueous solutions. The effect of various operational parameters such as contact time, temperature, concentration, agitation and pH on the adsorption of BPA was investigated using the batch adsorption study. It was found that Durian peel can be used as a low cost adsorbent for the removal of BPA in aqueous solution after treated with sulfuric acid. The effects of morphology, functional groups, and surface area of adsorbent, before and after pretreatment with sulfuric acid and reaction were investigated by using FESEM, FTIR, and BET. The present study indicates that durian peel had removed 69.63% of BPA with adsorption capacity of 4.178 mg/g for 24 hours. The result proved that this treated agricultural waste was promising material as an alternative adsorbent for the removal of BPA from aqueous solution. Kinetic study of the results gave a pseudo-second order type of mechanism while the adsorption characteristics of the adsorbent followed the Langmuir adsorption isotherm.

Keywords: BPA, durian peel, pseudo-second order, sulfuric acid, langmuir

Abstrak

Kajian ini mengkaji adsorben kos rendah kulit durian untuk pembuangan BPA daripada larutan akueus. Pelbagai faktor parameter seperti masa proses penjerapan, suhu, kepekatan larutan , kelajuan putaran dan pH pada penjerapan BPA telah dikaji dengan menggunakan eksperimen proses kelompok . Ia telah mendapati bahawa kulit Durian boleh digunakan sebagai adsorben kos rendah untuk penyingkiran BPA dalam larutan akueus selepas dirawat dengan asid sulfurik. Kesan daripada morfologi, kumpulan berfungsi dan luas kawasan permukaan penjerapan sebelum dan selepas rawatan dengan asid sulfurik dan tindak balas telah dikaji dengan menggunakan FESEM , FTIR , dan BET. Kajian ini menunjukkan bahawa kulit durian telah menyingkirkan 69,63 % BPA dengan kapasiti penjerapan 4,178 mg / g untuk tempoh masa 24 jam. Keputusan ini membuktikan bahawa sisa tumbuhan yang dirawat telah menjanjikan sebagai bahan penjerap alternatif untuk penyingkiran BPA daripada larutan akueus. Kajian kinetik memberikan jenis mekanisme pseudo-kedua manakala ciri-ciri penjerapan adsorben mengikuti penjerapan isoterma Langmuir.

Kata kunci: BPA, kulit durian, pseudo-kedua, asid sulfurik, langmuir

1.0 INTRODUCTION

Recently, awareness issues on new emerging contaminants are raised since it discovered in natural streams around the world due to human and industrial activities. New emerging Pollutant (NEP) including BPA was reported can affect human and aquatic life yet it has no regulatory standard. Other conventional methods are not really effective for all kind of NEP removal, thus this study investigated the adsorption process in the removal of Bisphenol a (BPA) [1].

Various actions had been implemented for removing emerging contaminants from drinking and waste water, such as reverse osmosis, membranes, filtration, coagulation/flocculation, advanced oxidation processes, and microorganism [2-5].

Agricultural waste comprise several basic elements such as lipids, lignin, cellulose, hemicellulose, hydrocarbons, extracts, proteins, starch, simple carbohydrates and water and also containing a variety of functional groups that present in the binding process and can sequestering of pollutant [6,7,8]. Cellulose elements are significance in adsorption process since it has potential sorption capacity for various pollutants [9,10]. Commonly, functional groups that present in agricultural waste are molecules alcohols, phenolic, amido, carboxyl, carbonyl, amino and esters etc. [8,11,12].

The agricultural wastes are low in cost, readily available and abundance resources and has unique chemical composition, thus it frequently used for investigated the removal of pollutants from aqueous solutions. Some kind of agricultural waste that are used such as orange peel [13, 14], hazelnut shell, almond shell, walnut, sawdust, poplar,[15], rice husk [16], sugarcane bagasse pith [17], coir pith [18], coconut bunch waste [19], sawdust [20], and papaya seed [21].

For this study, Durian peel was chosen as potential adsorbent for BPA removal in aqueous solution. Durian peel is not widely used in the adsorption process for NEP removal. Some of researchs that were studied such as removal of acid dye by durian peel [19], adsorption of basic green 4 dye by activated carbon durian peel [22], adsorption of basic blue 3 by durian peel [23] and ammoniacal nitrogen removal by activated carbon durian peel [24] slightly larger particles like activated carbon.

Adsorbents from agricultural waste are used either in their natural form, apply physical or chemical modification as pre-treatment [8,25]. Pretreatment of the adsorbents are for the purposes of removing all colors, metals and any soluble organic compounds in aqueous solutions and in order to enhance its ability of adsorption [26]. Different adsorbents are treated with different kind of modifying agents, such as organic compounds (epichlorohydrin, ethylenediamine, formaldehyde, methanol), mineral, dyes, acid and base solutions (calcium hydroxide, sodium carbonate, sodium hydroxide) and oxidizing agents and depending on types of adsorbate or pollutant to remove [8].

From the reviews, show that usually agricultural wastes were used in the adsorption of heavy metal and dye removal. Thought these agricultural wastes widely investigated for pollutant removal, there are still limited studies on new emerging pollutant (NEP) removal such as endocrine disrupting chemicals (EDC). Most of agricultural waste studies on pollutant removal were optimized by activated carbon which is high in cost of preparation; however, this study just treated the adsorbent with sulfuric acid to improve the adsorption capacity for BPA removal.

2.0 EXPERIMENTAL

2.1 Preparation of the Adsorbents

Durian peels were collected from marketplace in Johor Bahru. The sample was washed with tap water and repeated with distilled water to remove any impurities adhering to the surface and then the samples were cut into pieces and oven-dried at 105 °C for overnight. The dried samples were grinded and sieved through 30-mesh to get a consistent size of adsorbent powder. The adsorbent were then treated with sulfuric acid according to the former method [27]. The dried raw adsorbents were soaked in concentrated 98% H_2SO_4 (1:1 w/v), then were kept in a hot air oven at 105 °C for 12 hours. These samples were then washed with distilled water then were dried again in a hot air oven at 105 °C for hours. The treated adsorbents were stored in an airtight container for further experiments.

2.2 Preparation of BPA Solutions

The highest available grade of BPA (purity 95%, chemical formula $C_{15}H_{16}O_2$, MW = 228.29, wavelength = 278 nm) was purchased from Sigma-Aldrich (Milwaukee, USA). The stock solution of BPA was prepared by dissolve 0.02 g of BPA in 10% of ethanol (95%) and 90% of distilled water. Distilled water were used to dilute the stock solution of BPA depends to the desired of concentrations.

2.3 Characterization of the Adsorbent Materials

Surface texture and morphology of raw and treated durian peel was analyzed by the field emission scanning electron microscopy (FESEM JEOL 6335F-SEM, Japan) and elementary analyses were performed simultaneously using an EDX spectrometer. The Brunauer–Emmett–Teller (BET) surface areas and monolayer pore volumes of the adsorbents were determined by using provided software, surface analyzer (Quantachrome Instrument, USA). FTIR (Spectrum one, Perkin Elmer, USA) was used to analyzed the functional groups on the surface of raw and treated durian peel with the spectral range varied from 4000 to 400 cm^{-1}.

2.4 Effect of Temperature

The effect of temperature on BPA adsorption was carried out with 30 ml of the stock solution of 20 mg/L with amount 0.1 g of adsorbent were mixed into the 100 ml round bottom flask and kept for agitation at 150 rpm using rotary orbital shakers. The experiments were carried out at room temperature, 30°C, 40 °C, 50 °C and 60 °C for 24 h. After the batch adsorption method, the sample was analyzed by using the UV-Vis Spectrophotometer.

2.5 Effect of Initial pH Solution

The pH of the solution varied from 3 to 9, and the mass of adsorbent (0.1 g), the volume of solution (30 mL), the initial concentration of solution (20 mg/L), the temperature (room temperature) and the agitation (150 rpm) were kept constant for 24 h. The solution pH was adjusted with diluted 0.1 M HCl and 0.1 N NaOH.

2.6 Effect of Adsorbent Dosage

The adsorbent dosage were varied with amount (0.05, 0.1, 0.3 and 0.5 g) were mixed into 30 ml of 20 mg/L BPA in the 100 ml round bottom flask and kept for agitation at 150 rpm using rotary orbital shakers for 24 h. The experiment were run by the batch adsorption study and were analysed by using the UV-Vis Spectrophotometer

2.7 Effect of Agitation

The agitation of shaker were varied from 50 rpm, 100 rpm, 150 rpm, 200 rpm and 250 rpm, and the mass of the adsorbent (0.1 g), the volume of solution (30 mL), the initial concentration of solution (20 mg/L), the temperature (room temperature) and the initial pH solution (3) were kept constant for 24 h. Then, the sample was analyzed by using the UV-Vis Spectrophotometer.

2.8 Batch Adsorption Studies

The BPA adsorption isotherm was determined by using batch adsorption study at the initial pH value of 3. Initial BPA concentration was varied from 5 mg/L to 100 mg/L. In other to obtain a concentration of 100 mg/L BPA, 0.1g of BPA powder was dissolved in 10% of ethanol and 90% of distilled water, and then the solution was diluted to various concentrations depending on the experiment. Adsorption experiments were performed by adding 30 ml of BPA solution into 100 ml conical flask containing 0.1 g of treated adsorbent. The pH value has been adjusted by NaOH or HCl solution. The mixtures were sealed and shaken at a rate of 150 rpm at room temperature for 24 h. The solutions were then filtrated by using Whatman no. 1 filter paper and determined the BPA concentration by using a UV-Vis spectrophotometer (NANOCOLOR® VIS, Macherey-Nagel, Germany) at 277 nm. Ultimately, the solids obtained were oven-dried at 105°C for one daytime for analyzed by FESEM, FTIR, and BET.

3.0 RESULTS AND DISCUSSION

3.1 Morphology of durian peel

As hown in Figure 1, the photograph of raw durian peel has no pores. After treatment, more pore appear resulting the adsorbent was found to be quite efficient in getting rid of BPA. The sulfuric acid treatment responsible for the growth in the porosity of the durian peel adsorbent, which increased its ion-adsorption capacity, probably because the number of available binding sites increased [28,29,30].

A

B

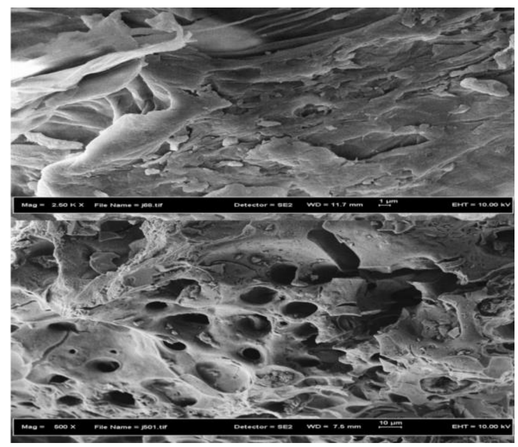

Figure 1 Field Emission Scanning Electron Microscope (FESEM) image of raw durian peel (A) and treated durian peel (B)

3.2 Effect of Temperature

The effect of the temperature on BPA solution was done with varied from 25 – 60 °C. The adsorption condition was 0.1 g adsorbent in 30 ml of 20 mg/L BPA and shaken at 150 rpm for 24 hours. As can be seen in Table 1, the percentage removal of BPA for durian peel decreased from 69.63% to 22.42% as the temperature increased. The adsorption capacity was also decreased with the increased of temperature. Thus, in this study, the temperature gives a significant effect on BPA removal, which the result indicated that the room temperature or temperature at 25°C was the best condition for BPA removal. This result is same with studied by Liu [32] who stated that low temperature are the most suitable for BPA removal by selected activated carbon adsorbent.

Table 1 Effect of temperature on BPA adsorption onto durian peel

Temperature (°C)	Durian peel	
	Percentage Removal (%)	Absorption Capacity q_e (mg/g)
25(Room Temp)	69.63	4.642
30	65.90	4.393
40	55.34	3.689
50	47.89	3.193
60	22.42	1.495

3.3 Effect of Initial pH Solution

The BPA removal efficiency slightly decreased when the pH values increased from 3 to 9, indicating that under acidic conditions, the binding attraction between BPA and the binding sites did not significantly change [33]. This means the current study indicated that the sorption capability of the sorbent did not increases by the increase in the negative charge of BPA. As the initial pH solution increases, the electrostatic repulsion between BPA and the sorbent becomes increasingly stronger and overcomes the binding affinity because the overall surface charge becomes more negative. Thus, this resulted low in percentage removal and adsorption capacity of BPA onto the adsorbents at alkaline conditions.

3.4 Effect of Adsorbent Dosage

The effect of the sorbent dose on BPA removal at an initial concentration of 20 mg/L, pH 3.0, agitation rate of 150 rpm at room temperature was determined. The effect of the adsorbent dosage on the adsorption of BPA was investigated by varying the dosage of adsorbent from 0.05 to 0.5 g/30 mL. The results show that the percentage removal increased when adsorbent added from 0.05 to 0.1 g, then gradually decreased when adsorbent dosage added from 0.1 to 0.5 g respectively. However, the adsorption capacities of all adsorbents decreased by increasing the adsorbent dosage. This result showed that the adsorption efficiency is dependent on the amount of adsorbent and volume of solution added. A relatively low dose approximately 0.01 g in 30 mL is sufficient for the treated adsorbent to achieve a reasonably good removal performance. Therefore, the optimum sorbent dose is 0.01 g for 30 mL, which was used for the subsequent experiments for economic purposes [33]. Tsai *et al*. [34] stated that the resulting in increased BPA percentage removal when the sorbent dose added because surface area of adsorbent becomes greater and amount of available binding sites for BPA were increased. However, the adsorption capacity of the adsorbent decreased as the adsorbent dose increased because with increasing sorbent mass concentration, the amount of BPA adsorbed onto per unit mass of a adsorbent were reduced. Similar trends have been reported for BPA sorption onto activated carbon [34] and malachite green sorption onto clayey soil [35].

3.5 Effect of Agitation

The effect of the agitation on BPA solution was done with varied from 50 – 250 rpm. The adsorption condition was 0.1g adsorbent in 30 ml of 20 mg/L BPA, pH 3 and contact time 24 hours. As can be seen in Table 2, the percentage removals of BPA and adsorption capacity for adsorbents were not constant by increase of agitation (rpm). The obtain results show that the different rpm gave the little bit differ of percentage removal and adsorption capacity. However, in moderate agitation which is 150 rpm, the results show the highest in removal and adsorption capacity of BPA. Thus, agitation 150 rpm show the most suitable for this study.

Table 2 Effect of agitation speed on BPA adsorption onto durian peel

Agitation (rpm)	Durian peel	
	Percentage Removal (%)	Absorption Capacity Qe (mg/g)
50	58.76	3.525
100	66.83	4.010
150	69.63	4.178
200	68.70	4.122
250	63.10	3.786

3.6 Effect of Initial Concentration BPA

The initial concentration of the BPA solution was varied from 5 - 100 mg/L. As can be seen in Figure 2 the percentage removal of BPA for durian peel increased from 29% to 80% as the initial BPA concentration increased from 5 – 40 mg/L, however percentage removal decreased at concentration 60 - 100 mg/L which were 75% to 63% removal. While, the adsorption capacity increased from 0.4375 mg/g to 19.0 mg/g with an increase in the initial BPA concentration. This was suggested due to the accumulation of BPA adsorbed by adsorbent per unit mass. The adsorption at different concentrations is rapid in the initial stages and gradually decreased after the optimum rate. The q_e values increased with increasing initial BPA concentrations. The initial concentration plays a significant role in determining the maximum sorption capacity of the adsorbents for BPA [33]. The initial concentration provides an important driving force to overcome all mass transfer resistances of the BPA between the aqueous and solid phases. Hence a higher initial concentration of BPA will enhance the adsorption process [36].

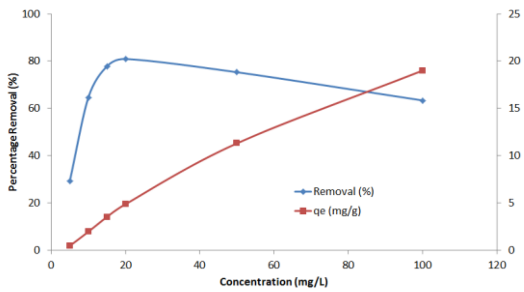

Figure 2 Effect of initial BPA concentration onto durian peel

3.7 Effect of Contact Time

Figure 3 showed that the BPA removal rate and adsorption capacity of durian peel adsorbent with contact of time of 38 h. The removal rate and adsorption capacity of the adsorbents were increased gradually over time. The adsorption test was observed over 38 h of adsorption time and the optimum adsorption equilibrium was reached at time 24th h. At the first 2 h, durian peel showed high adsorption capacity and removal of BPA. This might be because of morphology effect of the adsorbents and due to the availability of readily accessible sites. The stationary state of durian peel was reached after 24 h, indicating that the adsorbent was saturated [31]. After the optimal adsorption was achieved, at time 30th onward the percentage removal of BPA was slightly decreased. This might due to the release of some BPA molecules from adsorbent surface

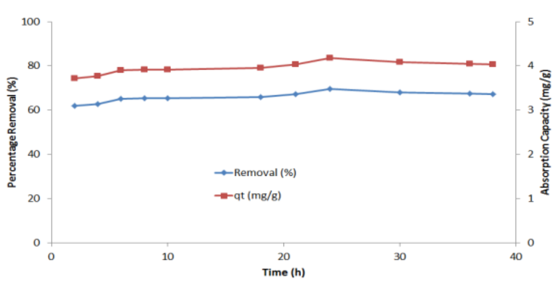

Figure 3 Effect of contact time on BPA adsorption by durian peel

3.8 Adsorption Kinetic

The kinetics data for BPA adsorption were calculated with the pseudo-first-order and pseudo-second-order models. Table 3 showed kinetics parameters of BPA adsorption onto durian peel. For pseudo-first-order model, graph were plotting by ln(q_e-q_t) vs t, while, for the pseudo-second-order models, graph were plotting against t/q_t vs t. The correlation coefficients (R^2), kinetics constant (K_1) and q_e values had been calculated from the data obtained. Based on the experiment, the amount of sorbed BPA at equilibrium (q_e) for durian peel was 4.178 mg/g, which was almost same as the calculated value for pseudo-second-order model which is 4.0469 mg/g. Besides, the R^2 in pseudo-second-order model gave a value of 0.9998 that indicated a better fit. While, the pseudo-first-order obtained the R^2 value of 0.8529 which was lower than the second order. Thus, this results showed that the adsorption of BPA obey the pseudo second-order model.

Table 3 The kinetic parameter of BPA adsorption

Kinetic Parameter	Durian Peel (q_e = 4.178 mg/g)
Pseudo-first-order	
q_e (calculated)	0.4475
K_1	0.0491
R^2	0.8529
Pseudo-second-order	
q_e (calculated)	4.0469
K_2	1.9619
R^2	0.9998
Intraparticle Diffusion	
C	3.6244
K_{diff}	0.0882
R^2	0.8626

3.9 Adsorption Isotherm

Table 4 The adsorption isotherm parameters of BPA adsorption

Isotherm parameter	Durian Peel
Langmuir	
q_m	34.0136
K_L	0.0449
R^2	0.9744
Fruendlich	
K_F	1.9422
n	1.4449
R^2	0.973

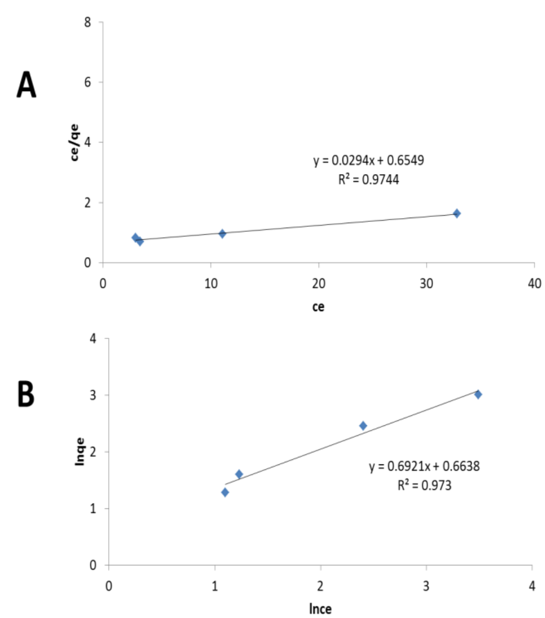

Figure 4 Langmuir model (A) and Freundlich model (B) for adsorption of BPA onto durian peel

Isotherm study is a basic technique for determining the nature of adsorption between the adsorbent and BPA. It indicates the distribution of BPA in the equilibrium phase. Langmuir and Freundlich are the most commonly used models for evaluating the experimentally acquired adsorption isotherms. Figure 4 shows the adsorption isotherm model of durian peel. For the Langmuir isotherm model, the graph was plotting by C_e/q_e vs C_e then the slope will be equal to $1/q_m$ and the intercept are equal to $1/(K_L q_m)$, while for Freundlich, the graph was plotting by $\ln q_e$ vs $\ln C_e$ and for temkin model, the graph was plotting by q_e vs $\ln C_e$.

As shown in the Table 4, the adsorption of BPA by adsorbents were suggested following the Langmuir model which obtained the R^2 equal to 0.9998, compared to Freundlich and Langmuir gave the R^2 value of below than 0.9 respectively. In the Temkin equation, A is the Temkin equilibrium binding constant (L/g) and B is the Temkin constant related to heat of sorption (J mol^{-18}) [19]. By comparing the correlation factors of the Langmuir, Freundlich and Temkin isotherm models, it is proved that the Langmuir model provides a better fit for explaining the adsorption of BPA onto the durian peel. , it is proved that the Langmuir model provides a better fit for explaining the adsorption of BPA onto all the adsorbents. This Langmuir model assumes surface containing a limited number of adsorption sites and apply monolayer adsorption with no transmigration of adsorbate in the smooth of surface [37].

4.0 CONCLUSION

This research was investigated the agricultural waste as potential adsorbents for BPA removal from aqueous solution. Durian peel had achieved the highest removal of BPA at time 24^{th} hours. Selection of an appropriate adsorbent is one of the key issues regarding the maximum removal of a given type of pollutant, depending upon the characteristic of the adsorbent and adsorbate. The outcomes of this study suggested that the durian peel as a potential adsorbent for BPA removal in aqueous solution. Adsorbent durian peels are economical and easily available resources. Thus, durian peel could be used as low-cost adsorbents which leading to environmental benefits such as reduce the waste material and monetary values of water and effluent handling.

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