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## IMMOBILIZATION OF RAMBUTAN (*NEPHELIUM LAPPACEUM*) PEEL AS A SORBENT FOR BASIC FUCHSIN REMOVAL

The potential of rambutan (*Nephelium lappaceum*) peel as a low-cost adsorbent in an immobilized condition to remove Basic Fuchsin from aqueous solution was studied. The effect of initial dye concentration, contact time, pH and adsorbent dosage for the dye removal was studied at room temperature (26±2 °C). Infrared spectrum of rambutan peel indicates the presence of C–O, –OH and C=C functional groups. The adsorption rate was found to be high at the first 30 min and the process reached equilibrium after 3 h with more than 80% of dye being removed. The uptake of Basic Fuchsin was more promising at pH slightly above 7. The study of sorption model and kinetic model showed that the adsorption of Basic Fuchsin on rambutan peel conformed to the Freundlich isotherm and pseudo-second order kinetics with a high correlation coefficient,  $R^2 > 0.98$ . This indicates the Basic Fuchsin adsorbs chemically on the heterogeneous surface of rambutan peel. The maximum adsorption capacity determined from the Langmuir isotherm was 108.696 mg/g. It seems that rambutan peel has the potential to substitute activated carbon as an alternative low-cost adsorbent in the treatment of the dye effluent.

### 1. INTRODUCTION

Dyes, commonly used in food, paper, textile, tanning and cosmetic industries, are usually classified based on their chemical structure or application. Three major classes of dyes are cationic, anionic and non-ionic ones. In the cationic dye, the major component of its structure is positively charged. It is also called basic dye because the introduction of alkaline substance will convert most of the cationic dye into water-insoluble dye bases. The anionic dye contains a negatively charged functional group in the molecule, usually the sulfonate group. It serves as a hydrophilic substituent which makes the

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dye dissociates completely under acidic conditions. Non-ionic or disperse dyes are basically neutral ones. Their molecules do not carry neither positive nor negative charge. Such dyes have very low water solubility, and they only disperse in a colloidal form. Non-ionic dyes are suitable for dyeing hydrophobic fabrics and polyester.

It has been estimated that there are about 10 000 textile dyes available in worldwide with the annual production of  $7 \times 10^5$  Mg [1]. However, due to the inefficiencies of the dyeing processes, approximately 10–15% of the used dyes can find their route to the ecosystem when being discharged directly to the environment [2]. The high percentage of textile dyes lost or discharged to the environment will cause serious pollution, especially to rivers or streams. A highly visible coloured wastewater is undesirable because it restricts light transmission and reduces the photosynthesis rate of the aquatic plants. Besides, dye itself are often toxic, carcinogenic, mutagenic, or teratogenic to aquatic life and human health. Therefore, an adequate treatment must be performed to remove dyes from wastewater.

The feasibility of using industrial waste, agricultural by-products and catalysts for various dyes removal has been studied extensively and literature survey revealed that these materials generally possess some unique features and serve as an attractive alternative for colored waste [3–9].

Rambutan (*Nephelium lappaceum*), belonging to the Sapindaceae family, is a tropical fruit that can be found easily in South-East Asia, especially in Thailand and Malaysia. It is a medium sized tree which can grow up to a maximum height of 20 m. Rambutan trees fruit twice a year, giving about  $5 \times 10^5$  Mg of harvest quantities in South-East Asia [10]. In addition, the consumption of fresh rambutan fruits or those from the canned process lead to high production of agriculture waste such as their seeds and peels. Disposal of large quantity of agriculture wastes leads to serious environmental problem. Reuse of the agriculture waste such as rambutan peel in other purposes may solve the disposal problem.

Therefore, in this study, attention is being focused on the possibility of utilizing this local fruit waste as the adsorbent for cationic dye, Basic Fuchsin. This cationic dye appears to be green in powder form and purple in solution. It is also known as Basic Violet 14 or Rosaniline chloride. Cationic dye was selected as the adsorbate in this study because it is widely used in textile and leather industries and known to be more toxic than other classes of dyes [11, 12].

## 2. MATERIALS AND METHODS

*Adsorbent.* Rambutan peels were collected from the local market, cut into small pieces and washed with tap water for several times to remove all the pollutants. After that, they were sundried until they turned into brown colour and became crispy. The dried peels were then blended into a powder form and the powder was sieved to obtain

particle size of less than 1 mm. The powdered rambutan peels were boiled for 1 h to avoid the colour leaching problem. Then the material was dried in the oven at 60 °C for 24 h to ensure complete removal of water. The dried powder (BRPP) was then stored in a glass container for future use.

*Adsorbate.* The synthetic dye, Basic Fuchsin was purchased from the Acros Organics. It has a molecular formula of  $C_{20}H_{19}N_3.HCl$  with the molecular weight of 337.84 g/mol. A 1000 mg/dm<sup>3</sup> stock solution of Basic Fuchsin was prepared and then dissolved to obtain the desired concentration for each experiment.

*Immobilization of the adsorbent.* The adsorbent was confined within a plastic canister and agitation was provided to enhance the dye uptake. Filtration or centrifugation steps were not needed towards the end of the experiment as the adsorbent can be easily removed from the plastic canister.

*Scanning Electron Microscopy (SEM).* The surface morphology of BRPP (before and after adsorption) was examined using a FESEM JSM 6701F (JEOL).

*Fourier transform infrared spectrophotometry (FTIR).* A FTIR spectrophotometer was used to analyse the functional groups present on the surface of BRPP. The BRPP was mixed with potassium bromide by using mortar and pestle. Then the mixture was subjected to high pressure using a hydraulic press. Small discs were formed and the FTIR analysis of the BRPP in solid phase was performed using a Perkin-Elmer System 2000 FT-IR spectrometer. The spectra of BRPP were recorded before and after adsorption.

*Batch adsorption studies.* The experiments were carried out at room temperature (26±2 °C). 1 dm<sup>3</sup> of Basic Fuchsin dye solution was poured into a clean aquarium (glass tank) with an aquarium pump. A pipe was connected from the pump to the immobilised container which contained 2 g of BRPP to allow a full contact between dye solution and the adsorbent and to homogenize the dye solution in the system. The supernatant was analyzed for its dye concentration using a Perkin Elmer UV-Vis double beam spectrophotometer. All measurements were made at the wavelength corresponding to maximum absorption for Basic Fuchsin,  $\lambda_{max} = 542$  nm. Sorption experiments were performed at the natural pH of the dye solution which is 5.7. pH of the dye solutions was measured at the end of the experiment and it was found to remain constant.

The percentage uptake of Basic Fuchsin was determined from the following equation:

$$\text{Uptake} = \frac{C_0 - C_e}{C_0} \times 100\% \quad (1)$$

where:  $C_0$  is the initial concentration of dye solution and  $C_e$  is its concentration at equilibrium.

*Effect of initial dye concentrations and contact time.* 2 g of BRPP was poured into the plastic canister in immobilized form and placed in a glass tank with 1 dm<sup>3</sup> of Basic Fuchsin solution. Then, ca. 5 cm<sup>3</sup> of sample solutions were withdrawn at the specified time intervals (0.5, 1.0, 3.0, 5.0, 10.0, 15.0, 30.0, 60.0, 120.0, 180.0, 240.0 min) for UV-Vis analysis. The experiments were carried out under identical experimental condition for all time intervals. Three various dye concentrations (100, 150, and 200 mg/dm<sup>3</sup>) were examined.

*Effect of pH.* Solutions of the Basic Fuchsin in the pH range from 2 to 10 were examined. pH was adjusted by adding HCl or NaOH to particular solutions. No noticeable color change of the dye solution was visible, most probably due to the concentration and the stability of the dye itself. Each sample contained 1 dm<sup>3</sup> of the dye solution (200 mg/dm<sup>3</sup>) and 2.0 g of BRPP. After adjusting pH, the solution was agitated for 4 h. Then approximately 5 cm<sup>3</sup> of the solution was collected to be analysed for the dye concentration.

*Sorption isotherm.* The dye solutions (100, 150, 200, 250 and 300 mg/dm<sup>3</sup>) were prepared and equilibrated for 4 h. The final dye concentration was then determined using a UV-Vis spectrophotometer.

### 3. RESULTS AND DISCUSSION

#### 3.1. CHARACTERIZATION OF BRPP

The IR spectra of BRPP in the range of 4000–400 cm<sup>-1</sup> before and after adsorption are shown in Fig. 1.

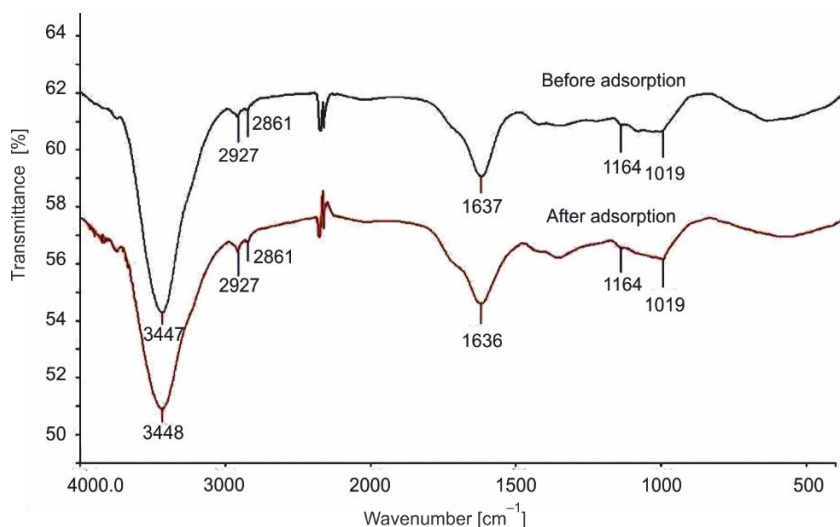


Fig. 1. IR spectrum of BRPP before and after adsorption

A strong absorption band in the range of 3600–3000  $\text{cm}^{-1}$  is visible, indicating the presence of hydroxyl groups in BRPP. The peak at 3447  $\text{cm}^{-1}$  can be assigned to the O–H stretching vibration. The peak at 1637  $\text{cm}^{-1}$  was assigned to the C=C stretching vibration. There are also two peaks at 1164  $\text{cm}^{-1}$  and 1019  $\text{cm}^{-1}$ , typical of C–O stretching vibrations, which confirm the presence of hydroxyl groups in rambutan peels. It is postulated that these functional groups participate in adsorption of the oppositely charged Basic Fuchsin molecules. However, the FTIR spectra of BRPP before and after adsorption are very similar to each other. This could be due to the limitations in the sensitivity of the instrument.

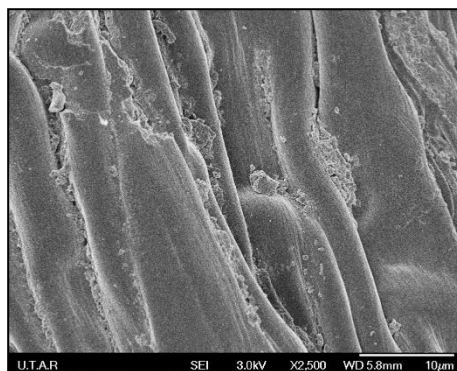
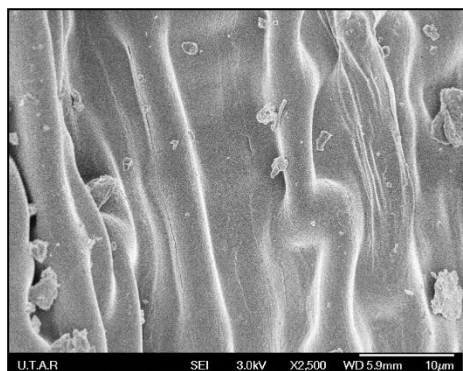


Fig. 2. SEM micrograph of BRPP before adsorption Fig. 3. SEM micrograph of BRPP after adsorption

The SEM micrographs of BRPP before and after adsorption are shown in Figs. 2, 3. It was found that the BRPP has a smooth surface and it is a non-porous material (Fig. 2). However, after the adsorption, apparently the surface of BRPP becomes more heavily loaded with the dye molecules (Fig. 3). Further confirmation can be performed by using atomic force microscopy (AFM).

### 3.2. EFFECT OF INITIAL CONCENTRATION AND CONTACT TIME

The percentage uptake of Basic Fuchsin decreased while the concentration of dye solution increased from 100  $\text{mg}/\text{dm}^3$  to 200  $\text{mg}/\text{dm}^3$  (Fig. 4). The adsorption rate of dye is very high at the first 30 min. Then the adsorption became slower and can be considered to reach equilibrium after 4 h. The high uptake rate at the beginning of the process is due to high availability of binding sites on the surface of BRPP at this stage. The binding sites were slowly occupied by the dye molecules and became saturated. Hence, the rate was decreased. A similar trend in the adsorption of Basic Fuchsin on  $\text{KMnO}_4$ -modified activated carbon has also been reported [13].

The kinetic study can be used to determine the adsorption mechanism of Basic Fuchsin onto BRPP. The experimental data was fitted into pseudo-first order kinetic

model by Lagergren [14] and pseudo-second order kinetic model by Ho and McKay [15]. According to Basha et al. [16] the pseudo-first order kinetic model defines the rate of intake of the binding sites to be proportional to the number of unoccupied binding sites. The linear equation of pseudo-first order model is expressed as follows:

$$\log(q_e - q_t) = \log q_e - \frac{k_1}{2.303t} \quad (2)$$

where:  $q_e$  is the amount of dye adsorbed at equilibrium, mg/g,  $q_t$  – the amount of dye adsorbed at a particular time, mg/g,  $k_1$  – rate constant of pseudo-first order reaction, 1/min,  $t$  – time, min.

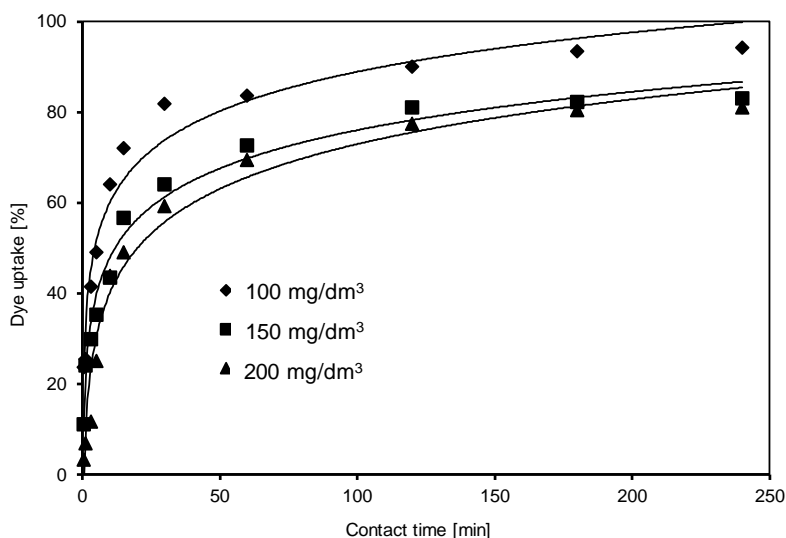


Fig. 4. Effect of initial dye concentrations and contact time on the percentage uptake of Basic Fuchsin by BRPP (2.0 g BRPP/dm<sup>3</sup>)

A linear plot of  $\log(q_e - q_t)$  vs. time for the adsorption of Basic Fuchsin onto BRPP at the concentration of 100, 150 and 200 mg/dm<sup>3</sup> is shown in Fig. 5. The rate constant  $k_1$  and the theoretical amount of dye adsorbed at equilibrium can be calculated from the slope and the y-intercept of the plot. Adsorption capacities and correlation coefficients based on pseudo first order kinetics are given in Table 1. As is seen, the theoretical value of  $q_e$  differed a lot from the experimental one. Hence, it can be concluded that the adsorption of Basic Fuchsin on BRPP does not fit to the pseudo-first order kinetic model.

The results from the effect of contact time were used to test the applicability of pseudo-second order kinetic model, which is based on the assumption that the rate limiting step may be due to chemisorption which associates with the valence forces through

the transferring between adsorbent and adsorbate [17]. The linear equation of this model is expressed as follows:

$$\frac{t}{q_t} = \frac{1}{h} + \frac{1}{q_e} t, \quad h = k_2 q_e^2 \quad (3)$$

where:  $t$  – time, min,  $q_e$  – amount of the dye adsorbed at the equilibrium, mg/g,  $q_t$  – amount of the dye adsorbed at a particular time, mg/g,  $h$  – initial rate of adsorption, mg/(g·min),  $k_2$  – rate constant of pseudo-second order kinetic model, g/(mg·min).

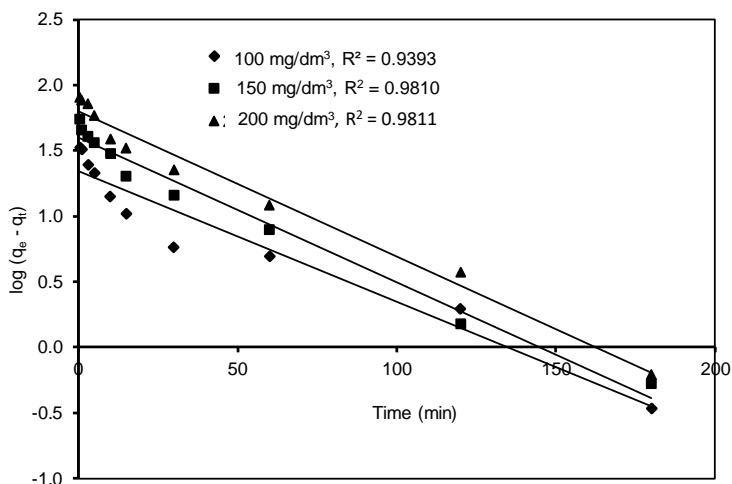


Fig. 5. Pseudo-first order kinetic plot for adsorption of Basic Fuchsin on BRPP (2.0 g BRPP/dm<sup>3</sup>)

Table 1

Pseudo-first order and pseudo-second order kinetic model parameters for various initial Basic Fuchsin concentrations at natural pH of dye solution 5.7 and adsorbent dose of 2 g/dm<sup>3</sup>

Initial concentration [mg/dm <sup>3</sup> ]	$q_{e, \text{exp}}$ [mg/g]	Pseudo-first order kinetic model			Pseudo-second order kinetic model			
		$q_{e, \text{cal}}$ [mg/g]	$k_1$ [1/min]	$R^2$	$q_{e, \text{cal}}$ [mg/g]	$k_2$ [g/(mg·min)]	$h$ [mg/(mg·min)]	$R^2$
100	44.1503	19.0596	0.02303	0.9393	44.6429	0.00498	9.9341	0.9994
150	63.4544	39.6734	0.02533	0.9811	64.9351	0.00229	9.6711	0.9991
200	84.2057	63.6063	0.02556	0.9810	89.2857	0.00087	6.9252	0.9991

A plot of  $t/q_t$  vs. time was based on the experimental results is shown in Fig. 6. The initial rate of adsorption can be calculated from the y-intercept of the plot. The calculated values were tabulated in Table 1. Based on Table 1, the correlation coefficient,

$R^2$  of each plot was found to be greater than 0.999 which is close to unity. The calculated and experimental values of  $q_e$  for various concentrations of Basic Fuchsin are very close which indicates that the adsorption of Basic Fuchsin on BRPP fits well the pseudo-second order kinetic model.

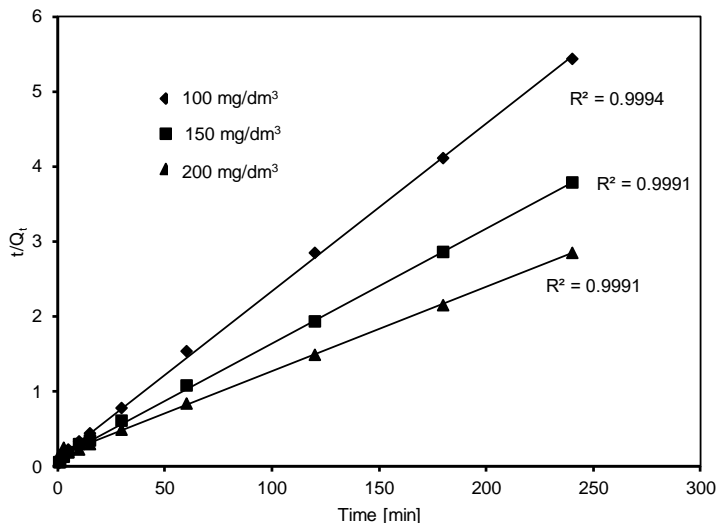


Fig. 6. Pseudo-second order kinetic plot for adsorption of Basic Fuchsin on BRPP (2.0 g BRPP/dm<sup>3</sup>)

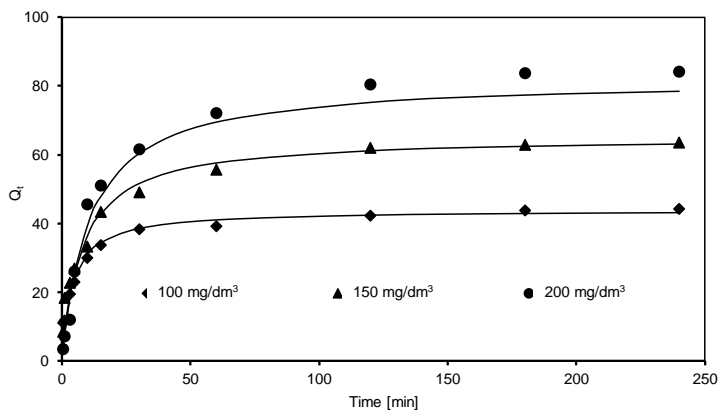


Fig. 7. Measured (points) and modelled (curves) pseudo-second order time profiles for Basic Fuchsin adsorption onto BRPP

By using the same model, the generalized predictive model for Basic Fuchsin adsorbed at any contact time and initial concentration within the given range is as follows:



$$q_t = \frac{tC_0}{0.1618C_0 - 6.2241 + (0.0035C_0 + 1.8091)t} \quad (4)$$

Experimental values and theoretical curves are shown in Fig. 7. It can be clearly seen that these results agree with each other with a slight deviation at 200 mg/dm<sup>3</sup>.

### 3.3. EFFECT OF pH

Plots of percentage uptake of the dye against pH are shown in Fig. 8. The uptake is low in acidic solutions and slowly increases upon increasing pH. A dramatic increase in percentage uptake of Basic Fuchsin was observed at pH 2–5 and only slightly increases at pH higher than 7. Similar observation has been reported for the effect of pH on adsorption of Basic Fuchsin on bottom ash and deoiled soya [18].

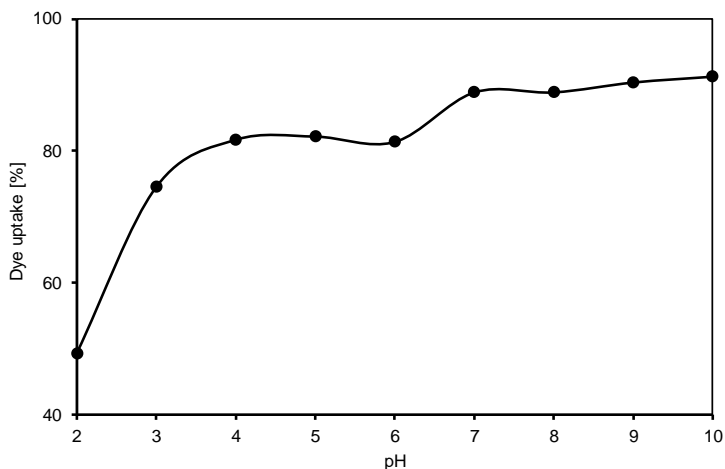


Fig. 8. Effect of pH of the solution on adsorption of Basic Fuchsin on BRPP (2.0 g BRPP/dm<sup>3</sup>, dye concentration 200 mg/dm<sup>3</sup>)

It has been suggested that at lower pH, carboxyl groups at the biosorbent surface which are responsible for binding the dye cations are predominantly protonated, therefore leading to a lower uptake of the dyes [19]. Besides, at low pH, hydrogen ions may also compete for the binding sites on BRPP and reduce their number for the dye molecules. Upon increasing pH, the amount of hydrogen ions decreases and there is less competition between the cationic Basic Fuchsin ion and hydrogen ions for the negatively binding sites on the surface of BRPP. Also, as the pH increased, adsorption became favourable due to the deprotonation of the carboxyl groups (–COO). This phenomenon favoured the sorption of positively charged dye due to electrostatic attraction. Hence, higher uptake of Basic Fuchsin by BRPP was observed at higher pH.

## 3.4. SORPTION ISOTHERM

Two common adsorption models which are Langmuir [20] and Freundlich [21] isotherms were applied to study the homogeneity and heterogeneity of the adsorbent surface and to evaluate the applicability of the adsorption process as a unit operation. Basically, the Langmuir isotherm corresponds to the monolayer adsorption process occurring at the specific homogeneous sites within the adsorbent with constant adsorption energy. The linear form of the equation is as follows:

$$\frac{1}{q_e} = \frac{1}{q_0} + \frac{1}{K_L q_0 C_e} \quad (5)$$

where:  $q_e$  is the amount of dye adsorbed onto BRPP at equilibrium, mg/g,  $q_0$  – maximum adsorption capacity, mg/g,  $K_L$  – a constant, dm<sup>3</sup>/mg,  $C_e$  – concentration of the dye in solution at equilibrium, mg/dm<sup>3</sup>. The constants for the Langmuir model of adsorption of Basic Fuchsin are presented in Table 2.

Table 2

Langmuir constants for the adsorption of Basic Fuchsin onto BRPP

Dye concentration [mg/dm <sup>3</sup> ]	$R_L$	$q_0$ [mg/g]	$K_L$ [dm <sup>3</sup> /mg]	$R^2$
100	0.08145	108.696	0.12026	0.9336
150	0.05156			
200	0.03844			
250	0.03159			
300	0.02625			

The adsorption capacities of Basic Fuchsin on various adsorbents are given in Table 3<sup>3</sup>. It is evident that rambutan peels demonstrated a large adsorption capacity towards Basic Fuchsin dye. Therefore, this waste material can be an efficient yet economical adsorbent for the removal of Basic Fuchsin from aqueous environment.

The Freundlich isotherm describes the adsorption process as physicochemical and multilayer adsorption with the interactions among the adsorbed molecules. It also implies that the adsorption occurs on the heterogeneous surface of the adsorbent. The linear equation of the Freundlich isotherm can be expressed as:

<sup>3</sup>Reported adsorption capacities are obtained under specific experimental conditions. For information, readers are encouraged to refer to original papers.

$$\log q_e = \log K_F + \frac{1}{n} \log C_e \quad (6)$$

where:  $q_e$  is the amount of dye adsorbed at equilibrium, mg/g,  $K_F$  – the Freundlich isotherm constant for adsorption capacity, mg/g,  $n$  – the Freundlich isotherm constant for intensity,  $C_e$  – concentration of dye adsorbed at equilibrium, mg/dm<sup>3</sup>.

Table 3

Adsorption capacities of Basic Fuchsin onto various adsorbents

Adsorbent	$q_0$ [mg/g]	Reference
Na-bentonite	147.49	[22]
Ca-bentonite	100.00	
KMnO <sub>4</sub> -modified activated carbon	270.27	[13]
Bottom ash	7.16	[18]
Deoiled soya	13.48	
Rambutan peel	108.70	this study

The calculated Freundlich isotherm constant for adsorption capacity,  $K_F$  and the Freundlich isotherm constant for intensity,  $n$  were: 22.842 and 2.7056, respectively. The heterogeneity factor,  $1/n$  is 0.3696 indicating that the adsorption of Basic Fuchsin on BRPP is favorable. The higher correlation coefficient,  $R^2$  of 0.9867 in the Freundlich isotherm indicated that the adsorption of Basic Fuchsin on BRPP is better explained by this isotherm which involved physiochemical and multilayer adsorption onto heterogeneous surface of BRPP.

#### 4. CONCLUSION

The FTIR analysis showed the presence of hydroxyl group on the surface of rambutan peels which may be possible binding sites for Basic Fuchsin. The effectiveness of adsorption of Basic Fuchsin on rambutan peel was influenced by few factors such as pH, contact time and initial concentration of dye solution. The experimental results indicate that the adsorption process attained equilibrium after 4 h and the percentage uptake of Basic Fuchsin increased upon decreasing initial concentration of the dye solution. The adsorption of Basic Fuchsin on rambutan peel was found to be higher in basic solution. The data obtained from the kinetic study showed that the adsorption of Basic Fuchsin fitted well with pseudo-second order kinetic model which is based on chemical sorption. Isotherm study showed that the adsorption process fitted better the Freundlich isotherm model as compared to the Langmuir equation.

The maximum adsorption capacity of rambutan peel towards Basic Fuchsin was determined to be 108.7 mg/g. As such, it can be concluded that the rambutan peel is a potential alternative low-cost adsorbent to substitute activated carbon on removal of Basic Fuchsin from aqueous environment.

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