Removal of Brilliant Green Dye from Aqueous Solution by using Banana leave as Adsorbent

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***Abstract*— Brilliant green (BG) dye is cationic dyes that are soluble in the water and commonly used for paper printing, veterinary medicine, textiles, cosmetic and food coloring. It is toxicity which affect to human health like irritation that causing vomiting, nausea and diarrhea. It also causes water pollution that directly affect the aquatic life by prevent light penetrating into the water. This study is focus to enhance the efficiency of removal brilliant green dye by using banana leaves as adsorbent and to determine the variables that affect the adsorption capacity. The banana leaves have been dried and grinded until become a powder. The parameters of the experiment are effect on the pH, effect of the initial dye concentration, effect of the contact time and effect of adsorbent dosage. The sample mixture of BG and banana leave were collected and analyzed by using UV-Vis spectrophotometer and FTIR instrument. The obtained data were analyzed to determine the percentage removal, adsorption capacity and adsorption isotherm. As a result, banana leave powder was hydroxyl group which functional group of alcohol. The optimum pH of solution was at pH 7 with the percentage removal of 98.98%. The solution reached equilibrium state in 120 minutes with adsorption capacity range of 9.86 mg/g to 49.37 mg/g. The optimum concentration of the percentage removal is at 97.33% to 98.73% at initial concentration of 50 to 250 mg/L. The percentage removal of the solution with different adsorbent dosage in the range of 0.5 to 2.5 g is 96.97% to 98.60%. Freundlich isotherm fitted the equilibrium data with correlation coefficient 0.995 which indicate multilayer adsorption. Therefore, banana leave has a potential to act as adsorbent in dye removal process and can be considered as low cost adsorbent material including banana leave waste.**

***Keywords*— Adsorbent, Adsorption capacity, Banana leave, Brilliant green dye, Isotherm**

I. INTRODUCTION

Water is one of the important sources for all living organisms to survive. However, there are many contaminants presence in the water that causes from the industries effluents such as organic dyes. Dyes are coloring agents that widely used in the industries of textiles, paper-making, printing, plastics, coating, food industries, cosmetics and mineral processing [5]. One of characteristic of dyes is it will completely soluble or partially soluble into the water. There are many negative impacts of the dyes to human, animal and environmental. Cancer, mutagenesis, teratogenicity and respiratory diseases are the effect that occurs from the dyes that harmful to human being [16]. The presences of dyes in the water cause the reduction of photosynthesis process by reducing the dissolved oxygen and disturb the aquatic life because of it prevent the light penetration [13]. Besides that, dyes is organic that has a different molecules structure which is non-biodegradable and dyes are difficult to treat.

Commonly, brilliant green dye is one type of dye that is used in the industry. It is category as cationic dye and mostly used for paper printing, textile dyeing, dermatological agent, veterinary medicine, intestinal parasite and fungus. According to [21], nitrogen oxides, carbon dioxides and sulfur oxides are hazardous product that produced from the heating process for decomposition of this dye. Dye disposed may also result in irritation to respiratory, nausea, vomiting and diarrhea. In this research study, brilliant green dye is use as simulated dye in the waste water. Therefore, there are many types of treatment that have been applied to treat the dye effluent into the water bodies.

The methods that have been applied to treat the dyes from the wastewater are flocculation, coagulation, oxidation, electrolysis, ion-exchange, photo catalytic-degradation, biomass, membrane separation and adsorption. Among these methods, adsorptions method are the most efficient method to treat the dyes that from different type of coloring material from the wastewater because it has a capability to removes large scale dyes and the adsorbent has a potential to be regenerate, recovery and recycling [3]. The adsorbent that used is cheaper and easy to get. Technically, adsorption method is economical and environmental friendly [12].

There are many types of waste product that has been recycle to act as adsorbent. Commonly, agricultural waste product has been used as adsorbent because it cheaper and no disposal problem [11]. In the previous study, clay material, red mud, fly ash, zeolites[10], and agricultural waste such as sawdust, , orange peel, guava leaf, and spent tea leaf have been used as alternative of adsorbent for removal of dyes .

Banana leaves are widely used in cooking and wrapping food which is plant based compound that known as polyphenols. Polyphenols is natural antioxidant that is free radical and can prevent from any disease. After using, it was discarded as waste and some of it remains in the cultivation area. It have been proven that, banana leaves have a capability in removal of methylene blue dye from aqueous solution [15]. In order to investigate the effectiveness of banana leaves as adsorbent, it was being study in the removal of brilliant green dye from aqueous solution and determines the variables that affect the adsorption capacity.

II. METHODOLOGY

A. Materials

Brilliant green dye (95% dye content) supplied by R&M Marketing (Essex, U.K), chemical formula $C_{27}H_{34}N_2O_4S$, Molecular weight 482.64 g/mol, was chosen as adsorbate in this study. Hydrochloric acid (HCl), Sodium Hydroxide (NaOH) and

distilled water were used as reagent throughout the experiment any data analysis that was carried out.

B. Preparation of Banana leaves as adsorbent

In this study, banana leaves waste was collected at cultivation area and were washed using distilled water to remove dust and impurities from its surface. The adsorbent was dried for 24 h at 60 °C inside the oven. Then, the dried leaves were grind and sieve to obtain the small particle size of powder and stored in the air tight container [11].

C *Preparation of Brilliant green dye solution as simulated dye wastewater*

Brilliant green dye was chosen as the simulate dye waste water. About 1 g of BG was dissolved in 1000 ml of distilled water to obtain the dye concentration of 1000 mg/L. Then, the solution was diluted to 50, 100,150,200 and 250 mg/L as the standard solution. The concentration of the solution was measured using UV-vis spectrophotometer at 623 nm [8].

D. Characterization

The banana leave powder was analyzed by Fourier Transform Infrared spectrophotometer in range 4000 to 400 cm^{-1} to identify the functional group on the surface of banana leave. The UV-vis spectra of standard curve BG were recorded at room temperature.

E. Batch Equilibrium Studies

Batch equilibrium studies were conducted to determine the efficiency of banana leaves powder for removal of Brilliant green dye from aqueous solution. The fixed amount 0.5 g of banana leave was put into each of conical flasks that contain BG solution of 50 to 250 mg/L and put into shaker for 1 h to 3 h to determine the equilibrium state. Then, the step was repeated with same concentration of BG dye solution and different adsorbent dosage. BG concentration was analyzed using UV-vis spectrophotometer at maximum wavelength λ_{max} = 623nm. The adsorption amount of BG on banana leave was calculated using following equation:

$$Q_t = \frac{\mathrm{Co}-\mathrm{Cf}}{\mathrm{w}} \mathrm{x\ V} \quad (1)$$

Where, Co and Cf are the initial and final concentration of BG dye (mg/L), V is the volume of the solution (L) and W is the weight of adsorbent (g).

The percentage removal of BG (%) was calculated using the equation:

$$\%\ \mathrm{Removal} = \frac{\mathrm{Co}-\mathrm{Cf}}{\mathrm{Co}} \mathrm{x}\ 100\% \quad (2)$$

The equilibrium data obtained have been used to analyze the isotherm model. Langmuir isotherm is one of the common isotherms that usually used for various adsorbate – adsorbent combinations for both liquid and gas phase adsorption. It is a monolayer adsorption. The isotherm can expressed as follow:

$$\mathrm{q_e} = \frac{\mathrm{q_m K_L C_e}}{1+\mathrm{K_L C_e}} \quad (3)$$

Where q_e (mg/g) is amount of adsorbate taken, q_m (mg/g) is the monolayer adsorption capacity (mg/g) to form a complete monolayer coverage on the surface bound, C_e (mg/L) is adsorbate concentration in solution at equilibrium and K_L is Langmuir constant related to affinity between adsorbate and adsorbent.

The Langmuir isotherm can arranged linearly by following equation:

$$\frac{1}{q_e} = \frac{1}{q_m K_L C_e} + \frac{1}{q_m} \quad (4)$$

The characteristics of Langmuir isotherm can be expressed in dimensionless constants separation factor for equilibrium parameter, R_L by following equation:

$$R_L = \frac{1}{1+K_L C_o} \quad (5)$$

Where, Co (mg/L) is the initial concentration of BG dye. The value of RL which is the light penetrating on the nature of the adsorption to be either unfavourable (R_L >1), favourable (0 < R_L < 1), linear (RL = 1) and irreversible (R_L = 0).

Freundlich isotherm is a multilayer sorption which is commonly used to describe heterogeneous system. The isotherm can be expressed by the following equation:

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

Where, K_F (mg/g)$(mg/L^{)-1/n}$ sorption capacity, n is constant related to sorption capacity and sorption intensity.

III. RESULTS AND DISCUSSION

A. Characterization of Banana leave

Fourier Transform Infrared Spectrophotometer (FTIR) device had been used to determine the functional groups that exist in the banana leave. The FTIR spectrum of banana leave powder was illustrate in the figure 1. The FTIR spectrum obtained a peak at 3280.87 cm^{-1} corresponds to the absorption of hydroxyl group which was O-H stretching with functional group of alcohols and phenols. The adsorption of brilliant green onto banana leaves powder occurred because of the presence of the structure of cellulose, hemicelluloses and lignin in the functional group [20]. The peak located at 2917.09 and 2849.17 cm^{-1} were assigned to C-H stretching was present in alkane group. The peak at 1734.85 cm^{-1} was characteristics of carbonyl group with C=O stretching which the interaction between carbonyl group of adsorbent with dye solution was occurred [15]. At the peak of 1601.96 cm^{-1} there was N-H bend which in the functional group amine. Lastly, the peak that observed at the surface of banana leaves was at 1030.84 cm^{-1} that were aliphatic amine with C-N stretch bond exists.

B. Effect of pH on dye removal

In this study, the effect of different pH value on the removal of brilliant green dye from the aqueous solution was studied. The values of pH in the dye solutions were adjusted by using 0.1 M hydrochloric acid solution and 0.1 M sodium hydroxide solution within the pH range between 2 to 11 at initial dye concentration of 50 mg/L and 0.5 g of banana leave powder.

It was observed that the removal of brilliant green dye was increased significantly as the pH value increased. The percentages removal and amount of dye adsorbed at pH 2,4,7,9 and 11 was obtained in table 1. Previous finding proved that, the repulsive force between adsorbate and adsorbent decreased in the higher pH value of dye solution resulting in increased of adsorption capacity of the dye [19]. The interaction between dye solution and biosorbent was increase as the pH of the solution increased the number of hydroxyl group that result in the increasing the negatively charge site.

The figure 2 indicates the percentage removal of the dye at different pH value. It can be seen that the percentage removal was higher at pH 7 which was 98.98 %. Then, adsorption process remains almost constant in the pH ranges from 7 to 11. Based on previous finding, the brilliant green dye solution concentration will decrease in the natural acidic pH solution and the adsorption process was higher at pH 7 which was higher sorption efficiency with 97 % of dye removal [14]. In conclusion, the optimum pH for removal of brilliant green dye from aqueous solution was 7.

Table 1: Data collected for different pH

pH	Percentage Removal (%)	Adsorption capacity (mg/g)
2	94.58	9.4576
4	95.66	9.5658
7	98.98	9.8982
9	98.49	9.8490
11	98.34	9.8343

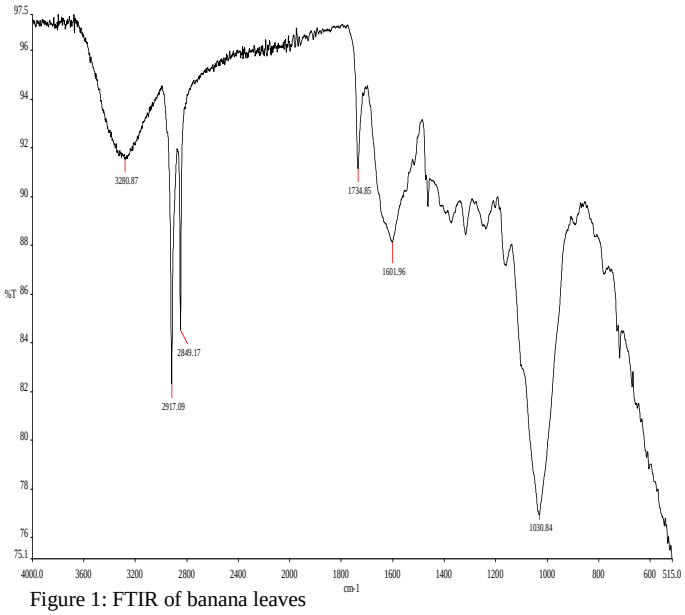

Figure 1: FTIR of banana leaves

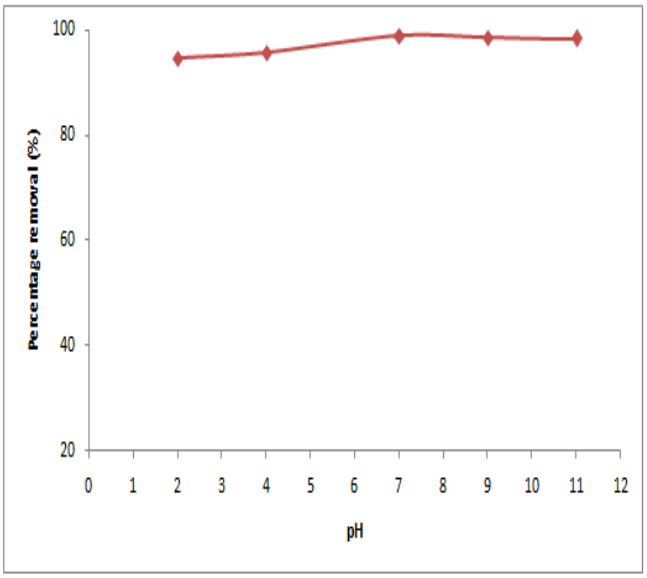

Figure 2: Percentage removal of different pH

C. Effect of contact time on dye removal

The effect of contact time on percentage of dye removal and adsorption capacity of brilliant green dye was studied at initial concentration of 50, 100, 150, 200 and 250 mg/L with fixed amount of adsorbent dosage of 0.5 g of banana leaves powder. Based on the data obtained, the percentage removal of dye was increased as the time interval increased.

The adsorption capacity at different initial concentration of brilliant green dye is shown in figure 3. The adsorption capacity has been observed to increase quickly at contact time of 60 minutes. This indicates that the adsorption process occurs rapidly. The adsorption process rapidly occurs at the initial contact time where there were available active sites at the surface of the adsorbent. Then, it gradually increases at contact time of 90 minutes and reach to the equilibrium at 120 minutes. Table 2 shows the adsorption capacity for the effect of contact time. The adsorption capacity increases from 9.86 mg/g to 49.37mg/g from 50 to 250 mg/L of initial dye concentration at equilibrium state.

The adsorption active site was limited when the concentration of brilliant green dye increases and takes a longer time to reach the equilibrium state [20]. According to the previous research, the adsorption capacity increases quickly at the contact time of 20 minutes for the removal of brilliant green dye using cactus fruit peel and reaches to the equilibrium after 250 minutes at initial brilliant green dye concentration of 25, 50, 200 and 500 mg/L [8] In conclusion, the adsorption capacity increases as the contact time increases where there were more dye can adsorbed onto the surface of the adsorbent.

Table 2: Data collected for adsorption capacity for 180 min

Time(min)	Initial dye concentration (mg/L) / Adsorption capacity (mg/g)				
	50	100	150	200	250
60	9.74	19.60	29.52	39.35	48.00
90	9.84	19.70	29.58	39.48	49.19
120	9.86	19.74	29.64	39.52	49.37
150	9.87	19.77	29.72	39.70	49.46
180	9.87	19.81	29.76	39.64	49.46

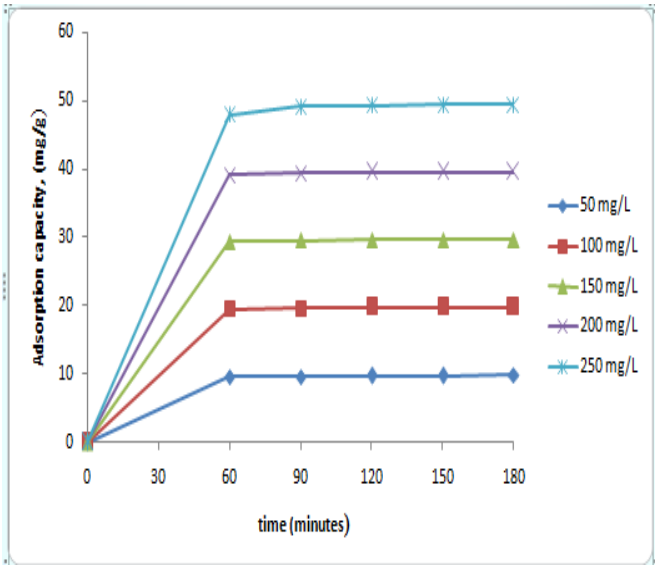

Figure 3: Effect of contact time on adsorption capacity

D. Effect of initial dye concentration on dye removal

The effect of initial dye concentration was conducted to evaluate the influences of initial brilliant green dye concentration on the adsorption capacity and percentage removal of dye from the aqueous solution. It was conducted with fixed amount of adsorbent dosage of 0.5 g with various concentrations of 50, 100, 150, 200 and 250 mg/L of brilliant green dye solution.

It can be observed from figure 4, that the percentage of dye removal was increased from 97.33% to 98.73% as the concentration of brilliant green dye increased from 50 to 250 mg/L at the equilibrium of 120 minutes. It also can be inferred from the figure 5 that the adsorption capacity increases with increasing the initial dye concentration from 9.86 to 49.37 mg/g. Table 3 shows the data collected for initial dye concentration. According to [6], the brilliant green dye molecules diffused onto banana leaves powder that result in higher concentration gradient between the brilliant green dye molecules. The increasing in the percentage removal of dye as the initial concentration of brilliant green dye increases. It was proved by previous research where the percentage removal of dye increases from 66.2% to 96% at initial concentration of brilliant green dye from 1 to 20 mg/L [7]. The number of collision between dye molecule and adsorption increased as the initial concentration of brilliant green dye increased will intensify the adsorption process However, based on the result obtained the increasing in initial dye concentration was not significantly affect the percentage removal. This might be happen due to the saturated surface of the adsorbent that cause the adsorption process of brilliant green dye onto banana leave not further occurred.

Table 3: Data collected for initial dye concentration

Initial dye concentration (mg/L)	Percentage removal (%)	Adsorption capacity (mg/g)
50	97.33	9.86
100	98.72	19.74
150	98.80	29.64
200	98.81	39.52
250	98.73	49.37

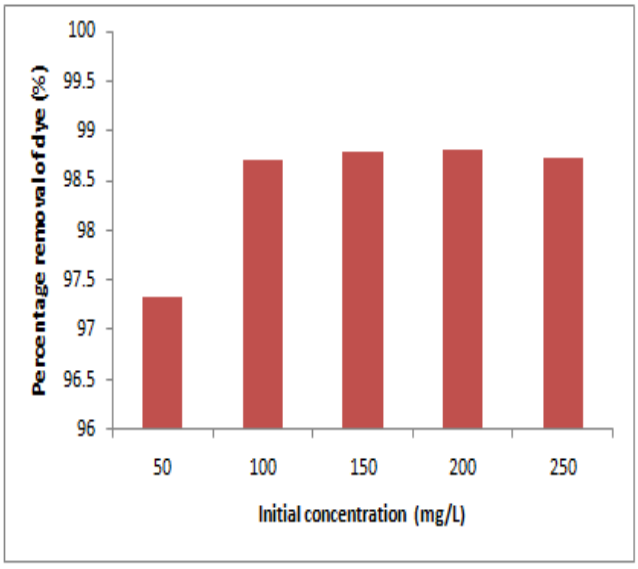

Figure 4: Percentage removal of different initial concentration

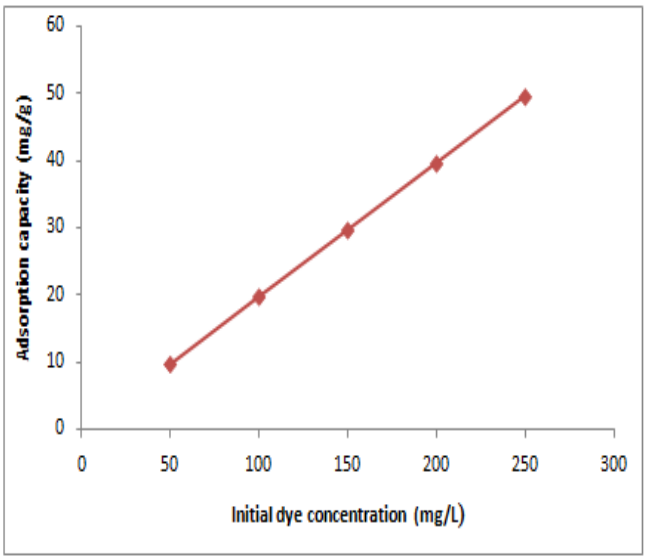

Figure 5: Effect of initial concentration on adsorption capacity

E. Effect of adsorbent dosage on dye removal

The effect of adsorbent dosage was carried out by varying the amount of banana leaves powder from 0.5 g to 2.5 g with 50 mg/L of initial dye concentration to evaluate the adsorption capacity and percentage of dye removal from the aqueous solution. Table 4 shows. From figure 6, it has been observed that the percentage removal of dye increased as the amount of banana leaves powder increased. According to the previous research, the percentage removal of dye was higher when there were higher quantities of adsorbent due to the presence of higher adsorption sites that lead to a better adsorption process [4].

However, the amount adsorption capacity was inversely proportional with adsorbent dosage. Figure 7 show the effect of the adsorbent dosage on adsorption capacity. It can be seen that, the adsorption capacity decreased from 9.6972 mg/g to 1.9721 mg/g as the adsorbent dosage increased. The adsorption sites remain unsaturated during the adsorption process as the amount of adsorbed dye decreased in the higher quantities of adsorbent dosage [1]. Besides that, the adsorbent particles were aggregate to each other in the higher amount of adsorbent dosage that results in smaller adsorption capacity and decreased of the total surface area of the dye adsorbed onto the adsorbent.

Therefore, the percentage removal of brilliant green dye from aqueous solution was increased due to the increased of adsorbent dosage at the optimum adsorbent dosage of 2.5 g with 98.60 % due to the higher number in sorption site. While, the adsorption capacity was inversely relationship with adsorbent dosage which it was decreased as the adsorbent dosage increased result in decreased in the number of active site in the adsorption process.

Table 4: Data collected for adsorbent dosage

Adsorbent dosage (g)	Percentage removal (%)	Adsorption capacity (mg/g)
0.5	96.97	9.6720
1.0	97.19	4.8786
1.5	98.13	3.2709
2.0	98.26	2.4566
2.5	98.60	1.9721

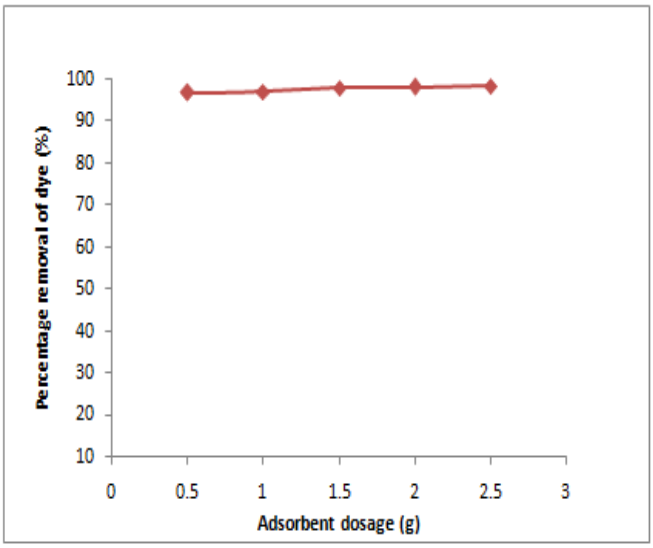

Figure 6: Percentage removal for different adsorbent dosage

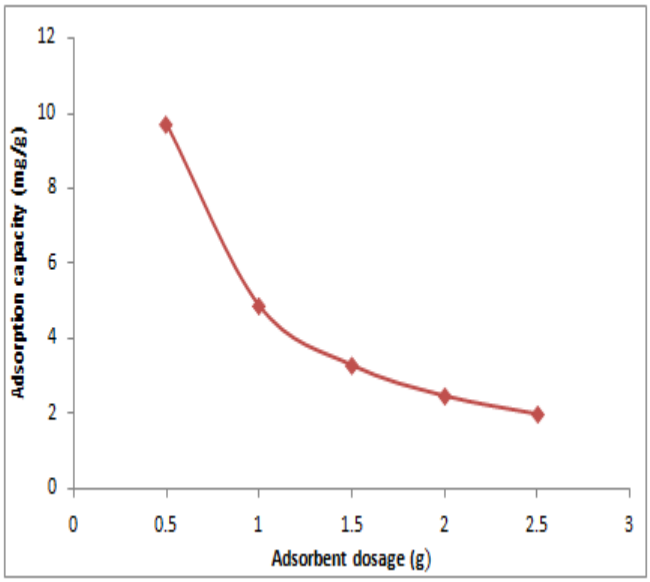

Figure 7: Effect of adsorbent dosage on adsorption capacity

F. Adsorption isotherm

There are a relation between concentration of adsorbate on solid phase and liquid phase with the isotherm equilibrium [17]. In this study, Langmuir isotherms and Freundlich isotherm were applied to determine the equilibrium of brilliant green dye adsorption. The best fit of this adsorption isotherm was identified by the correlation coefficient, R^2 value. Figure 8 and 9 shows the graph profile of Langmuir and Freundlich isotherm.

It can be seen that, Freundlich isotherm has the best fit correlations coefficient value of R^2 which was 0.995 for the adsorption of brilliant green dye. There were Van der waal forces between the adsorbent surface and adsorbate occurred in the adsorption process because of the negative charge at the adsorbent surface which affected by the increases of the value Kf and n [18]. Table 5 shows the data collected for Langmuir and Freundlich isotherm. For Langmuir isotherm, the correlation coefficient R^2 was 0.715 shows where the equilibrium of brilliant green adsorption happens. The data obtained shows that, the value of throw light R_L was -0.2247. According to previous research, it shows that brilliant green adsorption onto red clay fitted to Freundlich isotherm with R^2 value of 0.997[14]. Thus, banana leaves can be effectively to remove dye from effluents.

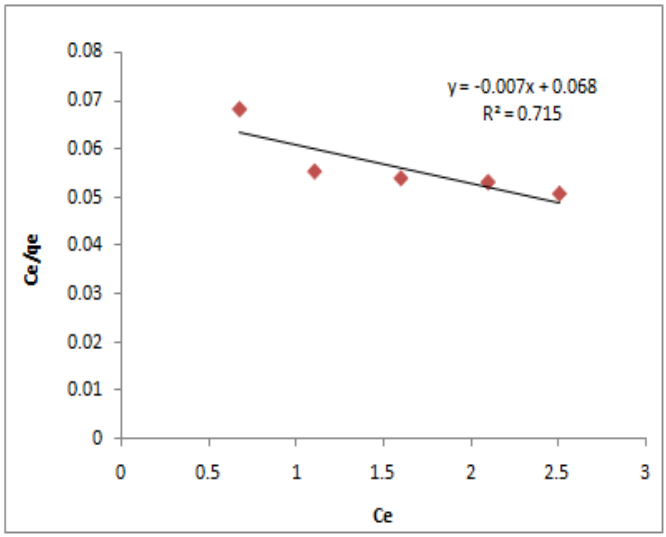

Figure 8: Langmuir plot

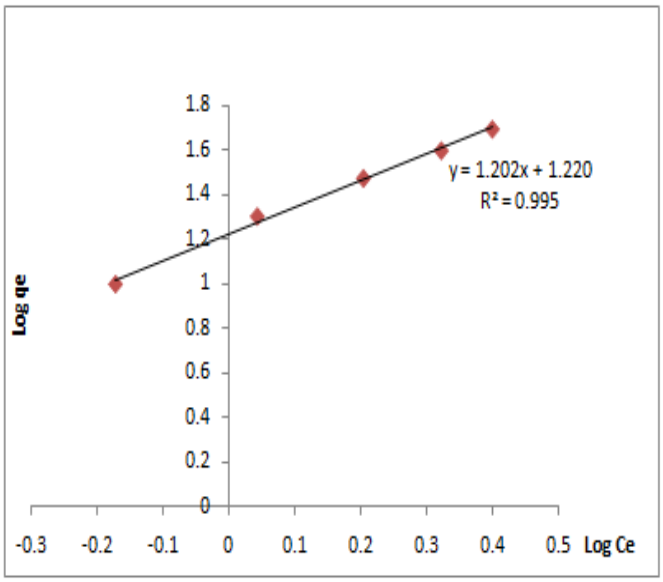

Figure 9: Freundlich isotherm

Table 5: Isotherm parameters for adsorption of Brilliant green dye onto banana leave

Isotherm	**Parameters**	**Values**	**R^2**
Langmuir	q_m K_L R_L	-142.86 -0.1029 -0.2247	0.715
Freundlich	n K_F	0.832 16.60	0.995

IV. CONCLUSION

Removal of brilliant green dye from aqueous solution using banana leave as adsorbent was investigated. Banana leave was characterized by the FTIR spectra which indicated the hydroxyl group where it is alcohol functional group that shows there are adsorption process occurred. The equilibrium time for adsorption brilliant green onto banana leave was found to be 120 minutes. The optimum pH of brilliant green dye was 7 with percentage removal of 98.98% and no significant change occurs above pH 7. This present study was revealed that the optimum dosage for banana leave to adsorb brilliant green was 2.5 g. The adsorption isotherm was well fitted by the Freundlich isotherm with correlation coefficient, R^2 0.995, which indicated adsorption occurred on the

multilayer. In order to improve the better result of removal the brilliant green dye in term of the effect of initial dye, study the effect of temperature and presence of catalyst to optimized the result. Last but not least, banana leave adsorbent has good adsorption capacity, good reusability and effective adsorbent for removal of brilliant green dye from aqueous solution. In directly, being a waste product, banana leave as adsorbent can resolve waste management.

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