



## REMOVAL OF BISMARCK BROWN R DYE FROM AQUEOUS SOLUTION BY *LAPLAP PURPUREUS* PLANT STEMS UTILIZED AS BIOSORBENT

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### ABSTRACT

Adsorption of dye utilizing biosorbent is an unusual method to remove the dye from industrial wastewater. Biosorbent arranged from *Laplap Purpureus* Stem Powder (LPSP) have been utilized as an adsorbent for the removal of Bismark Brown R dye from the aqueous solution. The biosorbent matter was analyzed by Scanning Electron Microscope (SEM) image and Fourier Transfer Infra Red (FTIR) spectroscopy. The batch adsorption experiment studies were carried out to evaluate various adsorbing effect of experimental parameters such as effect of pH, adsorbent dosage, equilibrium time and initial dye concentration. The data obtained from the experimental values were analyzed by Langmuir and Freundlich isotherm model. The adsorption kinetic studies were also analyzed by pseudo first order and pseudo second order models. The result reveals that the removal efficiency of biosorbent (LPSP) is very better for the Bismark Brown R dye from made up aqueous solution.

**Keywords:** Adsorption, *Laplap purpureus*, Biosorbent, Bismark Brown R, Isotherms, Kinetics

### 1. INTRODUCTION

Environmental pollution arises due to the release of industrial effluents are a main distress in growing countries. Generally, unused or partially used industrial effluents are being discharged directly to the natural ecosystem. This industrial effluents act as chief water pollutants. The most of the important category of pollutants derived from dyes, which are organic in nature and they are in the industrial effluents discharged from various industries like food, pharmaceutical, textiles, cosmetics, paper, leather, rubber and plastics. Man-made dyes are being produced on large amount than the natural dyes and are regularly used in textile industries. Due to their wide-ranging of applications, the man-made dyes can cause serious environmental problems and severe health risks. The dyes may be discharged into the water bodies is toxic to aquatic species as well as human beings, due to the presence of an aromatic or heavy metal in their structure. The contamination of dye in water is causes many healthiness troubles such as nausea, hemorrhage, ulceration and etc [1-2]. The various dye removal methods are using for purification of industrial effluents including oxidation, coagulation, floatation,

adsorption, membrane filtration and biological treatment. In the above all the methods the adsorption technique appears to have significant possible for the removal of dye from textile industrial effluents [3]. Adsorption technique is better in simplicity of design, low cost, easy to handle and insensitivity to toxic material. A huge number of suitable adsorbents such as activated carbon, polymeric resins and a variety of low cost adsorbents like non modified or modified cellulosic biomass, bacterial biomass have been studied [4]. Detection of a potential dye adsorbent should be in good concurrence with its dye binding capability, its requirements and limits with respect to environmental factors. The conversion of agricultural wastes into valuable materials with negligible generating pollutants is a prominent challenge and suggested for an industrial sustainable development in order to defend the environment [4]. Hence, biosorption is a promising technique for pollutants removal from their solutions. The materials of biological sources are used as a sorbents to remove dyes from the aqueous solutions. These 'biosorbents' have a variety of functional groups that can complex with the dyes. Researcher in the field of

biosorption proposes a number of advantages more than other techniques like the material can be found easily as by-products, unreacted or waste materials can be reused, no need of cost for growth media, simple methods, requires low cost investment, the process is rapid, easy to operate, greener synthesis and free from the physiological restraint of living cells [5]. Nowadays a huge number of biosorbents have been utilized to removal of dyes, such as *Daucus carota* [6], *Kappaphycus alvarezii*, *Gracilaria salicornia*, *Gracilaria edulis* [7], *Solanum tuberosum* plant waste [8], *Haloxylon recurvum* stem [9], *Cucumis sativus* fruit peel [10], Coconut coir dust [11], wood apple shell [12], *Casuarina equisetifolia* Needle [13], Bengal gram seed husk [14] Saw dust [15]. The effectiveness of adsorption process mostly depends on investment cost and removal ability of adsorbents used. Today agricultural waste materials are getting much more consideration as adsorbents due to its low cost and wide range of availability for the removal of dyes from textile industrial effluents. The objectives of present study were to evaluate the sorption characteristics of *Laplap purpureus* stem powder (LPSP), under suitable conditions used for the removal of dye from aqueous solution.

## 2. MATERIALS AND METHODS

### 2.1. Preparation of adsorbent

The *Laplap purpureus* plant stems were received from locality home garden later than harvest at Sevvaypatti village, Karambakudi (T.K), Pudukkottai (D.T), Tamil Nadu -614 614, India. The stems were chopped into small segments, systematically washed with tap water to eliminate dirt and then further washed with distilled water and after then dried in direct sun light for five days. The dried *Laplap purpureus* plant stems were ground as very fine powder in a domestic grinder and sieving to separate the particles size of <90  $\mu\text{m}$  by using manual experimental (Jayant Test Sieves) sieves. These separated *Laplap purpureus* stem powdered (LPSP) particles were kept in good conditioned air tight plastic bottle for use in adsorption studies.

### 2.2. Preparation of Adsorbate

Bismark Brown R dye having molecular formula  $\text{C}_{21}\text{H}_{26}\text{Cl}_2\text{N}_8$  was selected as the adsorbate. The Himedia grade of Bismark Brown R dye was used. The concentration of 1g/L stock solution of dye was prepared by precisely weighed Bismark Brown R dye dissolving in double distilled water. The experimental dye solution was obtained by diluting the stock solution in precise proportions to required initial dye concentrations. 4-[5-

(2,4-Diamino-5-methylphenyl) diazenyl-2-methylphenyl] diazenyl-6-methylbenzol-1, 3-diamine is the IUPAC name of the Bismark brown R dye. The structure of Bismark Brown R is shown in fig. 1. [16-17].

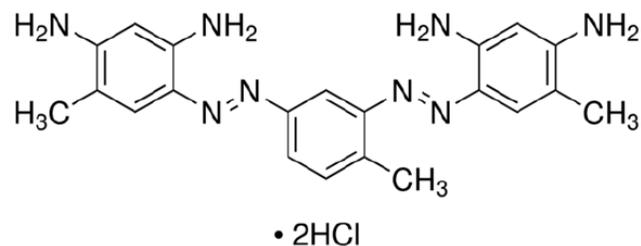


Fig. 1: Structure of Bismark Brown R Dye

## 2.3. Data Analyzing Methods

### 2.3.1. Adsorption Isotherm Studies

Adsorption isotherm is one of the most important physicochemical models in the description of adsorption process. It explains that how the adsorbent interacts by the adsorbate. Therefore it is essential in optimal the use of the adsorbent. The experimental data were tested by the familiar isotherm equations namely, Langmuir and Freundlich models.

#### 2.3.1.1. Langmuir Isotherm

Langmuir isotherm takes postulation that the sorption presents at particular homogeneous sites within the adsorbent. The common term for Langmuir equation is,

$$q_e = b q_{\max} C_e / (1 + b C_e)$$

The linear structure of isotherm equation has written as,

$$1/q_e = (1/bq_{\max}) (1/C_e) + (1/q_{\max})$$

$q_{\max}$  = Saturation capacity of the adsorbent by maximum dye uptake,  $b$  = Energy of adsorption variable  $C_e$  and  $q_e$  respectively.

#### 2.3.1.2. Freundlich Isotherm

Freundlich isotherm is an experimental equation based on a heterogeneous surface. The common form of Freundlich equation is,

$$q_e = k_f C_e^{1/n}$$

and the linear form is,  $\log q_e = \log k_f + 1/n \log C_e$  where the intercept  $\log k_f$  is a measure of adsorption capacity and slope  $1/n$  is the intensity of adsorption.

### 2.3.2. Kinetics Studies

Kinetic models were used to analyze the experimental data to explore about the potential rate controlling step and the mechanism of the adsorption such as the chemical reaction and mass transfer process. The transiting perform of batch adsorption process was studied by using

pseudo first order and pseudo second order kinetic models.

2.3.2.1. Pseudo First order

The possibility of adsorption data following Lagergren pseudo-first-order kinetic is given by the linearized eq.

$$\log (q_e - q_t) = \log q_e - (k_1/2.303)t$$

Where  $q_e$  (mg/g) and  $q_t$  (mg/g) refers to the amount of dye adsorbed per unit weight of adsorbent at equilibrium and at time  $t$ ,  $k_1$  is the rate constant of adsorption [18].

2.3.2.2. Pseudo Second order

This adsorption kinetic model equation was developed by 'Ho', studied to give details about the sorption capacity, the pseudo second order model can be written as,

$$t/q_t = 1/k_2 q_e^2 + 1/q_e t$$

Where  $t$  is the constant time (min),  $q_e$  (mg/g) and  $q_t$  (mg/g) are the amounts of dye adsorbed at equilibrium and at any time,  $t$  [19].

2.3.2.3. Batch Experiment

Adsorption experiments of dye solution were carried by treated with 500 mg of adsorbent dose and introducing 50 mL of stock solution of dye. The various parameters were performed like effect of pH, effect of adsorbent dose, effect of contact time and effect of initial dye concentration. After that the preferred times of treatment, the experimental samples were drained to remove the adsorbent and then adsorption progress was observed by using lambda 35 UV-visible Spectrophotometer at 420 nm is the wave length for maximum absorbance of  $\lambda_{max}$  for Bismark Brown R dye.

3. RESULTS AND DISCUSSION

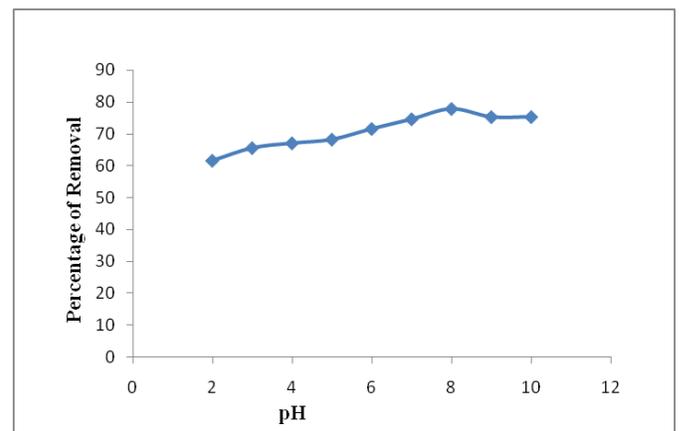
3.1. Effect of pH for dye solution onto adsorption

The pH of the aqueous solution of dye is the evidently main parameter for controlled the adsorption process. The experiments were completed with range of pH from 2 to 10, temperature is 30°C, contact time is 50 minutes, agitation speed is 360 rpm, initial dye concentration is 200 mg/L and the adsorbent dose is 300 mg. The results of the experiment are shown in the table 1. The graph has drawn between pH and BBR dye uptake is shown in the fig. 2. The figure shows that the biosorbent contains polymers with more number of functional group; therefore the net charge on the biosorbent is more pH dependent [20]. When increasing the pH of the system while the number of negatively charged sites (OH) also increases on biosorbent, due to increase in the hydroxyl

ion concentration where as the number of positively charged sites ( $H^+$ ) also decreases [21]. Hence, at higher pH is most favors for the uptake of positively charged (cationic) dye due to the surface of the adsorbent gets more negatively charged by losing protons. Here the dye uptake occurs due to increased electrostatic force of attraction between surface of dye and adsorbent [22]. Therefore, dye uptake at lower pH decreases due to less number of negatively charged sites at the LPSP surface. The lower sorption of BBR dye at lower pH was maybe due to the presence of the large number of  $H^+$  ions competing with the cationic groups of the dye on sorption sites [23]. The maximum sorption was observed at pH 8 for BBR (cationic or positively charged dye) dye. The decrease in the biosorption of BBR dye after pH was insignificant.

**Table 1: Effect of pH on dye uptake, Time 50 min, Biosorbent dose 300 mg, Volume of the solution 50 mL, Initial dye concentration 200 mg/L and Temperature 30°C**

pH	Percentage of Removal
2	61.50
3	65.52
4	67.03
5	68.20
6	71.57
7	74.63
8	77.91
9	75.35
10	75.35



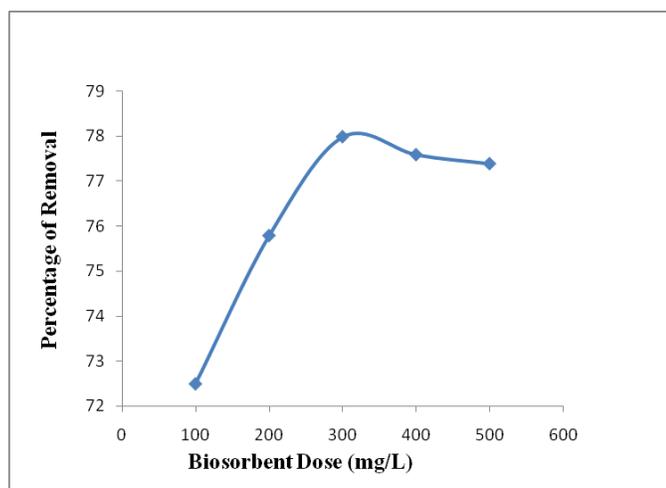
**Fig. 2: Effect of pH on dye uptake, Time 50 min, Biosorbent dose 300 mg, Volume of the solution 50 mL, Initial dye concentration 200 mg/L and Temperature 30°C**

### 3.2. Effect of biosorbent dose for dye solution onto adsorption

The adsorbent dose was also studied for the removal of BBR dye from aqueous solution. The experiment was carried out with adsorbent dose is varied from 100 to 500 mg with other parameters are constant such as pH 8, initial dye concentration is 200 mg/L, temperature is 30°C, contact time is 50 minutes, agitation speed is 360 rpm. The results of the experiment are shown in table - 2. The effect of biosorbent dosage for the removal of dye is shown in fig. 3. The figure representing that adsorption was mostly complete with biosorbent from 100 to 300 mg. The adsorption of dye uptake increase with increase in dose of the adsorbent, because of increases the adsorption surface area and availability of adsorption sites [24]. Moreover, above the 300 mg of the adsorbent dose weight, did not show any significant for the removal of dye, hence, 300 mg biosorbent dose was preferred for following experiments.

**Table 2: Effect of Biosorbent dose on dye uptake, Time 50 min, pH 8, Volume of Solution 50 mL, Initial dye concentration 200 mg/L and Temperature 30<sup>o</sup> C**

Adsorbent dose (mg)	Percentage of Removal
100	72.49
200	75.79
300	77.99
400	77.59
500	77.39



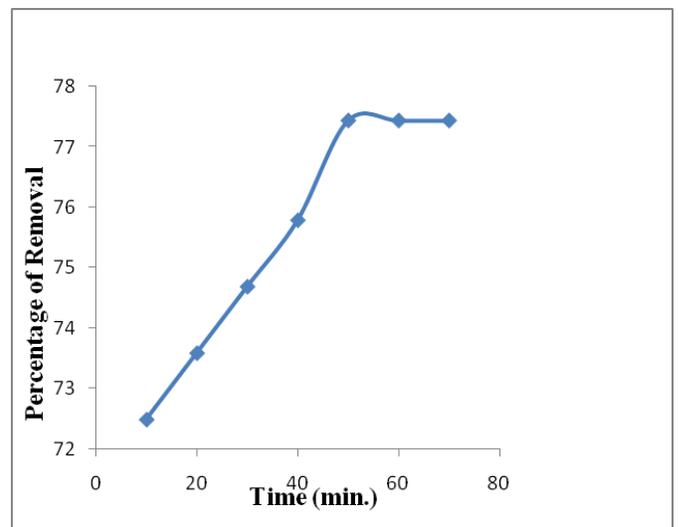
**Fig.3: Effect of Biosorbent dose on dye uptake, Time 50 min, pH 8, Volume of Solution 50 mL, Initial dye concentration 200 mg/L and Temperature 30<sup>o</sup> C**

### 3.3. Effect of contact time for dye solution onto adsorption

The most essential factor in batch adsorption studies are the effect of contact time. In this study all of the parameter other than contact time 10 to 70 minutes, including temperature is 30°C, adsorbent dose is 300 mg, pH is 8, initial dye concentration is 200 mg/L and agitation speed is 360 rpm were kept permanent. The table 3 is exhibits the experimental data and the fig. 4 shows efficiency of dye adsorption by effect of contact time.

**Table 3: Effect of contact time on dye uptake, pH 8, volume of solution 50 mL, Biosorbent dose 300 mg, Initial dye concentration 200 mg/L, Temperature 30<sup>o</sup> C**

Time in(min)	Percentage of Removal
10	72.49
20	73.59
30	74.69
40	75.79
50	77.44
60	77.44
70	77.44



**Fig. 4: Effect of contact time on dye uptake, pH 8, volume of Solution 50 mL, Biosorbent dose 300 mg, Initial dye concentration 200 mg/L, Temperature 30<sup>o</sup> C**

The time variant graph exhibits that in the preliminary stage of the dye adsorption is rapidly removed but while it reaches at equilibrium, it slows downward gradually. This is due to during the initial stage of adsorption process the availability of vacant surface sites and then a

certain time period the vacant sites of the adsorbent get occupied by dye molecules, as a result to form a repulsive force between the adsorbate on the adsorbent surface and bulk phases. The adsorption was takes place up to 50 minutes after that the equilibrium attainment, the percentage of adsorption of dye did not show any appreciable change with respect to time. This suggests that after equilibrium is attained, further treatment does not provide more removal [15]. In batch adsorption, the removal rate of the adsorbate in aqueous solution is mainly controlled by transport of dye molecules from the surrounding sites to the interior sites of the adsorbent particles [25]. The figure exhibited that a contact time of 50 minutes was sufficient to reach equilibrium and the adsorption no change with further increasing contact time, hence, the contact time has been preferred as 50 minutes for the continuous experiment.

**3.4.Effect of Initial dye concentration for dye solution onto adsorption**

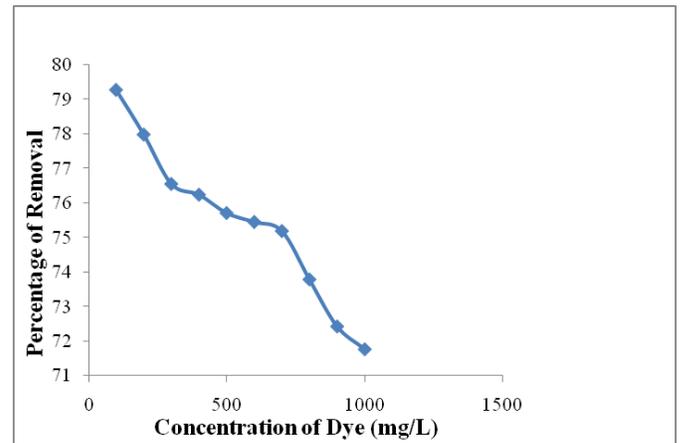
The experiment were carried out with a different concentrations of dye solution from 100 to 1000 mg/L and temperature is 30°C, adsorbent dose is 300 mg, pH is 8, contact time is 50 minutes, agitation speed is 360 rpm. The experimental data are shown in table 4 and the graphs were drawn between initial dye concentrations and dye uptake is shown in figure 5.

**Table 4: Effect of Initial dye concentration on dye uptake, Time 50 min, pH 8, Volume of Solution 50 mL, Biosorbent dose 300mg and Temperature 30°C**

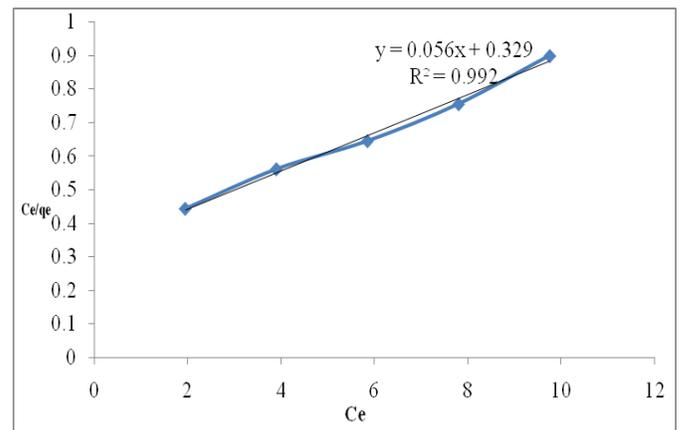
Initial dye concentration (ppm)	Percentage of Removal
100	79.28
200	77.99
300	76.56
400	76.25
500	75.72
600	75.46
700	75.20
800	73.80
900	72.44
1000	71.78

The figure exhibits that the effect of initial dye concentration is highly depend upon the immediate relation between the available binding sites on an adsorbent surface and the concentration of dye [26].

Usually the dye removal percentage is decreases with increase in concentration of initial dye, which may be reason for the saturation of adsorption sites in the adsorbent surface and the adsorption ability increased with an increase in concentration of the dye. In low concentration there will be vacant active sites on surface of the adsorption and when the initial dye concentration increases, the active sites not to be enough for adsorption of the dye molecules [27].



**Fig. 5: Effect of Initial dye concentration on dye uptake, Time 50 min, pH 8, Volume of Solution 50 mL, biosorbent dose 300 mg and Temperature 30°C**

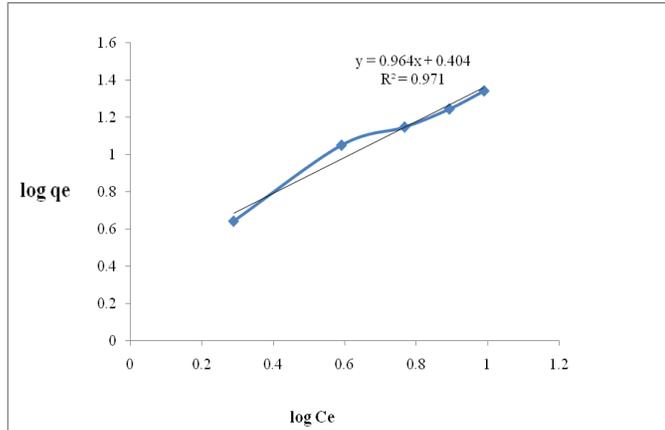


**Fig. 6: Langmuir isotherm plot of Bismark Brown R dye using *Laplap purpureus* plant stem powder**

**3.5.Adsorption Isotherm Models**

The constant  $q_{max}$  and  $b$  are the characters of Langmuir isotherm and can be determined from the Langmuir equation, a plot of  $1/q_e$  Vs  $1/C_e$  gives a straight line of a slope  $(1/q_{max})$  and intercepts  $1/q_{max}$ . So that the data matching with Langmuir isotherm. The linearity of the plot represents the application of Langmuir equation is supporting monolayer creation on the surface of the

adsorption. The variable from Freundlich equation,  $q_e$  and  $C_e$  are dye adsorbed and the equal dye concentration in solution. Langmuir and Freundlich plots were here using the tables 5 and their plots were shown in fig. 6 and fig. 7 respectively.

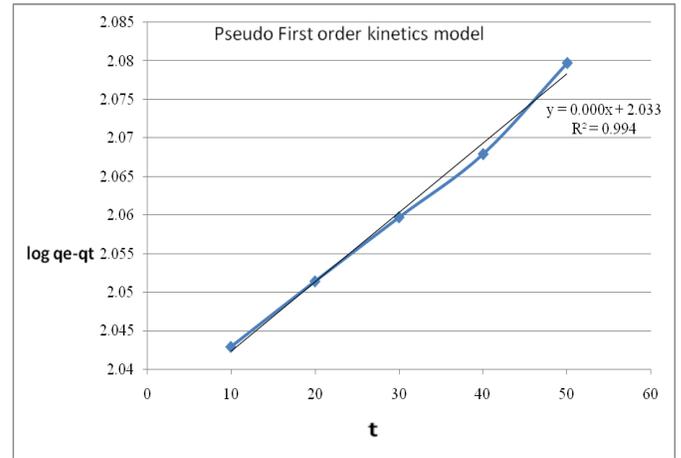


**Fig. 7: Freundlich isotherm plot of Bismark Brown R dye using *Laplap purpureus* plant stem powder**

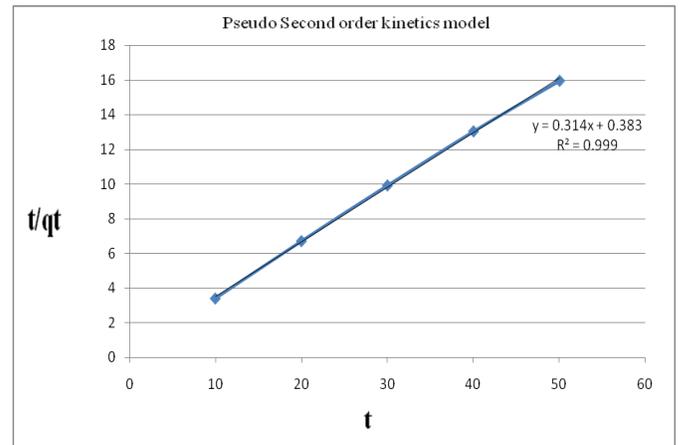
**3.6. Kinetic Models**

The sorption coefficient and the capacity of the equilibrium  $q_e$  can be determined from the linear plot of  $\log (q_e - q_t)$  versus time  $t$  from the fig. 5. It was evidence that the linear plot exhibits the applicability of the Lagergren equation;  $q_e$  values were present at table 6. The results represented that the concentration of dye has no significant effect. The correlation coefficient of  $r^2$  is 0.9943. If second order kinetics is applicable; the plot of  $t/q_t$  versus  $t$  should give a linear relationship (Fig. 9). The  $q_e$  and  $r^2$  values can be derived from the plots. The data values were reported in Table 6.

It was seen that the pseudo-second-order model fit very well and reporting a very high correlation coefficient of 0.9994 with  $q_e$  126.35.



**Fig 8: Pseudo first order kinetic model**



**Fig. 9: Pseudo second order kinetic model.**

**Table 5: Langmuir and Freundlich model parameters**

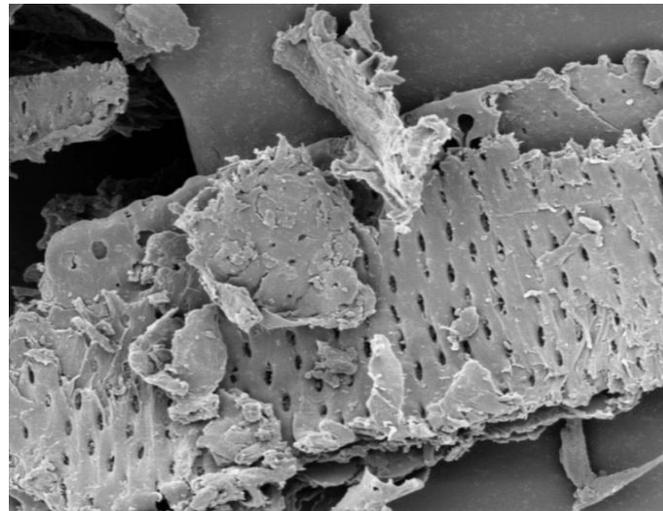
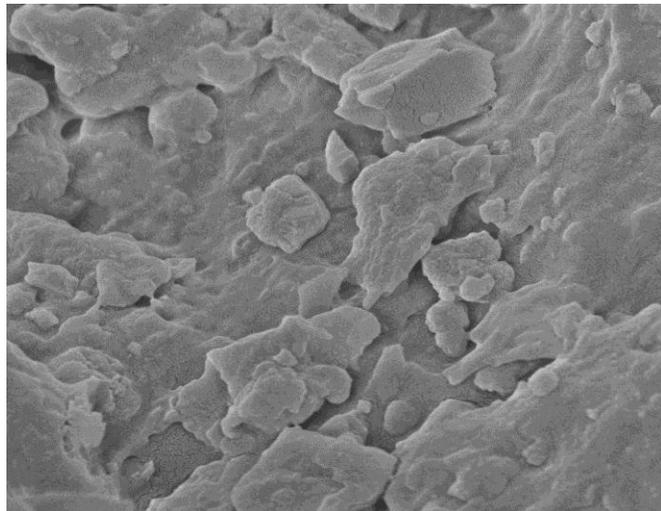
Temperature	Langmuir model			Freundlich model		
	$q_m(\text{mg/g})$	$b(\text{L/mg})$	$r^2$	$K_f(\text{mg}^{1-n} \text{g}^{-1} \text{L}^n)$	$n(\text{mg}^{1-n} \text{g}^{-1} \text{L}^n)$	$r^2$
30° C	17.59	0.0185	0.9924	2.5379	1.0348	0.9719

**Table: 6 Pseudo-first-order and second order kinetic parameters**

Pseudo-first-order			Pseudo-second-order		
$q_e(\text{mg/g})$			$q_e(\text{mg/g})$		
Theo.	Exp.	$r^2$	Theo.	Exp.	$r^2$
91.08	122.33	0.9943	111.17	126.35	0.9994

### 3.7. SEM Analysis

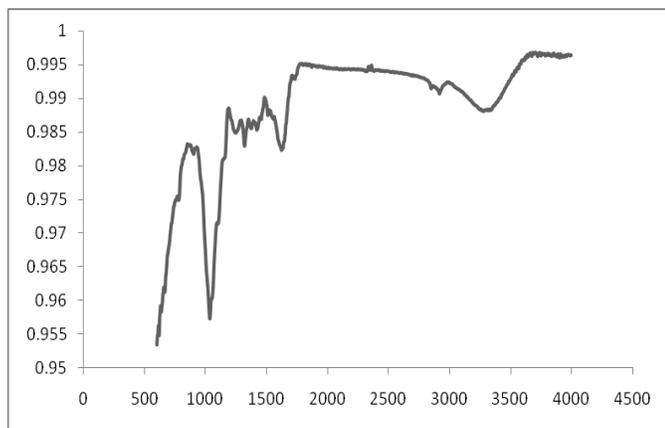
Morphological characters of the biosorbent can be analyzed by Scanning Electron Microscope (SEM) studies. It provides useful information about biosorbent. The SEM image of the raw LPSP biosorbent appears as the rough and uneven surface. This rough surface



**Fig. 10: SEM images of Raw LPSP and BBR dye Loaded LPSP**

### 3.8. FTIR of LPSP

The FTIR study provides the change in functional groups of biosorbent LPSP, spectra of the LPSP before and after the BBR dye adsorption shows in the figure 11a and 11b.

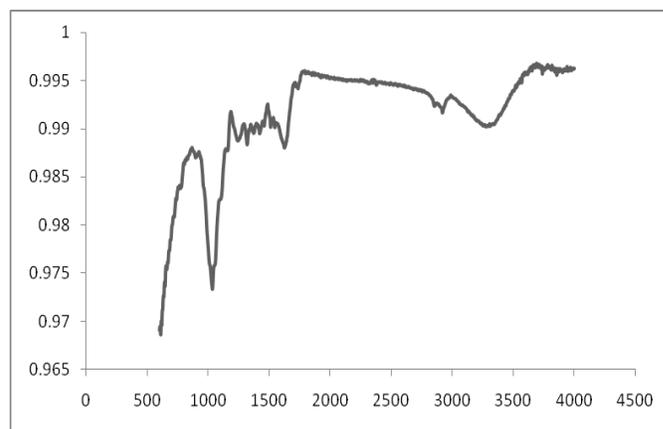


**Fig. 11a: FTIR spectra of unloaded LPSP**

FTIR spectrum of native biosorbent revealed a number of absorption peaks with the range of 600-4000  $\text{cm}^{-1}$ , which is only a sign of the complex chemical nature of this biosorbent. The aromatic compounds of the dye molecule showed characteristic absorption peaks at 2995  $\text{cm}^{-1}$  due to aromatic OH stretching. The absorption band at 1998  $\text{cm}^{-1}$  was due to CH stretching of alkyl group.

character must be considered as a reason for binding of BBR dye molecules. After the BBR dye loaded on LPSP biosorbent, the SEM image is considerable changes were observed in the structure of the biosorbent. The biosorbent appears as irregular surface and pores includes novel shiny particles after the adsorption process.

The absorption band at 3455  $\text{cm}^{-1}$  was due to NH stretching of amino group of dye molecule. The peaks appear at 1590  $\text{cm}^{-1}$ , 1480  $\text{cm}^{-1}$  was due to C-N-C stretching. The absorption spectrum of biosorbent treated with dye solution showed evident changes with respect to that of native biosorbent. Amongst these changes were the broadening of the absorption bands between 3455 and 1480  $\text{cm}^{-1}$  which suggests the superposition of numerous peaks that appeared in these regions. The band at 1480  $\text{cm}^{-1}$  and 3455  $\text{cm}^{-1}$  were due to the biosorbent binding with BBR dye molecule.



**Fig.11b: FTIR spectra of loaded LPSP**

#### 4. CONCLUSION

The results observed that the biosorbent received from locally home garden as costless material, *Laplap purpureus* plant stem powder (LPSP) have deserved in adsorption ability with respect to the removal of Bismark Brown R dye in the form of its aqueous solution. The conclusion made from the present studies shows that LPSP is an apt material for dye adsorption. pH, Adsorbent dose, Equilibrium time and Initial dye concentration are more favorable for the dye removal efficiency of the adsorbent. It was concluded that the sorption process is pH dependent and the upper limit adsorption capacity of BBR dye is at pH 8. The optimal dose weight was 300 mg. The best possible time was noted to be 50 min. and with 77.44% BBR dye removal efficiency. Present result shows that Langmuir model is better fit than Freundlich model for the adsorption equilibrium data. In the examined concentration range 50 to 250 mg/L, the results also reveals that, it follows pseudo second order than pseudo first order kinetic model. SEM and FT-IR spectral characterization clearly reveals that the adsorption of BBR dye onto biosorbent LPSP. Therefore, LPSP can be utilized for removal of BBR dye in the form of aqueous solution.

#### 5. ACKNOWLEDGEMENT

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