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# To Study Adsorption Capacity of Methylene Blue by Powdered Biomass of Ailanthus excelsa

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**ABSTRACT**

**The potential feasibility of powdered biomass of Ailanthus excelsa. for removal of methylene blue from aqueous solution was investigated. The effects of various experimental parameters such as Biomass dose, Contact time, and concentration of MB solution, pH, and temperature were investigated. The extent of methylene blue removal increased with the increased in contact time, The maximum removal observed at 12 to 15 minutes. The optimum pH is 2. The sorption increases with decreasing temperature. Adsorption data was better fitted to the Langmuir and Freundlich isotherm with the $R^2$ values. Langmuir and Freundlich equations were found to have the correlation coefficient value in good agreement. Adsorption of MB onto Powdered Biomasas of Ailanthus excelsa. followed pseudo second order kinetics with $R^2$ value 0.9996. The calculated values of $\Delta H°$, $\Delta S°$ and $\Delta G°$ were found to be 15.225.17 KJ/mol, -0.04656 KJ/mol and 1.8235 KJ/ mol, respectively. The equilibrium data were also fitted to the Freundlich equation and it was observed that the sorption process is spontaneous and exothermic in nature. The results in this study indicated that the untreated powdered biomass is cheap, easily available, ecofrieindly very good biosorbent for removal of organic dye methylene blue.**

**Key words: Adsorbent, Adsorption, Methylene Blue dye, Powder biomass, Ailanthus excelsa excelas, Langmuir, Freundlich and Temkin is otherm**

## INTRODUCTION

A large number of Industrial Effluents from dyeing and other related industries are containing highly colored species. Over 10,000 dyes with a total yearly production over 10 $X10^6$ MT are commercially available in industrial effluents. It is estimated that approximately 20% of the dye stuffs are lost in industrial effluents during manufacturing and processing operations. [1,2,3] Various physical, chemical and biological methods, including adsorption, biosorption, coagulation/flocculation, advanced oxidation, ozonation, membrane filtration and liquid–liquid extraction have been widely used for the treatment of dye-bearing wastewater. The advantages and disadvantages of every removal technique have been extensively reviewed. [4, 5] Highly colored wastes are not only esthetically unpleasant but also hinder light penetration and may disturb the ecosystem. Moreover, dyes itself fare toxic to some organism. Methylene blue (MB) is a cationic dye having various applications in chemistry, biology, medical science and dyeing industries. Its long term exposure can cause vomiting, nausea, anemia and hypertension. [4, 6] The removal of dyes in an economic way remains an important issue for researchers and environmentalists. Adsorption is a very effective separation technique in terms of initial cost, simplicity of design, ease of operation and insensitive to toxic substances. Activated carbon is the most efficient adsorbent used for dye removal. But it is expensive to produce and regenerate. [5, 7] Kale A. A. , (2020).

Biosorption of Hg2+ions by Sulphonated biomass of Stalks of Prunus cerasus, [8] .Ailanthus excelsa. sa, commonly known as tree of heaven, is a large deciduous tree found in India and Sri Lanka.[9] In Tamil, it is also known as Pi-Nari Maram due to its disagreeable odor. The trees are grown along the edges of fields and rivers to mark boundaries and prevent soil erosion.[9-10] The tree has several uses in medicine as the gum and the bitter, aromatic leaves are reported to have medicinal properties. The bark is a febrifuge and can be used as a treatment against asthma, bronchitis and dysentery.[9] The leaves and bark are also in good repute as a tonic that is used after labor. The juice of the leaves and fresh bark is used as a remedy for after-pains.[10] The tree is also used in matchstick industry,[11] as fodder for goats,[12] and is one of the best tree used to trap Suspended Particulate Matter. M. Auta, B.H Hameed, (2011) reported the Preparation of waste tea[13] Nitrated biomass. Using potassium acetate as an activating agent for adsorption of Acid Blue 25 dye.

The O.Tunc, H. Tanacı, Z. Aksu (2009) has been studied the Potential use of cotton plant wastes [14] for the removal of black B reactive dye. V.K.Gupta, D.Pathania, S.Agarwal, P.Singh, (2012) reported the Adsorptional photo catalytic degradation of methylene blue onto pectin-Cu Sn[15] a nanocomposite under solar light. Various physical, chemical and biological methods, including adsorption, biosorption, coagulation/flocculation, advanced oxidation, ozonation, membrane filtration and liquid–liquid extraction have been widely used for the treatment of dye-bearing wastewater. The A.A.Kale has been reported study sieved agro waste of Cicer arientinum. The B.H. Hameed (2009), M.A.M. Salleh, D.K. Mahmoud, W.A. Karim, A. Idris (2011), worked on Preparation, characterization and evaluation of adsorptive properties of orange peel based Nitrated biomass. Via microwave induced K2CO3 activation [16-17]. Highly efficient removal of synthetic dyes from wastewater has been attempted by various mathods.among them adsorption is a promising one because of its low cost and High efficiency, particularly for biomass sorbent derived from agriculture residuse. The adsorptive capacity of ailanthus excelsa. sa material to environment protection via adsobting diverse organic and inorganic pollutants from aqueous solution to clan wastewater has been widely studied.to data ,diverse plant material or their modified from have been explored as biosorbents for removal of metal ions or organic pollutants from aqueous solution. Ailanthus excelsa. sa, commonly known as tree of heaven, is a large deciduous tree found in India and Srilanka. In Tamil, it is also known as Pi-NariMaram due to its disagreeable odor. The trees are grown along the edges of fields and rivers to mark boundaries and prevent soil erosion. We take the tree of biomass for experimental purpose. I have take trunk of tree and that wood and powder in the powdered machine, then that biomass take in the 500 micron mesh for uniform size of powder material.

## MATERIALS AND METHODS

### Preparation of dye solution

Methylene blue (C16H18N3SCl.3H2O) was obtained from E.Merck, India and was used for sorption study. The solution of required concentration was prepared by dissolving the required amount of MB dye in distilled water.The dye stock solution was prepared by dissolving accurately weighed dye in distilled water to a concentration of 500 mg/L. The experimental solution was obtained by diluting the dye stock solution in accurate proportions to different initial concentrations like 15 mg/L, 20mg/L, 25mg/L and 30mg/L.

### Preparation of Biosorbent:

The locally available Plant of Ailanthus excelsa. sa was collected. The stalks of Ailanthus excelsa.were washed with distilled water to remove adhered impurities from its surface. The stalk part was dried at $100^0$C for 12 hr. The dried stalks was then crushed, milled and sieved through 500 micron particle size mesh to obtain powdered biomass which is used for MB removal study.

### Adsorption experiments

Adsorption of Methylene blue (MB) on powder Ailanthus excelsa. biomass of was carried out using a adsorption batch experiments method. The effect of contact time, adsorbent dose, pH, temperature and concentration of MB dye were investigated. The adsorption study was carried out with four different initial concentrations like 15mg/L, 20mg/L 25mg/L, 30mg/L MB. The effect of contact time was used to determine equilibrium time for the adsorption. For the experiment, 25 ml of MB solution was added into a conical flask containing 0.100 g of powder biomass of Ailanthus excelsa. s, stirred on magnetic stirrer for the time of 2,4,6,8,10.15,30,and 120 minutes respectively. The flasks were agitated in an isothermal water-bath shaker at 100 rpm and $27^0$C until the equilibrium is reached. After decantation and filtration, the equilibrium concentrations of dye in the solution were measured at 665 nm using UV-visible spectrophotometer. The pH of solution was adjusted with 1N HCl and 1N NaOH solutions. And this is filtered and absorption measured with double beam spectrophotometer. In the pH study, the pH of MB solution was adjusted in the range of 2-10 by adding 0.2 N HCL and 0.2 N sodium hydroxide and same experiment were carried out absorption is measured.

The same experiment is carried out to investigate the effect of adsorbent dosage on MB adsorption. The various amounts of adsorbent dose in the range of 0.05 g to 05 g were added to different concentrations of MB such as 0.15 mg/L,0.20mg/L,0.25 mg/L, and 0.30 mg/L MB. After stirring the flasks for predetermined time intervals,

filtered and absorption of each concentrations were measured. The isotherms study was carried out by varying the dosage of the adsorbent with the same concentration of MB solution. Each solution (25ml) was treated with (0.05-0.5) g of adsorbent. All experiments were conducted in duplicate and the controls (with no adsorbent) were simultaneously carried out to ensure that adsorption was by powder biomass and not by the container .The amount of dye adsorbed and percentage removal of MB were calculated using Eqs. (1) and (2), respectively:

$$q_e = (C_0 - C_e)\frac{V}{M} \quad (1)$$

$$\%\text{Removal} = \frac{C_i - C_e}{C_i} \quad (2)$$

Where:
qe amount of dye in mg per gram of adsorbent.Ci and Ce are respectively initial concentration and equilibrium time of MB (mg/l).V volume of solution. M mass of adsorbent.

**Adsorption measurement and isotherm.**

**Langmuir isotherm**

The Langmuir (1916) sorption isotherm is applied to equilibrium sorption assuming monolayer sorption onto a surface with a finite number of identical sites .constitution and fundamental properties of solids and liquids. The Langmuir equation is written as

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

The shape of this isotherm can also be expressed in terms of separation factor (RL), which is given as follows [6]:

$$R_L = \frac{1}{1 + K_L C_0} \quad (4)$$

Where KL is Langmuir constant (L/mg) related to the affinity of binding sites and the free energy of sorption. qe is dye concentration at equilibrium onto bio sorbent (mg/g). Ce is dye concentration at equilibrium in solution (mg/l).qm is dye concentration when monolayer forms on biosorbent (mg/g).

**Freundlich isotherm**

The A.E. Nemr, W.O. Abdel, E.S., AmanyA. Khaled, (2009) Removal of direct blue-86 from aqueous solution by new Nitrated biomass. developed from orange peel[19]. The Freundlich equation for heterogeneous surface energy systems shown by is given by Eq. (5).

$$\ln q_e = \ln K_F + \frac{1}{n} \ln C_e \quad (5)$$

The KF and n are Freundlich constants, determined from the Plot of ln qe versus ln Ce. The parameters KF and 1/n are related to sorption capacity and the sorption intensity of the system. The magnitude of the term (1/n) gives an indication of the favorability of the sorbent/adsorbate systems.P.K. Malik, (2003) Use of Nitrated biomass. Prepared from sawdust and rice-husk for sorption of acid dyes: a case study of acid yellow36.

**Tempkin isotherm**

This isotherm was studied by X.S. Wang, Y. Qin. (2005) Equilibrium sorption isotherms for of Cu2+ on rice bran. The linearized Tempkin equation is given by the following equation.

$$q_e = \beta \ln \alpha + \beta \ln C_e \quad (6)$$

where $\beta = \frac{RT}{b}$

T is the absolute temperature in Kelvin, R is the universal gas constant (8.314 J/mol K), and b is the
Tempkin constant related to heat of sorption (J/mg). The Tempkin constants a and b are calculated from the slope and intercept of qe versus ln Ce.

## RESULT AND DISCUSION

**Adsorption Isotherm**

**Effect of adsorbent dose and initial dye concentration.**

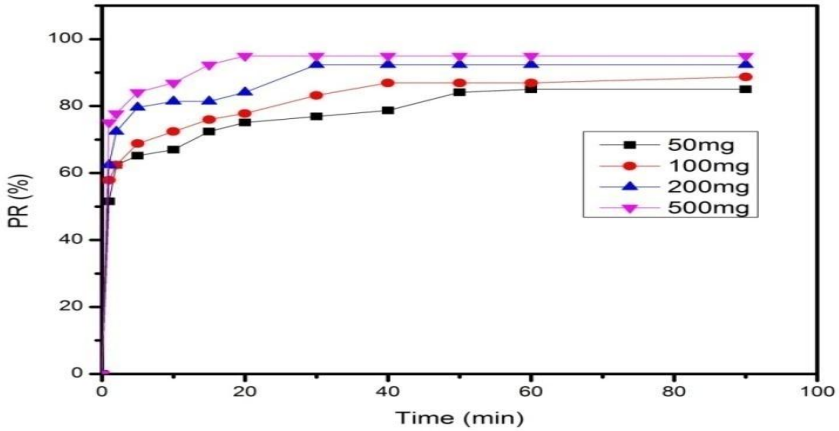



**Figure: 1 Effect of adsorbent dose on MB removal**

**Effect of Time**

on the rate of adsorption by powder biomass of Ailanthus excelsa. was carried out. The figure-1 effect of time indicates that the removal of dye (adsorbate) initially increases with time but attains equilibrium within 15-30 minutes for all types of biomass dose. The adsorption process was found to be very rapid initially, and a large amount of the total concentration of dye was removed in the first 15 minutes. Though it was observed that adsorption of dye increased with an increase in dye concentration in the solution, which shows that removal of dye is dependent upon the concentration of the dye solution. But as a whole the percent removal decreases with the increase in dye concentration as shown in Figure-2.

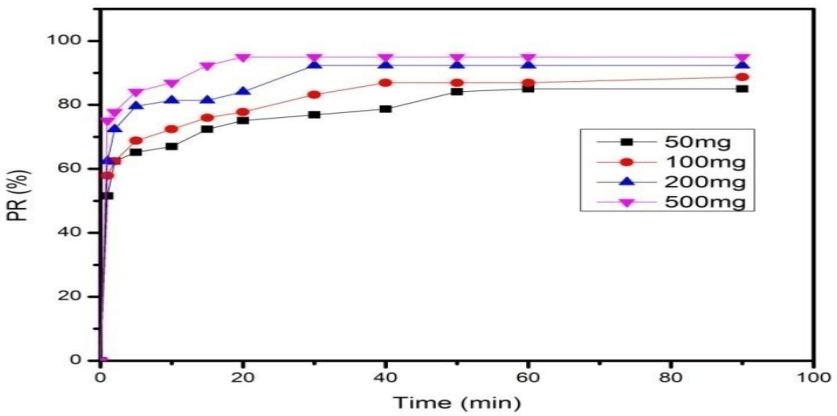



**Figure 2 Effect of Time on MB Removal**

**Effect of concentration**:

Further increase in dye concentration showed no significant changes in removal efficiency (Fig.2). This is due to the fact that with increased dye concentration, the driving force for mass transfer also increases. At low concentrations there will be unoccupied active sites on the adsorbent surface. Above optimal MB concentration, the active sites required for the adsorption of dye will lack (Barkaal., 2011; Iqbal et al., 2011). The various amounts of adsorbent dose in the range of 0.05 g to 05 g were added to different concentrations of MB such as 0.15 mg/L,0.20mg/L,0.25 mg/L, and 0.30 mg/L MB. After stirring the flasks for predetermined time intervals, filtered and absorption of each concentrations were measured. The effect of varying the powder biomass on aqueous dye

solution are presented in experiment show a decreasing trend in dye concentration at a faster rate as the adsorbent mass is increased. 500 mg powder biomass gave the greater removal at all levels of the adsorbent dose. An equilibrium percentage removal rate of 92.2% and was achieved with 500 gm/l in 60-90 minutes of adsorbate concentration of 25 mg/L. Initially the rate of increase in the percent dye removal has been found to be rapid which slowed down as the dose increased. This phenomenon can be explained, based on the fact that at lower adsorbent dose the adsorbate (dye) is more easily accessible and because of this, removal per unit weight of adsorbent is higher. (shown in fig-3)

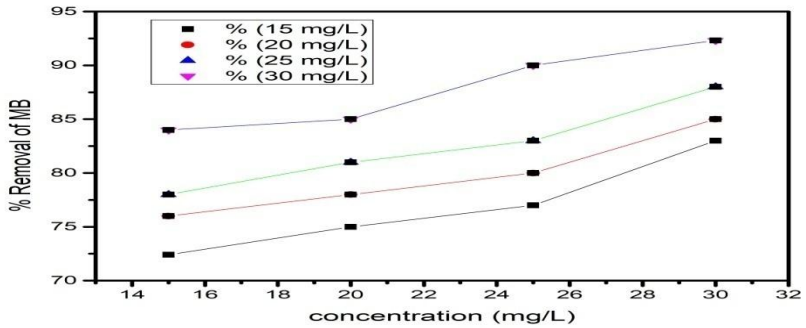



**Fig. 3 Effect of concentration of powder biomass**

For a particular experiment, the rate of adsorption decreased with time, it gradually approached a maximum adsorption, and owing to continuous decrease in the concentration driving force and it also indicate that the adsorbent is saturated at this level. In addition it is observed that initial rate of adsorption was greater for higher initial dye concentration because as the resistance to the dye uptake decreased, the mass transfer driving force increased. The time variation adsorption increases continuously and seems to be smooth which, is indicative of the formation of monolayer coverage on the surface of adsorbent powder biomass could remove a maximum of 92.2 %Blue at initial dye concentration of 25 mg/L, while for dye concentrations of 30 mg/L, the adsorption of the dye was well above 92.2 % in 60-90 minutes. Both cases were studied at natural pH, temperature 15°C and adsorbent dose of 500 gm/l. Though it was observed that adsorption of dye increased with an increase in dye concentration in the solution, which shows that removal of dye is dependent upon the concentration of the dye solution. But as a whole the percent removal decreases with the increase in dye concentration as shown in Figure-2.

**Effect of pH**

Solution pH is an important monitoring parameter influencing the sorption behavior of adsorbate onto biosorbent surface due to its impact on both the surface binding-sites of the biosorbent and the dye solution chemistry. In the present study, the effect of pH on biosorption of MB onto sieve biomass was studied over a pH range of 2–10. Results are shown in Fig. 4. The amount of dye removed at equilibrium decreases with decreasing pH, appreciably up to pH 2.0. With further increase in pH, there is no significant increase in the amount of dye removed. Maximum removal is observed at pH 2.0. Hence, all further experiments were carried out at pH 2.0 The pH of the aqueous solution affects both the surface charge of the biosorbent material as well as the degree of ionization of the dye molecule. Feathers mainly contain N–H, C=O and C–H functional groups on their surface (Aguayo-Villarreal et al. 2011). Protonation of these functional groups at low pH values renders a net negative charge to the biosorbent surface while deprotonation of the functional groups at high pH values render it positively charged. The pKa of MB is 0.04 (Weng et al. 2009); hence, it is completely ionized at pH [0.04 and exists as cationic species. At low pH values, there exists a strong electronegative repulsion between the positively charged dye ions and the negatively charged biomass surface resulting in high dye uptake capacity. On the contrary, as the pH of the dye solution increases, considerable decreases in adsorptive removal of dye is observed due to between negatively charged sites on the biosorbent and the dye cations. Similar results were previously reported for biosorption of MB from aqueous solution.

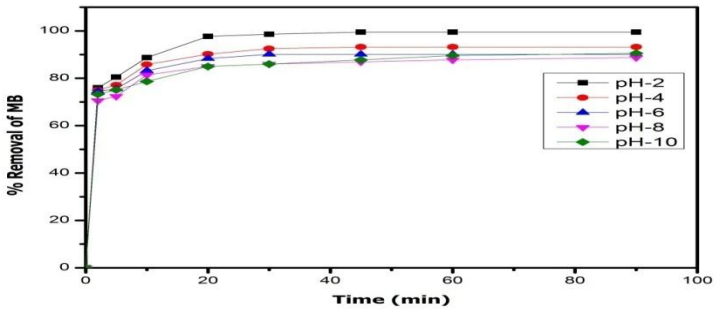



**Fig. 4 effect of pH**

**Effect of temperature:**

Temperature has pronounced effect on the sorption removal of dyes from aqueous solutions. As such, the effect of temperature on the biosorption process of MB was studied in the range of 288 to318 K and the results are depicted in Fig. 5. The figure shows that the dye uptake capacity decreases with increasing temperature. This finding suggests that MB uptake process was exothermic in nature. The negative correlation between temperature and dye biosorption capacity may be due to the weakening of bonds between the dye molecules and the active site of the biosorbent (Chowdhury and Saha 2010a). Also with increasing temperature, the solubility of MB increases. Consequently, the interaction forces between the solute and the solvent are stronger than those between the solute and the biosorbent. As a result, the solute is more difficult to adsorb (Chowdhury and Saha 2010a). Similar findings have been reported for biosorption of MB by Brazil nut shells (de Oliveira Brito et al. 2010) and Ulothrix sp. (Dogar et al. 2010).

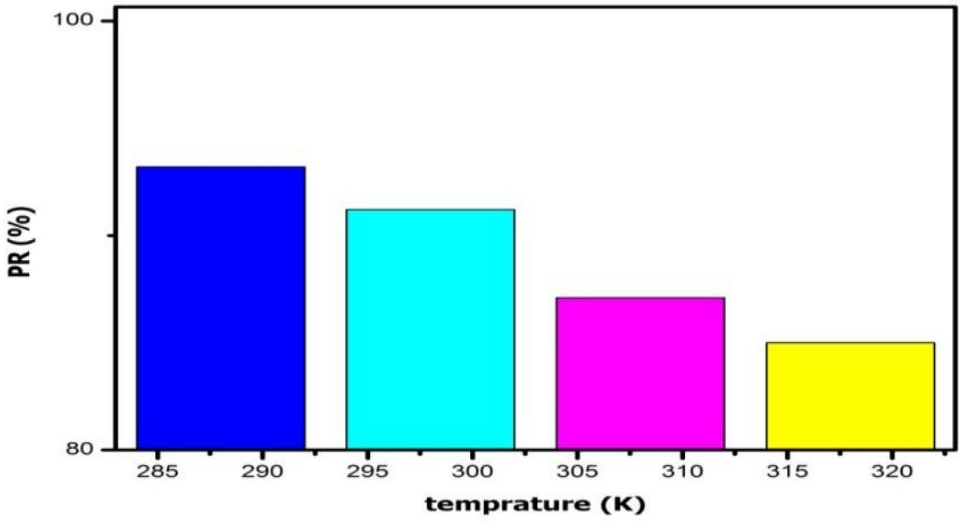



**Fig. 5 Effect of temperature**

**Adsorption kinetics.**

Adsorption is a physiochemical process that involves the mass transfer of a solute (adsorbate) from the fluid phase to the adsorbent surface. A study of kinetics of adsorption is desirable as i provide information about the mechanism of adsorption, which is important for efficiency of the process. The applicability of the pseudo-first-order and pseudo-second-order model was tested for the adsorption of MB onto sieved biomass. The best-fit model was selected based on the linear regression correlation coefficient, $R^2$ The first-order kinetic model The rate equation is one of the most widely used adsorption rate equations for the adsorption of solute from a liquid solution. The pseudo-first-order kinetic model of may be dqt/dt = k1(qe − qt) Integrating this equation for the boundary conditions t = 0 to t = t and q = 0 to q = qt, gives: ln(qe − qt) = ln qe − k1t where qe and qt are the amounts of MB adsorbed (mol g−1) at equilibrium and at time t (min), respectively, and k1 is the rate constant of pseudo-first-order adsorption (min−1). The validity of the model can be checked by linearized plot of ln(qe − qt) versus t. The rate constant of pseudo-first-order adsorption is determined from the slope of the plot. The values of k1 and qe at different concentrations, initial solution pHs, and temperatures. The second-order kinetic model is expressed as.

dqt dt = k2(qe − qt) 2

Rearranging the variables in the above Eq.

dqt (qe − qt) 2 = k2 dt ------

Taking into account, the boundary conditions t = 0 to t = t and q = 0 to q = qt, the integrated linear form of above Eq.can be rearranged to obtain as:

t/ qt = 1 /k2q2 e + t/ qe

The initial adsorption rate, h (mol g−1 min−1) is expressed as

h = k2q2 e

where the initial adsorption rate (h), the equilibrium adsorption capacity (qe), and the second-order constants k2 (g mol−1 min−1) can be determined experimentally from the slope and intercept of plot/qt versus t. The k2 and h values under different conditions were calculated and listed in Table 1. As discussed above, the validity of the model of pseudo

first order and the pseudo-second-order kinetic model can be checked by each linearized plot. If the second-order kinetics is applicable, then the plot of t/qt versus t should show a linear relationship. The linear plots of t/qt versus t show good agreement between experimental (qe (exp)) and calculated (qe (cal)) values (Table 1). The correlation coefficients for the second-order kinetics model (R2) are greater than 0.999, indicating the applicability of this kinetic equation and the second order nature of the adsorption process of MB onto sieved biomass. Similar phenomena have been observed for MB adsorption on coir pith carbon, cedar sawdust and crushed brick, perlite, sepiolite and wheat shells18.

**Table: 1 Kinetic data of MB Sorption**

| | | |
|---|---|---|
| Pseudo first order model<br>k1 ($min^{-1}$)<br>Slope=0.07916 | qe (mg/g)<br>7.37768 | R2<br>0.8100 |
| Pseudo second order model k2 (g/(mg min<br>Slope=07956 | 5.62271 | 0.9996 |
| Intraparticle diffusion model kd (mg/g min)<br>3.69 | C (mg/g)<br>2.19 | 0.305 |
| Thermodynamic parameters<br>$\Delta H^0$(kJ/mol)<br>15.22517 | $\Delta S^0$ (J/mol K)<br>-46.53 | DG°(KJ/mol)<br>1.8235 |

Calculation for adsorption of MB on sieved biomass Parameters Kinetic models T (◦C) [C0] (mol L−1) × 104 pH SS (rpm) [I] (mol L−1) PS (m) Pseudo-first-order model Pseudo-second-order model R2 qe(cal) (mol g−1) × 105 qe(exp) (mol g−1) × 105 k2 (g mol−1 min.

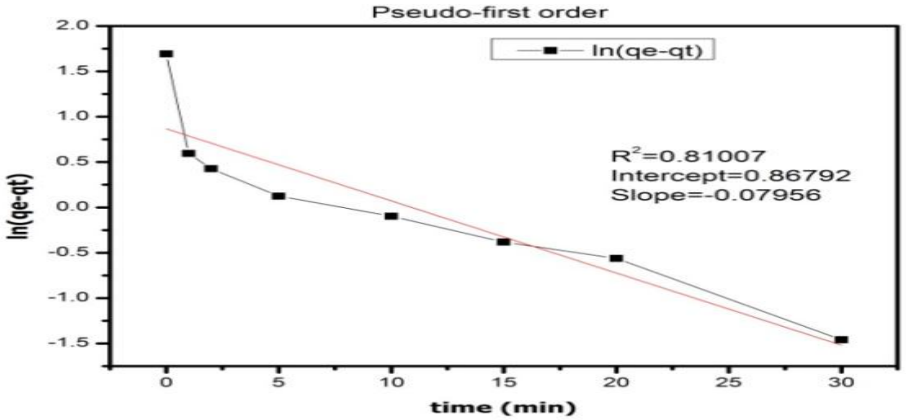



**Fig. 6 Adsorption kinetics**

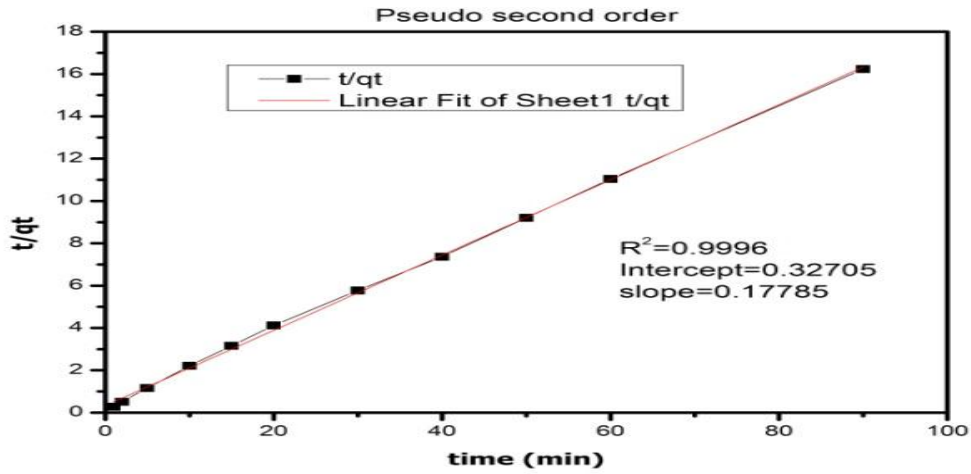
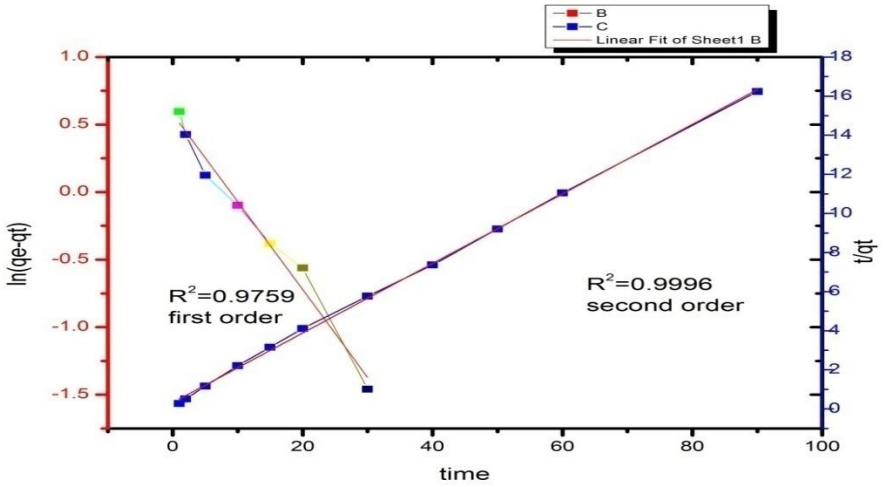



**Figure 7 Kinetics of Adsorption**




**Adsorption Isotherms:**

The adsorption equilibrium of MB dye was investigated over a range of MB dye concentrations at 15 mgL,20 mg/L,25 mg/L & 30 mg/L the results are presented in Figure 7, which gives the relationship between the equilibrium adsorption capacity ($q_e$, mg=g) and the equilibrium MB dye concentration ($C_e$, mg=L). These adsorption equilibrium data were used to check the well-known adsorption isotherm models, the Langmuir (1918) and Freundlich (1906) isotherm models. To determine the maximum monolayer adsorption capacity of STOP, the Langmuir adsorption isotherm model was used to describe the adsorption equilibrium experimental results as follows

$$\frac{1}{qe} = \frac{1}{qm} + \frac{1}{qmKL}\frac{1}{Ce}$$

where $q_e$ is the adsorption capacity at equilibrium (mg=g), $q_m$ is the maximum monolayer adsorption capacity (mg=g), $K_L$ is the constant related to the free energy of adsorption (L=mg), and $C_e$ is the concentration of MB dye in the solution at equilibrium (mg=L). The Langmuir adsorption isotherm model parameters and the related coefficient of determination ($R^2$) values are summarized in Table I. It can be seen that the Langmuir isotherm model slightly fitted the experimental data according to the coefficient of determination values. The estimated maximum monolayer adsorption capacities ($q_m$) of MB dye increased slightly with increasing temperature. To check the multilayer adsorption of MB dye onto powdered biomass, the adsorption experimental data were further applied to the Freundlich adsorption isotherm model. The Freundlich adsorption isotherm model is an empirical equation that assumes that the adsorption onto a heterogeneous energetic distribution of the active sites is accompanied by the interaction between the adsorbed species. The linear form of the Freundlich adsorption isotherm model is as following.

$$\log q_e = \log K_f + \frac{1}{n} \log C_e$$

where $K_F$ ((mg=g)(L=mg)$^{(1=n)}$) is the Freundlich equilibrium constant and $n$ (g=L) is the exponent in the Freundlich equation.

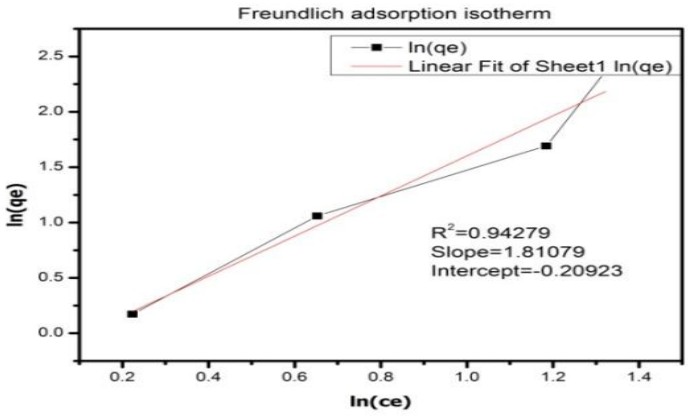
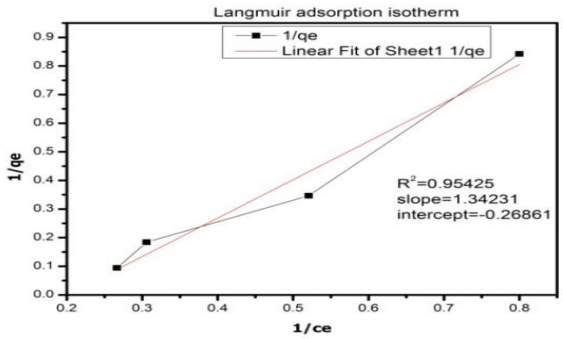



**Fig-8 Adsorption Isotherms**



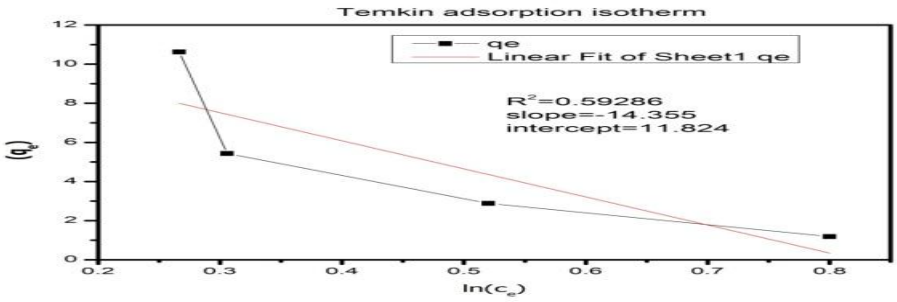




**Table -2 Results of various isotherm plots for the adsorption of MB onto powder biomass.**

| **Models** | Isotherm Constant | | | |
|---|---|---|---|---|
| Langmuir | $q_m$(mg/g)<br>40.10 | KL (L/mg)<br>0.038 | RL<br>.0.06-0.35 | $R^2$<br>0.9542 |
| Freundlich | n<br>2.24 | (mg/g)<br>5.41 | <br>$8.4 \times 10^{-3}$ | $R^2$<br>0.9427 |

## CONCLUSION

In this work shows promising adsorption capacity for methylene blue removal by powdered biomass of Ailanthus excelsa .The maximum sorption for MB solution concentration (0.100 g/25 ml), sorbent dosage (0.5 g/25 ml), contact time (15 min) and temperature (280 K) were observed. The maximum removal of methylene blue dye was attained at pH 2.0 The equilibrium data were fitted well in the Langmuir, Freundlich isotherm models which confirmed that the sorption is heterogeneous and occurred through physico–chemical interactions. The rate of sorption was found to obey pseudo-second order kinetics with correlation coefficient value R2 is 0.9996.

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