

THE EFFECTIVENESS OF NATURAL ADSORBENT FOR REMOVAL OF DYE USING TWO ISOTHERM MODELS

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ABSTRACT

The low-cost and more efficient adsorbents of *Azadirachta Indica* (Neem) are charcoal, bark; leave and root were investigated for its optimistic de-colorization of wastewater containing a dye. The effect of various parameters such as contact time, pH, temperature and adsorbent dosage has been carried out in this study. Two isotherm models were developed for compliance with Langmuir and Freundlich isotherm. The results of the goodness fit test based on Root Mean Square Error (RMSE), R² for Langmuir and Freundlich models were given as, for pH (0.9745 – 0.99919), (0.4831 – 0.9999) for Adsorbent Dose is (0.9034 – 0.9221), (0.4689 – 0.9884) for contact time (0.9494 – 0.9513), (0.4677 – 0.9990) and temperature (0.9264 – 0.9349), (0.3755 – 0.5849) respectively. Both models were found to be best fit on decolorization of dye with contact time, pH, temperature and adsorbent dosage. pH 5 value indicated that adsorbent was effective in the removal of dye.

Keywords: Adsorption isotherm, decolorization, adsorbent parameters and dye

INTRODUCTION

Generally, a dye is a substance that bears affinity to the substrate that they are applied upon. In most cases, they are applied in aqueous solution (Vankatesan *et al.*, 2016; Saturaki *et al.*, 2015; and Musah *et al.*, 2018). Therefore, there is need of a mordant to improve its binding with fabric. It appears to be colored because of the wavelengths of light they absorbed but these absorbed wavelengths of light tend to differ from one another (Hu, *et al.*, 2020; Manga *et al.*, 2020; Sheba *et al.*, 2016; Mohameed *et al.*, 2018; Khote *et al.*, 2019).

For high production propose, dye generate colored waste water resulting in pollution of the immediate environment. Factories such as textile, paper and food industries, tanneries and electroplating, discharge colored wastewater (Musah *et al.*, 2018 and Soumya *et al.*, 2018). Color or dye being one of the recalcitrant, continue for long distances in flowing water reducing, photosynthesis and inhibit growth of aquatic biota by blocking out sunlight and utilizing dissolved oxygen (Edet *et al.*, 2010; Vankatesan *et al.*, 2016; Saturaki *et al.*, 2015; Musah *et al.*, 2018 and Soumya *et al.*, 2018). Some dyes may cause, skin irritation, allergic, dermatitis cancer and mutation in man.

Several strategies in recent years have been proposed for the utilization of vegetable wastes, including the recovery of energy (Sheba *et al.*, 2016 and Obaid *et al.*, 2020) or value-added products or the use as adsorbent materials (Musah *et al.*, 2018 and Soumya *et al.*, 2018). The removal of textile dyes from colored effluent is

one of the major concerns in the textile industries with a number of techniques introduced to treat the textile wastewater. The conventional methods are expensive (Edet *et al.*, 2010; Vankatesan *et al.*, 2016; Saturaki *et al.*, 2015; Musah *et al.*, 2018 and Soumya *et al.*, 2018). A number of adsorption studies were carried by various researchers as adsorption is quite popular for its simplicity and efficiency (sheba *et al.*, 2016 and Obaid *et al.*, 2020). Activated carbon is commonly used in adsorption assessment, however, raw material of this technique is relatively expensive (Pandhare *et al.*, 2013). As a result, implementation of activated carbon in large effluent treatment is rare (Islam *et al.*, 2021; Mhemeed *et al.*, 2018).

Adsorption is the most suitable method for dye removal from industrial effluents, due to its commercial acceptability at low cost with high performance. Activated carbon is the most effective adsorbent widely employed to treat wastewater containing different classes of dyes, recognizing the economical drawback of commercial activated carbon (Hu, *et al.*, 2020; Sheba *et al.*, 2016; Mohameed *et al.*, 2018; Khote *et al.*, 2019).

Besides the activated carbon, recent research has shown the use of other low-cost adsorbents used in the areas of special interest. These include clays, silica gel, sawdust and peat, (Hu, *et al.*, 2020; Sheba *et al.*, 2016; Mohameed *et al.*, 2018; Khote *et al.*, 2019). In the selection of any adsorbents, researchers need to be mindful of the following Economic advantages, performance efficiencies and environment. Thus, researchers generally use low-cost adsorbents (Edet *et al.*, 2010; Vankatesan *et al.*, 2016; Saturaki *et al.*, 2015; Musah *et al.*, 2018 and Soumya *et al.*, 2018).

The study was undertaken to evaluate the efficiency of *Azadirachta Indica* (Neem) for charcoal, bark, leave and root as an adsorbent for the removal of dye from aqueous solutions (Hu, *et al.*, 2020; Sheba *et al.*, 2016; Mohameed *et al.*, 2018; Khote *et al.*, 2019). The kinetic, equilibrium and thermodynamic data deduces from the isothermal models on adsorption capacity were carried out to have more understanding of the absorption process, (Musa *et al.*, 2018; Soumya *et al.*, 2018; Natrayan *et al.*, 2022). The effect of adsorption parameters such as initial dye concentration, temperature, pH, adsorbent dose and contact time has been studied.

MATERIALS AND METHODS

The experiment of dye decolorization for *Azadirachta Indica* (Neem) Charcoal, Bark; Leave and Root, was conducted at the Laboratory of Department of Science Laboratory Technology, School of Science and Technology, Abubakar Tatari Ali

Polytechnic, Bauchi, Bauchi State, Nigeria.

Preparation of Samples

Azadirachta Indica (Neem) Leave, Bark and Root were collected from twigs of matured Neem tree from within Abubakar Tatari Ali Polytechnic, Bauchi, Bauchi, Nigeria. The leaves and bark were washed repeatedly by using distilled water and then exposed it to sun to remove moisture and soluble impurities. The samples were again washed with 1N HNO₃ and allowed to air dry under shade at room temperature until they become crisp. The four Adsorbent where dry for five days and the samples were grounded in a domestic mixer-grinder for the purposed of research analysis. Analytical grade was obtained from dye dealers at Aminu Street, Igbo quarters in Bauchi metropolis, Bauchi, Nigeria. The stock solution was prepared by dissolving 4g of dye salt and 8g of NaOH into a 400ml of hot water. The dye bath was collected in a clean reagent bottle before dyeing. The dye effluent was also collected in a clean reagent bottle after dyeing of a 45g polyester fabric. Dye adsorption experiments were carried out in batch mode by taking 50 mL solution of dye and treated with a dose of 0.1 g of adsorbent. The variable studies were pH, temperature, adsorbent dose, and contact time. The solution was stirred by magnetic stirrer (Hu *et al.*, 2020; Saturaki *et al.*, 2015; Paudel *et al.*, 2020; Khote *et al.*, 2019; Edet *et al.*, 2020; Farouk *et al.*, 2015). The values of percentage removal and amount of dye adsorbed were calculated using the following relationships.

$$\text{Percentage removal} = \left(\frac{C_i - C_f}{C_i} \right) \times 100 \quad \dots (1)$$

$$\text{Amount adsorbed} = \frac{C_i - C_f}{m} \quad \dots (2)$$

Where,

- C_i = Initial dye concentration ($mg\ l^{-1}$),
- C_f = Final dye concentration ($mg\ l^{-1}$),
- m = Mass of adsorbent ($g\ l^{-1}$).

Methods

The method of Vijayakumar, *et al.*, (2014) was adopted for this study. The removal of synthetic dye from textile dye effluent for each parameter: time, pH, adsorbent concentration and temperature were carried out. The batch experiments were carried out for the effect of time at a time interval of 24 hours for *Azadirachta Indica* (Neem) leave, bark and root for three (3) days.

The effect of pH was also ascertained at 5.0, 7.0 and 9.0 pH value for leave, bark and root respectively. Adsorbent concentration at various temperatures of 10 °C, 15 °C, 25 °C, 35 °C and 45 °C of a textile effluent was examined with a dosage rate of 2.5g, 5.0g, 7.5g 10.0g and 12.5g per 100ml solution of dye effluent for Neem leave, bark and root respectively.

Errors Analysis

In this research, we assessed the degree of linear relationship between the two isotherm models and the adsorbent parameter's by checking the level of error and coefficients of the goodness of fit (R^2) statistically. The Root Mean Square Error is calculated as follows (Mhemeed *et al.*, 2018 Manga *et al.*, 2021 and Islam *et al.*, 2021).

$$RMSE = \left[\frac{1}{N} \sum_{i=1}^N (y_i - x_i)^2 \right]^{0.5} \quad \dots (3)$$

$$R^2 = 1 - \frac{\sum_{i=1}^N (y_i - x_i)^2}{\sum_{i=1}^N (y_i - \bar{y})^2} \quad \dots (4)$$

Where by N is total interval numbers, y_i is the observed model frequency, x_i adsorbent parameter value. This case if RMSE value is low while R^2 is large the result more valid or proposed model will be accepted (Pobočiková *et al.*, 2018).

RESULTS AND DISCUSSION

Four parameters were selected for conducting a field adsorption study to lower color in the textile industries wastewater includes adsorption dosage (g), temperature 10°C, contact time (Hours) and pH.

Figure 1 shows the effect of temperature 10°C on the process of removing of dye solution with charcoal, bark, leave and root 1g/1L. The temperature changed with a range of (0 – 10°C). The result indicated that as the temperature rises from (0 – 10°C). The adsorption capacity increases. This is due to the fact that increase in the temperature will increase ions mobility in a large number but sudden decrease will also rapidly decrease in the mobility ions on charcoal, bark, root and leave. Since further increase in temperature may result in welling effect (Mhemeed *et al.*, 2018 and Islam *et al.*, 2021).

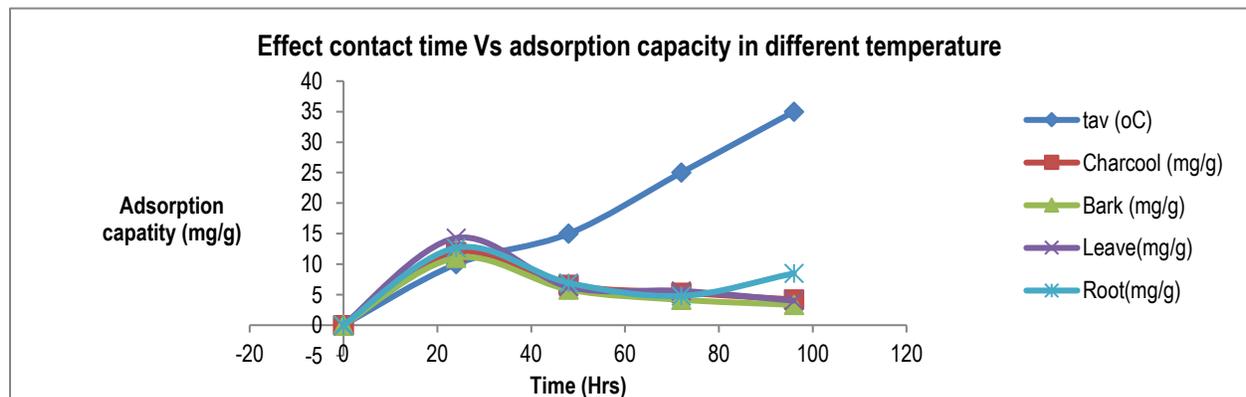


Figure 1. Variation on contact time Vs adsorption capacity in different temperature

Figure 2 shows the effect of pH at different contact time (Hours)

was investigated by a pH value of 5, 7 and 9 the removal was found to be minimum but high value was obtained as the initial pH of dye solutions were increased at above 5. The pH was maximum at 5 as shown on the graph of q_e (mg/g) Vs time (Hours). Decrease in the adsorption is influence by the surface charges on the adsorbent which in turn is influence by the pH on the solution. The result showed that availability of negatively charged groups at the

adsorbent surface are necessary for the adsorption of basic dyes which we see in pH 7 and 9 in this regard there may be accumulation of net positive charge in the adsorption. Thus more negative charge was available as the pH increases (Mohamed *et al.*, 2016).

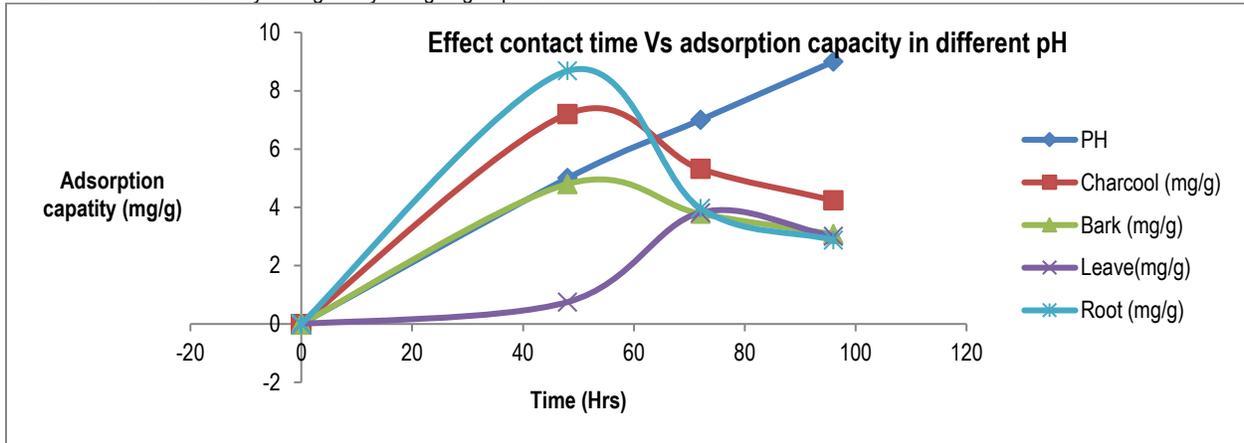


Figure 2. Variation on contact time Vs adsorption capacity in different pH

Figure 3: Shows the effect of adsorption dose for charcoal, bark, leave and root. From the result, the parameters are said to be efficient and highly effective for dye decolorization, with a varying range of 2.5g/100ml to 27.5g/100ml. From the graph the best dose

of adsorbent is 25g/100ml. The plot shows trend within the parameters as the contact time increases the adsorption dose rises or as the adsorption dose decreases the contact time also decreases. From the aforementioned the used of this parameter for dye decolorization is cost efficient.

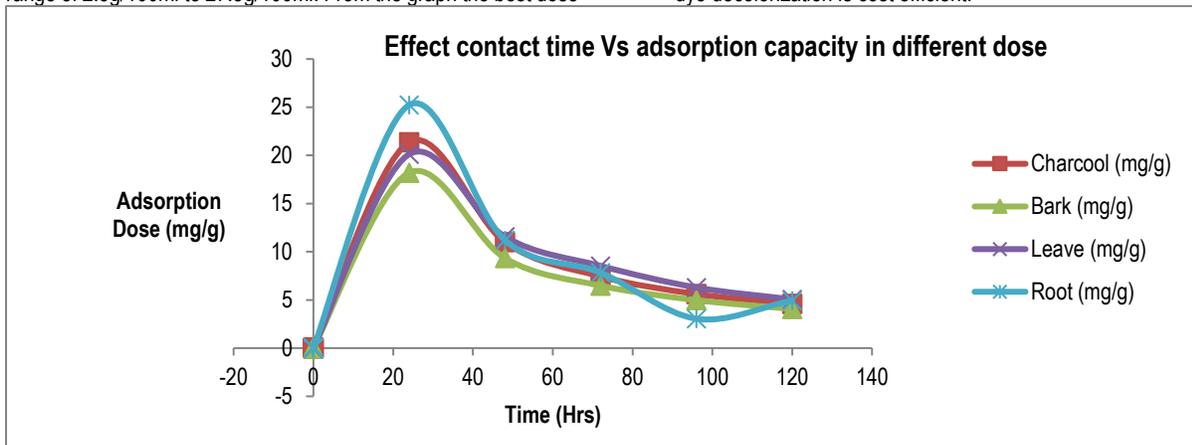


Figure 3. Variation on contact time Vs adsorption capacity in different dose

Figure 4 shows the effect of contact time with absorbent dose in (mg/g) of dye solution with pH 5 investigation. Initially the colour removal increase with increase in time and becomes lower in later

stages still saturation is allowed. At equilibrium the rate of decolourisation was 90% at a contact time of 110hours. This is due to saturation of the active site which does not allow further adsorption to occur (Musa *et al.*, 2018).

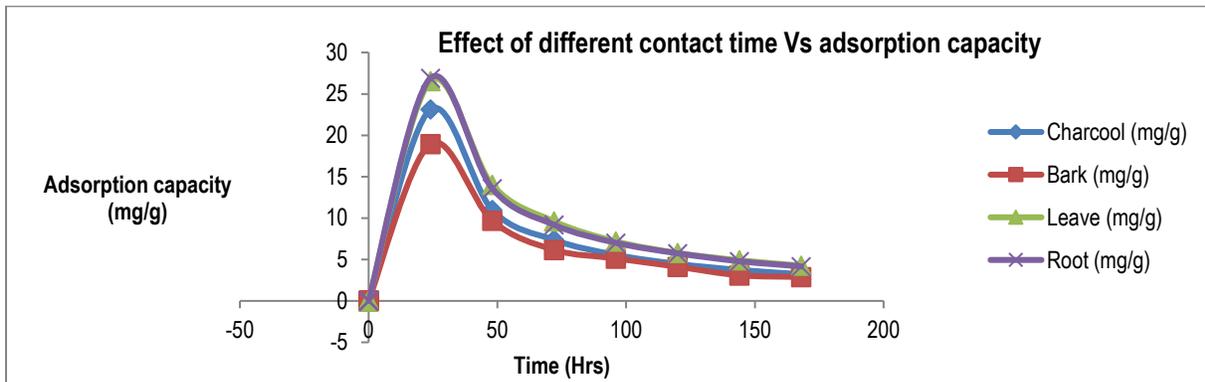


Figure 4. Variation of different contact time Vs adsorption capacity

Adsorption Isotherm

Freundlich adsorption isotherm

The result of the Freundlich adsorption isotherm was presented based on different processing parameters such as adsorbent dosage, temperature, contact time and pH to assess their fitness in term of the linear model with Freundlich adsorption isotherm (Farouk *et al.*, 2015, Hu *et al.*, 2020, Islam *et al.*, 2021 and Musa *et al.*, 2018)

The Freundlich equation was employed for the adsorption of dyes on charcoal, bark, leave and root on the adsorbent. The isotherm linear model is presented as.

$$\log q_e = \log K_f + \frac{1}{n} \log C_e \quad \dots (5)$$

Figure 5 shows the linear fitness of $\log q_e$ against $\log C_e$ based on temperature with a correlation coefficient of ($R^2 = 0.3755$ to 0.5849) indicates that the adsorption of dyes on charcoal, bark, leave and root are positively correlated which weakly obey Freundlich adsorption isotherm. The optimum temperature was found 15°C with a R^2 value of 0.5849 (Sheba *et al.*, 2016; Vankatesan *et al.*, 2016; Edet *et al.*, 2020).

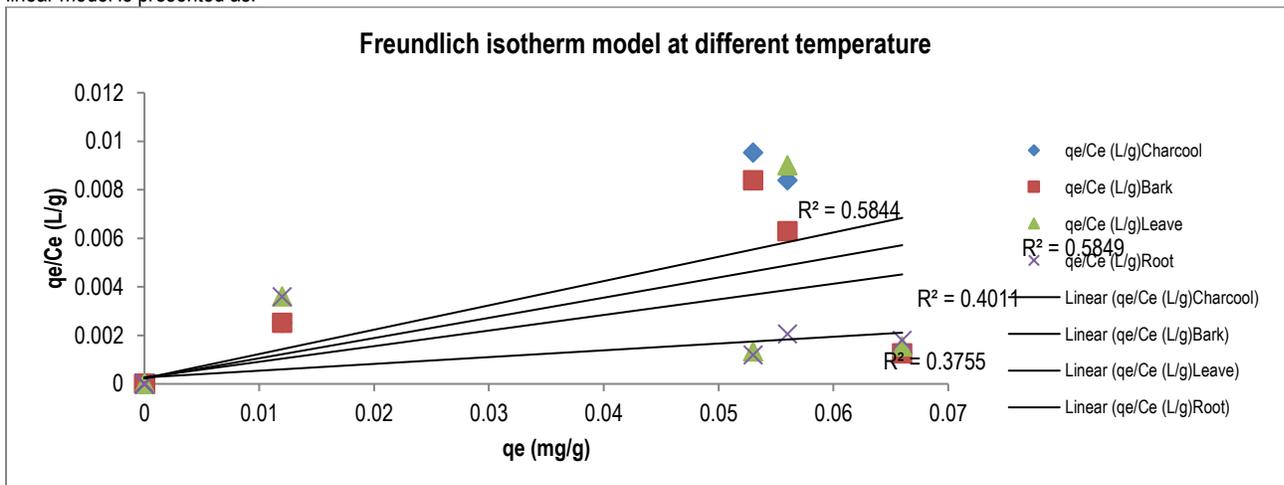


Figure 5. Linear model of Freundlich isotherm based on temperature

Figure 6 shows the linear fitness of $\log q_e$ against $\log C_e$ based on pH with a correlation coefficient of ($R^2 = 0.4831$ to 0.9999) indicates that the adsorption of dyes on charcoal, bark, leaves and root are positively correlated and strongly obey Freundlich

adsorption isotherm. The optimum pH was found to be $\text{pH } 9$ with a R^2 value of 0.999 (Sheba *et al.*, 2016; Vankatesan *et al.*, 2016; Edet *et al.*, 2020).

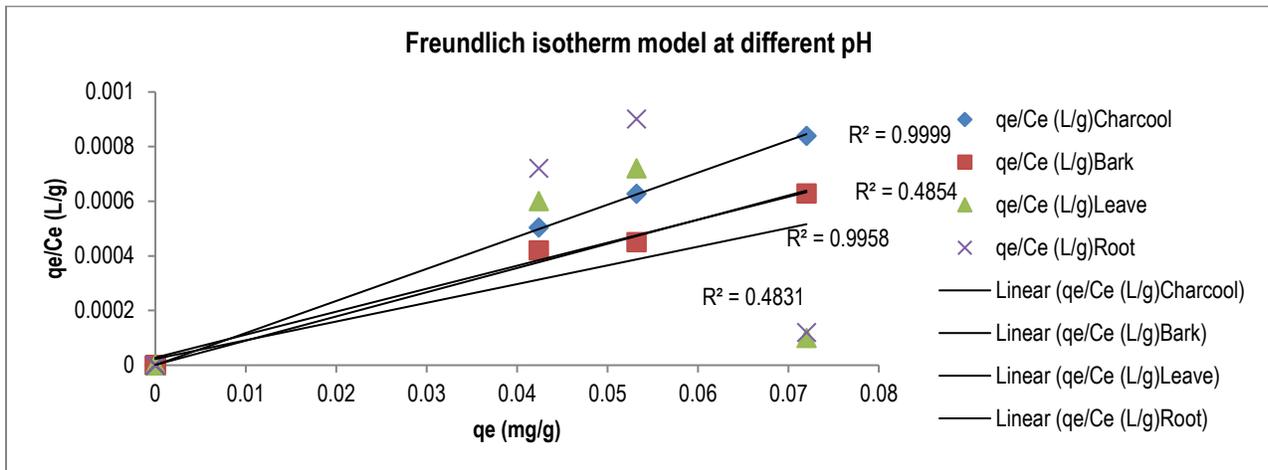


Figure 6. Linear model of Freundlich isotherm based on pH

Figure 7 shows the linear fitness of $\log q_e$ against $\log C_e$ based on adsorbent dose with a correlation coefficient of ($R^2 = 0.4689$ to 0.9884) indicates that the adsorption of dyes on charcoal, bark, leaves and root are positively correlated and

strongly obey Freundlich adsorption isotherm. The optimum adsorbent dose was found to be $7.5\text{g}/100\text{ml}$ with a R^2 value of 0.9884 (Sheba *et al.*, 2016; Vankatesan *et al.*, 2016; Edet *et al.*, 2020).

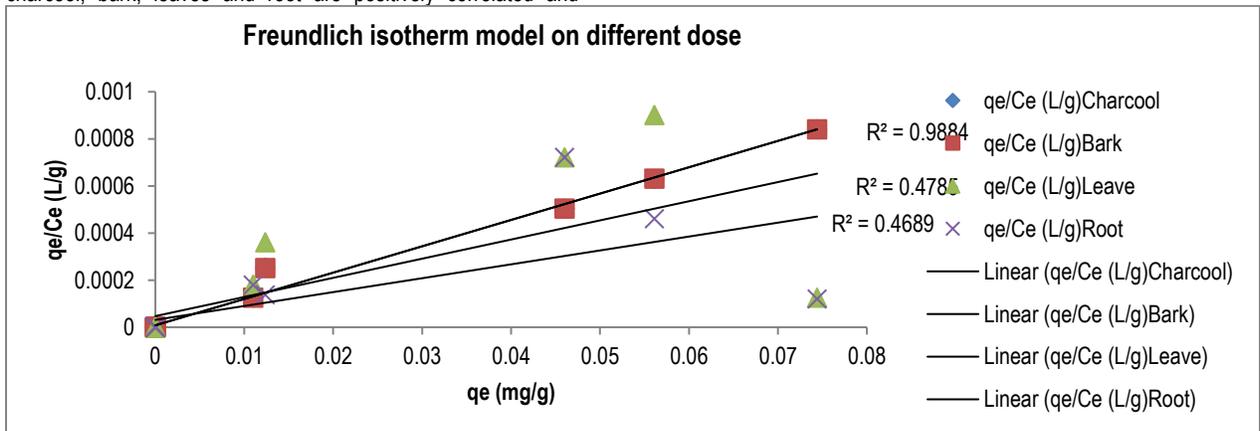


Figure 7. Linear model of Freundlich isotherm based on adsorbent dosage

Figure 8 shows the linear fitness of $\log q_e$ against $\log C_e$ based on contact time with a correlation coefficient of ($R^2 = 0.4677$ to 0.999). This indicate that the adsorption of dyes on charcoal, bark, leaves and root are positively correlated and

strongly obey Freundlich adsorption isotherm. The optimum contact time was found to be 72 (Hrs) with a R^2 value of 0.999 (Sheba *et al.*, 2016; Vankatesan *et al.*, 2016; Edet *et al.*, 2020).

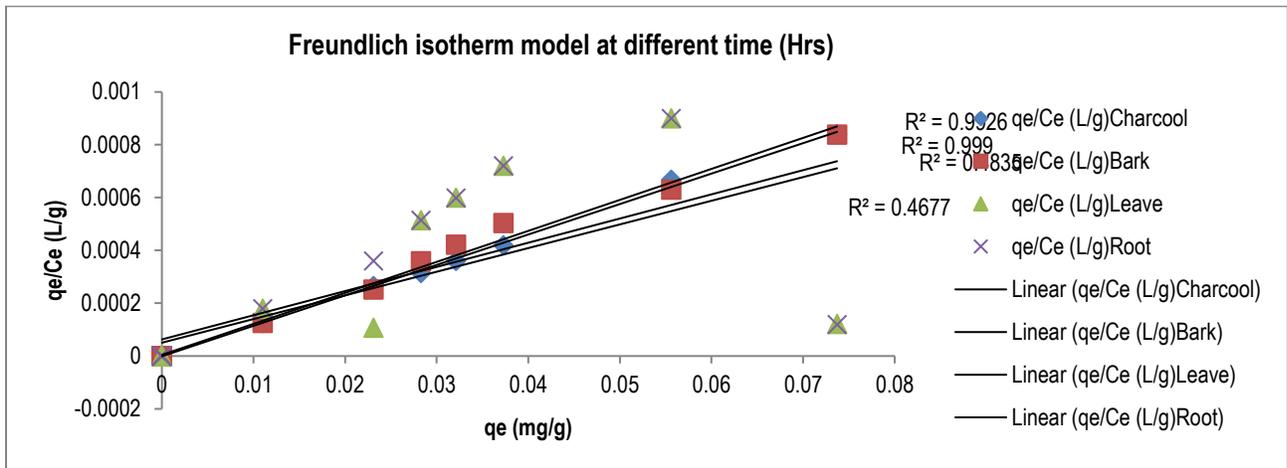


Figure 8. Linear model of Freundlich isotherm based on time contact.

Langmuir adsorption isotherm.

This portrays quantitatively the formation of a monolayer adsorbate on the outer surface adsorbent and after that no further adsorption takes place. Thereby the Langmuir represents the equilibrium distribution of metal ions between the solid and liquid phases. The Langmuir isotherm is valid for monolayer adsorption onto the surface and no transmigration of adsorbate in the plane of the surface (Obaid *et al.*, 2020; Paudel *et al.*, 2020; Saturaki *et al.*, 2015). Therefore, the Langmuir isotherm linear model is given by:

$$C_e/q_e = 1/(q_m b) + C_e/q_m \quad (6)$$

Figure 9 shows a linear plot of C_e/q_e Vs $1/q_e$ Vs $1/C_e$, q_e Vs q_e/C_e and q_e/C_e Vs q_e or $\ln q_e$ against $\ln C_e$.

They are applicable to Langmuir isotherms with a correlation coefficient of R^2 (0.9264 – 0.9349). The optimum temperature for isotherm was found to be 15°C with a R^2 value of 0.9349. The R^2 values indicate that temperature is strongly positively correlated and its obey Langmuir isotherm (Soumya *et al.*, 2018; Musa *et al.*, 2018).

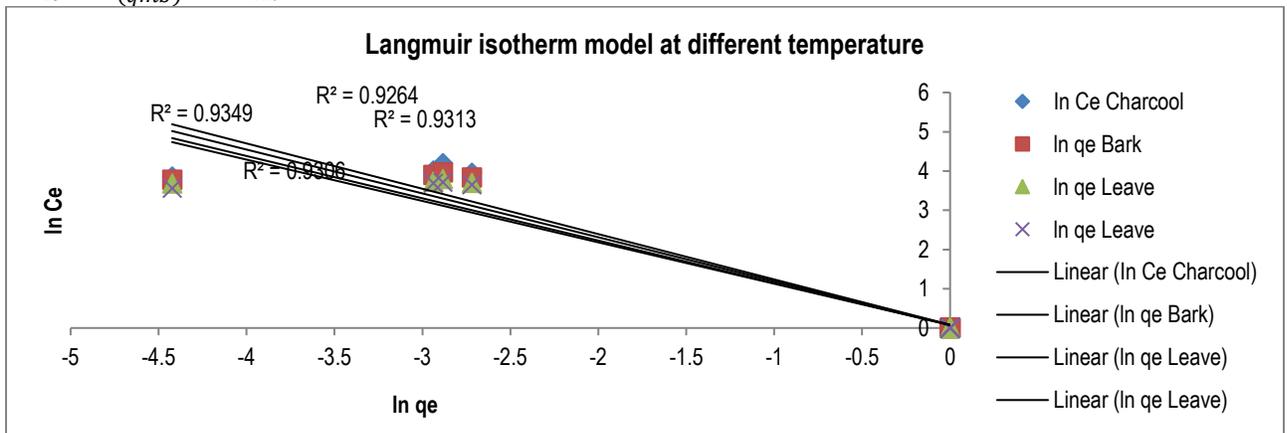


Figure 9. Linear model of Langmuir isotherm based on temperature

Figure 10 shows the plot of $\ln q_e$ against $\ln C_e$. They are applicable to Langmuir isotherms with a correlation coefficient of R^2 (0.9745 – 0.9919). The optimum pH for isotherm was found to

be (7) with a R^2 value of 0.9919. The R^2 values indicate that pH is strongly positively correlated and it obey Langmuir isotherm (Khote *et al.*, 2019; Natrayan *et al.*, 2022; Pobocikoya *et al.*, 2018).

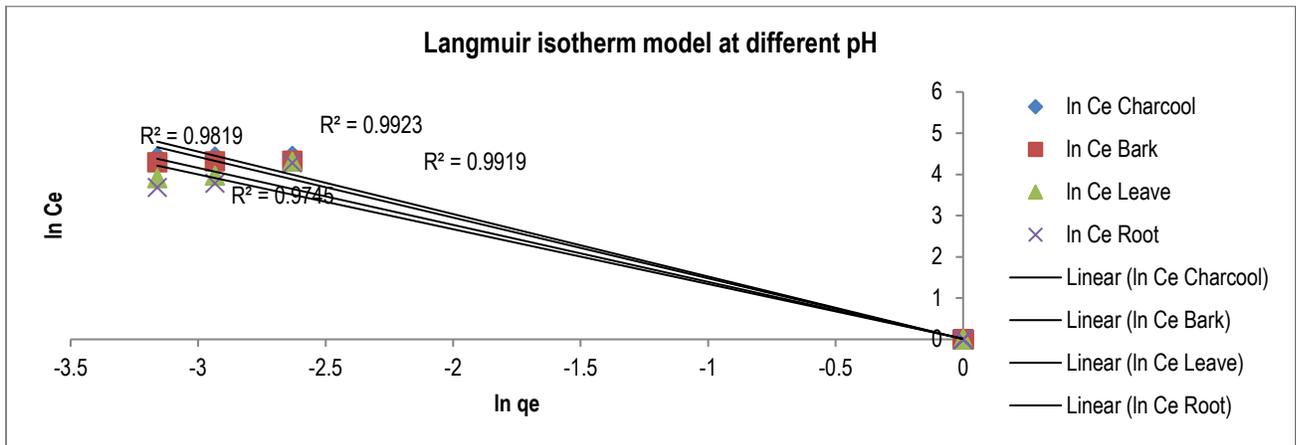


Figure 10. Linear model of Langmuir isotherm based on pH

Figure 11 shows the plot of $\ln q_e$ against $\ln C_e$. They are applicable to Langmuir isotherms with a correlation coefficient of R^2 (0.9139 – 0.9197). The optimum adsorbent dosage for isotherm was found to be (7.5g/100ml) with a R^2 value of 0.9197. The

R^2 values indicate that adsorbent dosage is strongly positively correlated and it obey Langmuir isotherm (Khote *et al.*, 2019; Natrayan *et al.*, 2022; Pobocikoya *et al.*, 2018).

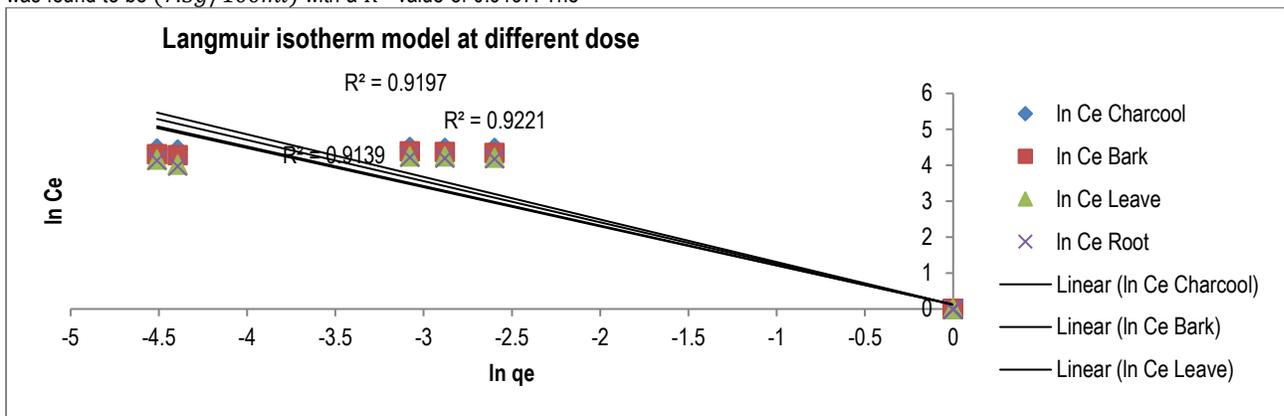


Figure 11. Linear model of Langmuir isotherm based on adsorbent dosage

Figure 12 shows the plot of $\ln q_e$ against $\ln C_e$. They are applicable to Langmuir isotherms with a correlation coefficient of R^2 (0.9513 – 0.9496). The optimum contact time for isotherm was found to be (72Hrs) with a R^2 value of 0.9496. The R^2 values

indicate that contact time is strongly positively correlated and it obey Langmuir isotherm (Khote *et al.*, 2019; Natrayan *et al.*, 2022; Pobocikoya *et al.*, 2018 and Manga *et al.*, 2021).

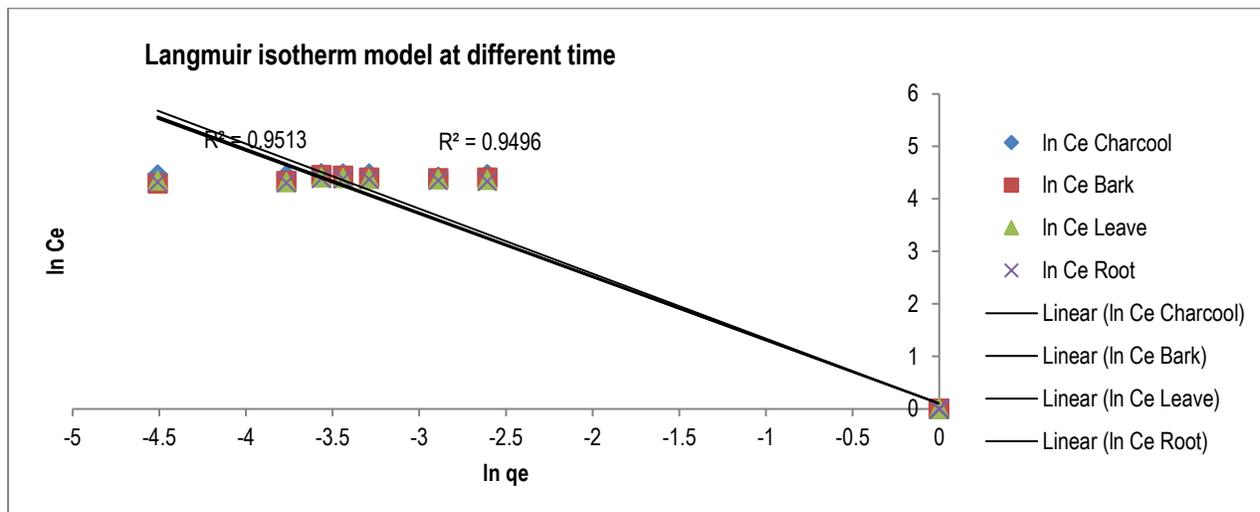


Figure 12. Linear model of Langmuir isotherm based on contact time Both isotherm models are well fitted with the equilibrium data, which described less error. Another important point indicated is that the best temperature for isotherm is 15°C.

CONCLUSION

In this study, we find out the optimum bio-adsorbents in the removing color in textile industry wastewater. Two isotherm models were developed based on Langmuir and Freundlich isotherms. The results of Root Mean Square Error (R^2) for assessing the linear relationship between the two models which are Langmuir and Freundlich models were given as, for pH (0.9745 – 0.99919), (0.4831 – 0.9999) for Adsorbent Dose is (0.9034 – 0.9221), (0.4689 – 0.9884) for contact time (0.9494 – 0.9513), (0.4677 – 0.9990) and temperature (0.9264 – 0.9349), (0.3755 – 0.5849) respectively. The ability of color removing at a pH 5 from bio-adsorbents in a textile industry wastewater on various adsorbent dosage, various contact time, various pH and various temperature were properly observed during the study.

The following **conclusions** can be drawn from the observed results:

1. The maximum efficiency of *Azadirachta Indica* (Neem) for charcoal, bark, leave and root on dye removal were obtained at an optimum pH of 5, optimum contact time of 110Hours, optimum temperature of 15°C and optimum dosage of 25g/100ml.
2. The Langmuir and Freundlich isotherm linear model were found to be best fits isotherm model.
3. Finally, the tendency for bio adsorbents for removing colour at a pH 5 in a textile industry wastewater by various adsorbent dosage, various contact time, various Temperature and various pH on charcoal, bark, leave and root from aqueous solution is found to be cheaper. It can be used as a cheap substitute for commercial adsorbent for the removing of dye from textile wastewater for a better environment.

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