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Facile and efficient strategy for removal of reactive industrial dye by using tea waste

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ABSTRACT

The present study deals with the study tea waste as an effective functional adsorbent for the removal of reactive blue 21(RB21) from aqueous solutions. The batch adsorption experiments were influenced by several parameters such as contact time, solution pH and mass of adsorbent and initial dye concentration. The results indicated that the maximum removal of RB21 dye by tea waste was after 90 minutes of contact time and at the nearly neutral pH of 6.0. Moreover, two isotherm models, namely Langmuir and Freundlich adsorption isotherm were used to describe the adsorption equilibrium of RB21 dye onto tea waste. The data obtained was in good agreement with Langmuir model than that of Freundlich model, showing a monolayer adsorption capacity of 28.99 mg/g. Furthermore, it was observed that tea waste did not require any supplementary pre-treatments; such as activation before the application. Collectively, this work highlights the promise of pristine TW, ability to harness it for adsorption of dyes, while also prioritizing areas for future research and development (e.g., collection of TW from local vendors and resulting in environmental friendly disposal of the same). The presented strategy exhibited excellent adsorption capacity of TW for the removal of RB21 from the aqueous solutions. Nonetheless, in most parts of the globe TW from local vendors is available without any cost, regeneration is not required and the saturated adsorbent can be disposed by incineration.

Keywords: Tea waste; adsorbent; isotherm models.

Introduction

In recent decades, the use of different commercially available synthetic dyes and pigments has been extensively used in industries (e.g., textile, leather, paper, cosmetics, pharmaceutical and food industries) to impart color in the fabricated products. Apparently, in developing Asian countries an array of structurally diverse, synthetic and/organic chemical wastes (e.g., from industries and unplanned cities) are abundantly used and discharged to the waterbodies without proper treatment. Further, these contaminants are translated into drinking water where their removal proves challenging with the conventionally available treatment approaches. All the aforementioned industries consume large amount of water for maintaining their high production rate. This in turn leads to discharge the bulk amount of wastewater every year [**1**]. The discharge of wastewater can result in water pollution and can directly deteriorate the water quality through damaging aquatic plants, microbes and fish species, reducing sunlight penetration [**2-3**], depleting dissolved oxygen (DO), increasing biological oxygen demand (BOD) and chemical oxygen demand (COD) [**4**]. Moreover, the colored wastewater may contain chemicals which are not only toxic but also are carcinogenic, mutagenic and teratogenic to humans and aquatic life [**5-7**]. Therefore, it is essential to remove the dyes and pigments from industrial effluents prior to discharge to a waterbody.

Reactive dyes are characteristically azo-based chromophores and are coupled with different types of reactive groups. Currently, reactive dyes are extensively used in textile industries to color cotton through covalent bond formation. However, sometimes are also used to impart color in wool and nylon fabrics. Moreover, due to their characteristic bright color, low energy requirement and easy processing techniques employed when used; makes them advantageous over other direct dyes. Therefore, they are rigorously used in most of the textile industries. Thus, the removal of these reactive dyes from industrial wastewater is of great importance for protecting the natural environment [**8**].

Advanced technologies (e.g., microfiltration, solvent extraction, electrochemical treatment, coagulation, polymeric flocculation, adsorption, nano-filtration, chemical precipitation, electrochemical de-colorization, membrane separation, and reverse osmosis) are well-suited for the removal of these so-called emerging pollutant classes. Therefore, numerous conventionally available physical, chemical and biological methods have been used for the treatment of these dyes. For example, microfiltration [**9**], solvent extraction [**10**], electrochemical treatment [**11**], coagulation [**12**], polymeric flocculation [**13**], adsorption [**14**], nanofiltration [**15**], chemical precipitation [**16**], electrochemical decolorization [**17**], membrane separation [**18**], and reverse osmosis [**19**], are commonly employed. In this connection, ZnO nanoparticles have been fabricated by microwave technique for rapid decolorization of Congo red dye [**20**]. Similarly, the lanthanum doped bismuth ferrite nanoparticles were also investigated for photocatalytic properties [**21**]. All these methods have some limitations, for instance, toxic sludge production, economic constraints, time consuming, incomplete ion and/or dye removal. Besides, the residual dyes in sludge are non-biodegradable in nature and remain unchanged for a longer period. Among all the previously mentioned techniques, adsorption has been reported as a well-known equilibrium separation process [**22-23**]. Furthermore, adsorption is considered as an effective methodology for the removal of dyes and hazardous organic compounds from textile effluents [**24**]. Recently, commercially activated carbon (CAC) is the most commonly employed adsorbent by the textile industries. Due to its large surface area along with high sorption capacity, makes it an effective adsorbent against hazardous organic and inorganic pollutants. However, despite having these benefits, its extensive use is limited because of its high-cost and difficulties in regeneration [**25-27**]. Therefore, many researches have been conducted for intensive studies to figure out a suitable and low-cost adsorbent which can replace the CAC. To obviate this barrier, numerous low-cost adsorbents have been tested for the dye removal. For instance, agricultural waste such as bagasse, fruit peels (e.g., banana and orange), coir pith, peanut hulls, rice by-products (e.g., straw and husk), saw dust, sepiolite, etc [**28**]. Additionally, in order to accelerate the capabilities of natural agricultural wastes few of the modifications had been suggested. This includes the graft copolymers using cheap potato starch and methacrylic acid for removal of metal ions and organic dyes from aqueous system [**29**]. Similarly, polysaccharide guar gum was hybridized with silica which resulted in water insoluble porous material [**30**]. This resulted in high surface area capable of removing cadmium (II) from aqueous solution [**31**]. The coconut coir dust as a low-cost adsorbent was used for the removal of methylene blue [**32**]. The removal of metal contaminants from chemically modified rice husk was also exploited [**33**]. The pineapple stem (i.e., waste generated from agricultural waste) was used as an adsorbent for the removal of hazardous dye methylene blue from its aqueous phase [**34**]. It is noteworthy in mentioning that synthetic materials using nano-engineered materials were used to removal of dyes from industrial effluents [**35**]. These nano-engineered techniques display a challenge of having high-cost and difficulty in availability. However, none of studies have attempted to investigate the utility of low-cost tea waste from local tea vendors for efficient removal of industrial dyes. Tea waste is comparatively cheap and easily available in developing Asian countries.

In the present adsorption studies, using tea waste (TW) has been employed as adsorbent for the removal of model dye as reactive blue 21 (RB21) from its aqueous solution. Previously, TW was used as adsorbent for acidic and basic dyes [**1-2**]. Moreover, TW was also used as adsorbent for the removal of reactive dyes from textile waste [**36-37**]. The low-cost potential of TW as adsorbent for the removal of azo-dyes was much more appreciated [**38-39**]. The alkali treated foumanat TW was used as an efficient adsorbent for methylene blue adsorption and the TW utilized was obtained from agriculture biomass [**40**]. TW is considered as a low-cost adsorbent because its availability is very high in the developing Asian countries. Moreover, its disposal is another problem related to solid waste management in the developing countries. So, utilization of such waste for removal of RB 21 will help to some extent in waste disposal problem alongside serving as a low-cost adsorbent without regeneration. Furthermore, it has functional groups like -COOH, -OH, etc. Due to the presence of structural hydroxyl protons, carboxylic protons and lactone protons in TW are able to adsorb dyes by its ion-exchange behavior [**6**]. Although the utilization of different forms of TW is recognized previously to adsorb the few dyes [**41-46**]. However, the influence of TW on degradation of RB 21 has not yet been investigated. Moreover, the previous studies used TW from agriculture biomass, which is comparatively expansive than the TW from local tea vendors. Also, utilizing the TW from tea vendors can boost in earning of their livelihood and encourage the resourceful disposal of solid waste. Further, to best exploit the known, high sorption capacity of TW toward dye compounds, we explored the cost effective methodology to investigate the catalytic properties of TW from local tea vendors. Our goal herein was to identify the optimal TW material for exhibiting sorption capacity and optimum parameters for removal of RB21. Accordingly, we evaluated the optimum concentration, influence of pH, mass of adsorbent, addition, two adsorption isotherm models such as Langmuir and Freundlich adsorption isotherm models were used to explain the adsorbent characteristics. In this study, the influence of several parameters such as contact time, pH, adsorbent mass and initial dye concentration while treating with TW were studied under equilibrium condition. Equilibrium isotherm data were plotted in Langmuir and Freundlich equations and constants of isotherm equations were determined for further confirmations.

Materials and Methods

Reactive Blue 21 (RB21) used in this study was obtained from Dystar (Kocaeli, Turkey) for the adsorption experiment. The chemical structure and characteristics of RB21 are shown in (**Fig. 1** and **Table 1**), respectively. Furthermore, all the chemicals used for adsorption study were of analytical grade and were used directly without purification.

Characterizations

UV-vis spectrophotometer (DR/4000U Spectrophotometer, HACH, USA) was used to measure the dye concentration. The pH measurements were obtained using a portable multiparameter meter (Sension 156, HACH, USA). Magnetic stirrers were purchased from Alabtech lms-1003 (Republic of Korea) used for all adsorption experiments.

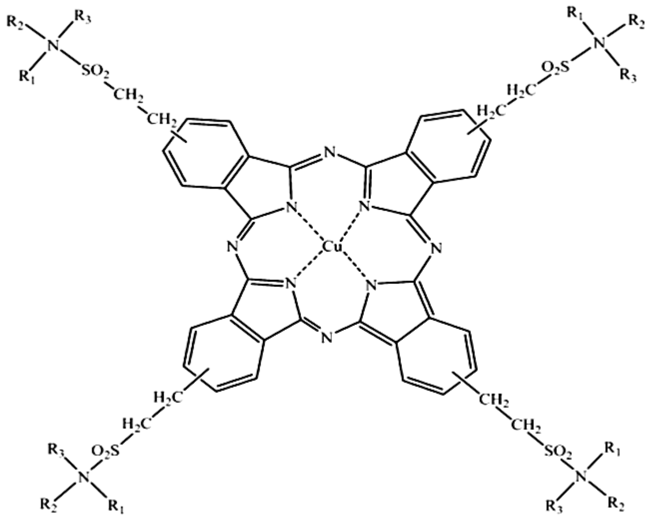
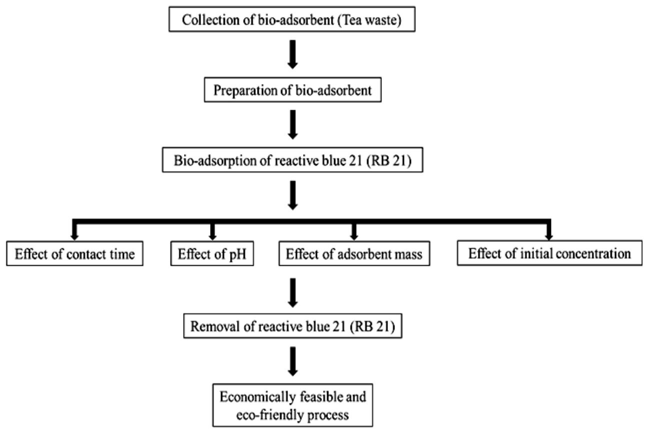
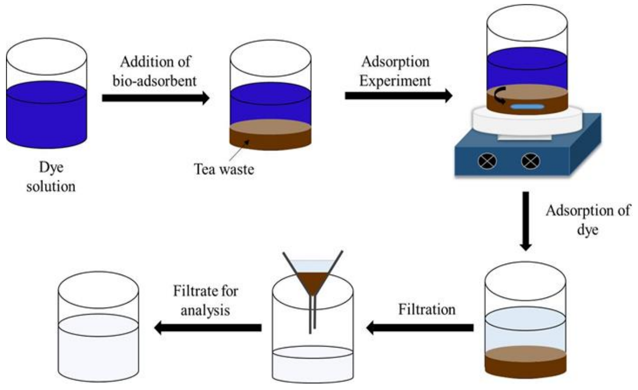


Fig. 1. Molecular structure of Reactive Blue 21 (RB21).



Scheme 1. Flow diagram for the used strategy using tea waste (TW) for dye adsorption.



Scheme 2. Schematic diagram of the dye adsorption analysis.

Procedure

The TW collected from local tea vendors was used as adsorbent for removal of RB21 from aqueous solutions in this study. Among all the adsorbents used to remove dye contaminants, the TW can be considered as economically feasible as it originates from wastage byproduct from local tea vendors in developing Asian countries. The (**Scheme 1** and **Scheme 2**) illustrates the possible strategy to treat dye solutions using TW from local tea vendors. In order to have a proper blend of different tea brands, the TW were collected from at least 10 local tea shops. In brief, the preparation of adsorbent was performed by boiling the TW in water at (80 °C) for 1 h for the removal of hydrolysable tannins and other residual colored components. After that, the decolorized and cleaned TW was dried in over at (80 °C) for 24 h. Then, the dried material was grinded to a fine powder using electrical grinder. The fine powder was washed with distilled water for several times until the used water appeared colorless with neutral pH and negligible conductance. Generally, a colorless solution of TW was observed after at least 5 washing cycles. Finally, the adsorbents were packed in hermetically sealed containers, labeled and stored in refrigerator for further use. The preparation of dye standard solution was formulated using a stock solution of reactive blue 21 (RB21) by dissolving a mixture of 0.1 g of dye in a 1000 mL volumetric flask, followed by diluting in de-ionized water. Dye test solutions were prepared through proper dilution of the stock solution to the desired concentration. De-ionized water was used to prepare all of the solutions in this study. The dye solutions were prepared by stirring them at room temperature and filtered prior to use.

Table 1. Chemical characteristics of reactive blue 21 (RB21).

Parameter	Dychufix Turquish Blue G
Class	Copper phthalocyanine
Color index name	Reactive Blue 21
Reactive group	Sulphatoethylsulphone
pH	5.12
COD (mg/l)	46.93
TOC (mg/l)	18.62
λ_{max} (nm)	626
Molecular weight (g/mol)	576.10

Adsorption experiments

Adsorption studies were performed in batches at room temperature (i.e., 25 °C). All the batch experiments were performed in triplicate and the results are the means with a relative standard deviation of less than 5 %. In addition, a control experiment without adsorbent was carried out simultaneously to ensure that adsorption in the samples was done by the adsorbent. After the desired contact period, for each batch experiment, the aqueous phases were separated from the materials. Thereafter, the dye concentration in the supernatant was measured using an UV-Vis spectrophotometer. The following adsorption experiments were carried out in batches:

The first batch was investigated in these studies were for the effect of contact time. The contact time ranged from 30 to 240 minutes. The initial concentration of dye was 20 mg/L, and the initial solution pH was 6 (adjusted using HCL and NaOH) and adsorbent dose was 1 g/100 ml. The second batch investigated was for studying the effect of pH.

The pH ranged from 2 to 14. The initial dye concentration was 20 mg/L. The adsorbent dose was 1 g/100 ml. This batch operated at optimum contact time resulted from previous batch (i.e., batch 1). The (batch 3) used to study was for the effect induced by adsorbent mass. The adsorbent dose ranged from 0.5 to 5 g/100 ml at optimum contact time from (batch 1) and optimum pH from (batch 2). The initial concentration was 20g/L. The (batch 4) consisted for studying the effect of initial concentration. The concentration varied from 5 to 30 mg/L at optimum contact time revealed from (batch 1, optimum pH from batch 2 and optimum the amount of adsorbent from batch 3), respectively. Furthermore, the (batch 5) consisted of adsorption isotherm for RB21 onto TW. Both the Langmuir and Freundlich adsorption isotherm models were employed to describe the equilibrium nature of the RB21 adsorption onto TW. In all the above analyses, the measurement of equilibrium adsorption, q_e or x/m was calculated by using the following equation:

$$q_e = \frac{(C_o - C_e)\ X\ V}{\mathrm{m\ X\ 1000}} \tag{1}$$

whereas, q_e is the amount of dye adsorbed at equilibrium time (mg/g). The C_o and C_e are the initial and equilibrium concentrations of the dye (mg/L). The V is the volume of dye solution (ml) and m is the mass of the adsorbent (g).

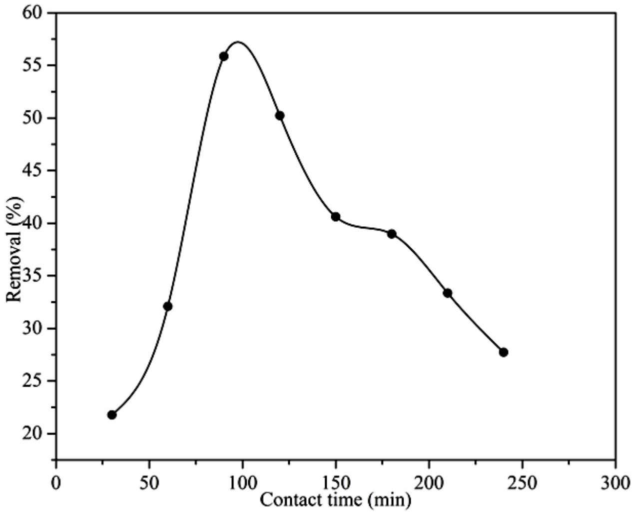


Fig. 2. Effect of contact time on the removal of reactive blue 21 (RB21) by adsorption onto tea waste (TW).

Results and discussion

The effect of contact time on the removal of the dye was studied at 6 pH and room temperature (25 °C) using conditions that were previously mentioned in (batch 1). The obtained result is presented in (**Fig. 2**) for tea waste. As expected, it was observed that removal efficiency initially increases logarithmically with an increase in the contact time. For an initial concentration of 20 mg/L, the removal efficiency increases until 90 minutes of time. A removal efficiency of over 55 % for tea waste was found after 90 minutes. Thereafter, the removal efficiency was decreased due to weak binding force between adsorbent surface functional groups and adsorbate [**44**]. Therefore, 90 minutes was chosen as best contact time for the subsequent batches.

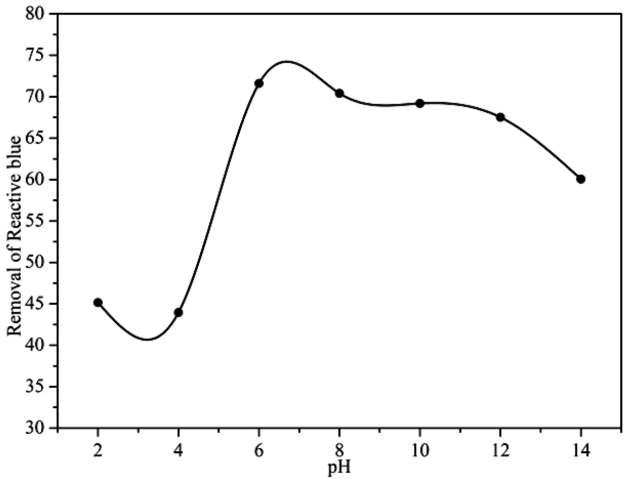


Fig. 3. Effect of pH on the removal of reactive blue 21 (RB21) by adsorption onto tea waste (TW).

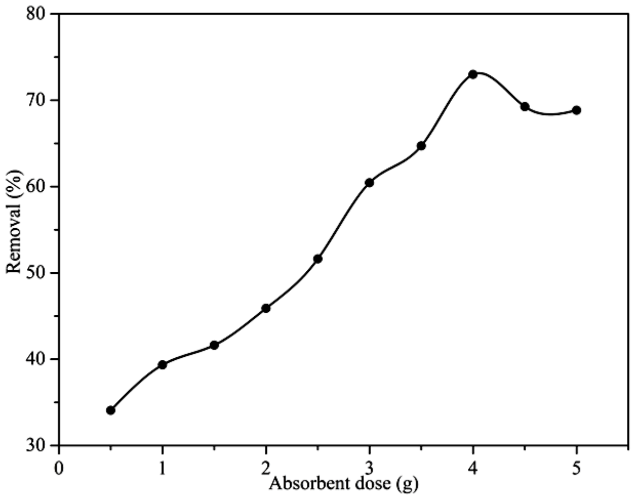


Fig. 4. Effect of amount of adsorbent on the removal of reactive blue 21 (RB21) by adsorption onto tea waste (TW).

Effect of pH

The pH of dye solution has been identified as the most important parameter affecting dye adsorption onto the bioadsorbents. The influence of pH is not easy to study, especially under strong acidic conditions. Strongly acidic pH results in formation of protonated species which possibly leads to a change the dye structure along with protonation of the negatively charged adsorbent surface sites. However, it may also be worth in mentioning that the acidic pH of a system can result in better adsorption for the dyes. In this regard, the (**Fig. 3**) shows results of effect of pH on the removal of dye. In this case, it was observed that about 71.6 % of RB21 was adsorbed at pH 6 and then decreased at higher pH. The rationale may be due of electrostatic repulsion, the highly negative charged adsorbent surface sites did not facilitate the adsorption of deprotonated RB21. Moreover, previous studies revealed that the excess OH^- ions in alkaline solution form a competitive atmosphere with anionic ions of RB21 for the adsorption sites resulted in lower removal efficiency [**47-49**]. Therefore, pH 6 was selected as an ideal pH for further experiments.

Effect of mass

The adsorption experiments were repeated at various amounts of adsorbents in the range of (0.5 to 5 g/100 ml) at the room temperature (25 °C) to study the adsorbent dose dependent behaviors of dye adsorption. In this context, the (**Fig. 4**) shows that about 73 % of RB21 were removed with (4.0g/100 ml) of TW. The adsorption capacity of an adsorbent depends on the surface activity that is the specific surface area available for dye surface interactions. However, after a specified adsorbent dose (i.e., 4g/100 ml) the equilibrium adsorption capacity of tea waste slightly decreased with increasing amount. The rationalle is due to limited dye concentration for this amount of adsorbent dosage [**50**]. Another possible reason could be the unsaturated adsorbent surface sites available during the adsorption reaction. Therefore, 4.0 g/100 ml of TW was chosen for the next batch of experiments.

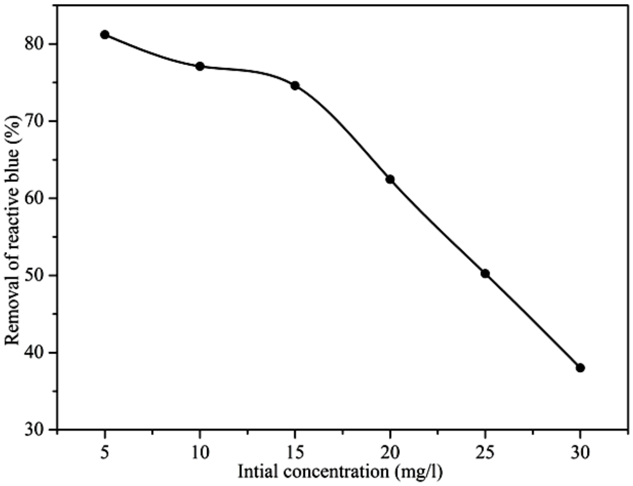


Fig. 5. Effect of initial concentration on the removal of reactive blue 21 (RB21) by adsorption onto tea waste (TW).

Effect of initial dye concentration

The adsorption of the dye, RB21 over TW, was investigated at initial concentration ranging from (5 to 30 mg/l) at the room temperature (25 °C) using the aforementioned optimum conditions. As a general trend a decrease of the removal efficiency with the increase in dye concentration in the solution is evident from (**Fig. 5**). This figure clearly demonstrates that the adsorption of RB21 by the TW was almost 81 % at low concentration and about 38 % at higher concentration. The same phenomena were observed in earlier studies and it was concluded, that lack of active sites on the adsorbent together with increasing competition for the available binding sites among the ions, resulted in lower removal efficiency at higher concentrations [**51**]. Our findings are in complete agreement with results from these previous findings. Furthermore, from the above observation, it is evident that more dyes were left as un-adsorbed in solution at higher concentration levels.

Adsorption isotherms

The adsorption isotherm is important for the understanding of adsorbent behavior between the liquid phase and the solid phase, once the system reaches an equilibrium state. Among several adsorption models, in our study, we used two classical models, namely Langmuir and Freundlich adsorption isotherm models. These adsorption isotherm models were used to describe the equilibrium of adsorbed dye (C_e) at room temperature (25 °C) and a constant pH (pH 6).

Langmuir adsorption isotherm model

In this case, the adsorption isotherm model is based on the assumption that specific identical sites of the adsorbent actively participate in the adsorption process [**52**] and that once a dye molecule adsorbed at the site, no other molecule can be adsorbed at the site on the same time. Moreover, this model is mostly used in monolayer sorption. The Langmuir equation is given by the following relation [**53**]:

$$q_e = \frac{q_m K_a C_e}{1 + K_a C_e} \qquad (2)$$

The linearized form of Langmuir isotherm that can be written as:

$$\frac{1}{q_e} = \frac{1}{K_a q_m} \cdot \frac{1}{C_e} + \frac{1}{q_m} \qquad (3)$$

where, q_e is the amount of dye adsorbed at equilibrium time (mg/g), C_e is the equilibrium concentration of dye in solution (mg/L), q_m is the maximum adsorption capacity (mg/g) and K_a is the isotherm constants for Langmuir (L/mg). Langmuir constants, q_m and K_a are related to maximum adsorption capacity and energy of adsorption through Arrhenious equation, respectively [**54**]. Further on, q_m can also be interpreted as the total number of binding sites that are available for sorption. When $1/q_e$ is plotted against $1/C_e$, a straight line with slope $1/K_a q_m$ is obtained and intercept is corresponds to $1/q_m$.

Table 2. Langmuir and Freundlich isotherm model constants for the sorption of reactive blue 21 (RB21) onto tea waste (TW).

Langmuir isotherm model constants			Freundlich isotherm model constants		
R^2	q_m (mg/g)	K_a (L/mg)	R^2	n	K_F [mg/g(L/g)$^{1/n}$]
0.71	28.99	1.8×10^{-3}	0.69	0.81	3.3×10^{-2}

Fig. 6 demonstrates experimental data and the linear form of the Langmuir isotherm (Eq. 3). The sorption constant, K_a and the saturated monolayer sorption capacity, q_m onto TW shows good result. The linear isotherm constants, q_m, K_a and R^2 are also indicated in (**Table 2**). Therefore, it ratifies that the Langmuir isotherm is followed in the adsorption process.

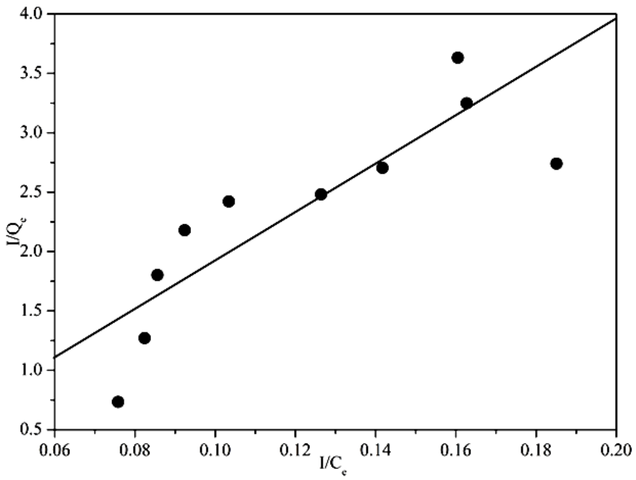


Fig. 6. Langmuir isotherm of reactive blue 21 (RB21) adsorbed onto tea waste (TW).

For further confirmation, the important characteristics of the Langmuir isotherm were expressed in term of a dimensionless constant separation factor for equilibrium parameter, R_L. The separation factor, R_L can be determined from Langmuir equation by using the following relation:

$$R_L = \frac{1}{1 + K_a C_o} \tag{4}$$

where, R_L values indicate the type of adsorption to be irreversible (RL=0), favorable ($0<R_L<1$), linear ($R_L = 1$) or unfavorable ($R_L>1$), K_a is the Langmuir constant and C_0 is the initial RB21 dye concentration (mg/l). Moreover, the results of R_L presented in (**Table 3**), indicate obtained R_L values were in the range 0-1. Thereby, it confirms that the TW is favorable for RB21 dye adsorption under the studied conditions.

Table 3. Intraparticle diffusion rate parameter and diffusion coefficient at different initial dye concentration.

Initial dye concentration (mg/l)	R_L
5	0.99
10	0.98
15	0.97
20	0.96
25	0.95
30	0.94

Freundlich adsorption isotherm model

The Freundlich adsorption isotherm model is a multilayer adsorption model. It is based on a physical hypothesis that a heterogeneous adsorption surface possesses unequal available sites with different energies of adsorption. The Freundlich adsorption isotherm model is represented as follows [**55**]:

$$q_e = K_f C_e^{1/n} \tag{5}$$

The linearized form of Freundlich isotherm that can be written as:

$$\ln q_e = \ln K_f + \frac{1}{n} \ln C_e \tag{6}$$

where q_e is the amount of dye adsorbed at equilibrium time (mg/g), C_e is the equilibrium concentration of dye in solution (mg/L), K_f is the capacity of the adsorbent and n is the adsorption constant intensity. The plot of ln q_e versus ln C_e is used to determine the K_f and n from both intercept and slope. In general, the correlation coefficient (R^2) value from the linear regression elucidates the information on which model can be selected to give the best-fit.

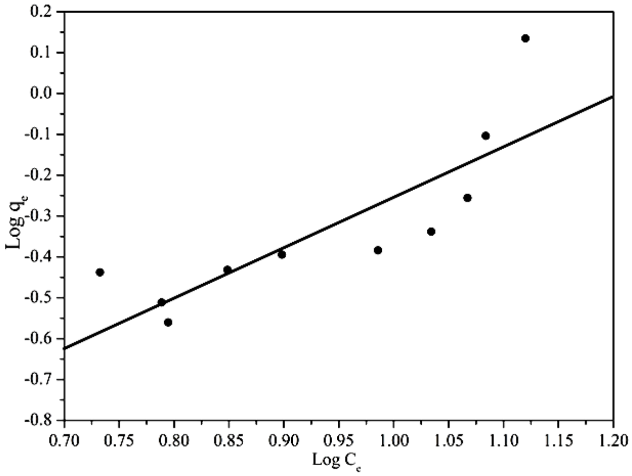


Fig. 7. Freundlich isotherm of reactive blue 21 (RB21) adsorbed onto tea waste (TW).

The linear Freundlich isotherm for the adsorption of the RB21 dye onto TW is presented in (**Fig. 7**). Examination of the plot suggests that the linear Freundlich isotherm is also a good model for the adsorption of RB21 dye. The (**Table 2**) shows the linear Freundlich sorption isotherm constants, K_f and n, and the correlation coefficient (R^2). Based on the R^2 value, the linear form of the Freundlich isotherm model appears to produce a reasonable model for adsorption.

Conclusion

In conclusion, this study demonstrates that TW can be effectively utilized as a functional adsorbent for the removal of reactive blue 21(RB21) from aqueous solutions. The batch adsorption experiments were influenced by several parameters such as contact time, solution pH, mass of adsorbent and initial dye concentration. The maximum removal efficiency of RB21 dye by TW was observed after 90 minutes of contact time and at an initial pH of 6.0. The removal efficiency decreased with increasing the dye concentration and increasing dose of adsorbent. In addition, two isotherm models, namely Langmuir and Freundlich adsorption isotherm were used to describe the adsorption equilibrium of RB21 dye onto TW. The data were in better agreement with Langmuir model than Freundlich model, showing a monolayer adsorption capacity of (28.99 mg/g). Moreover, TW is easily and profusely available, do not require supplementary pretreatment steps such as activation before applications and shows good removal efficiency for RB21 from aqueous solutions. However, an appropriate method for collection and preservation of tea waste should be considered. Further, experimental works are required on desorption and regeneration of the adsorbent and application of the adsorbent for industrial effluents. However, an appropriate method for collection and preservation of tea waste should be considered.

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