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Textile dyes as a major problem for the environment: Comparative removal of the highly toxic textile dyes from aqueous solution

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ABSTRACT

Environmental pollution is one of the most important and main problems that the world is facing today. Global production and use of chemical compounds, many of which are resistant to biodegradation, have increased significantly in recent decades. These compounds enter the environment after consumption, and therefore, it is necessary to provide solutions to reduce consumption and remove pollutants from the environment. Removing the textile dyes of crystal violet (CV) and methylene blue (MB) from aqueous solution by CNT/TiO_2 surface was studied. We considered several physio-chemical parameters for example, contact time, primary concentration dye (5-50 mg L^{-1}), adsorbent amount (0.01-0.1 gm), as well as temperature (15, 35, and 50 °C) in technique batch-adsorption. The output appeared absorbing two favorable dyes MB, CV at the basic pH. Moreover, removal present elevated by the increased amount of adsorbent, while decreased by uprising the primary concentration of dye, and agitation time bath of the two dyes. The adsorption capacity (Qe) upraised by the increased contact time and initial concentration of dyes, and then declined by the adsorbent weight and the procedure of temperature. The equilibrium data were estimated utilizing Freundlich and Langmuir isotherms. The Freundlich isotherm describes the best uptake of MB and CV dyes. The adsorption of CV and MB in the present study on the CNT/TiO_2 was heterogeneous with multi-layers. The results have shown immense potential, and it was found that the correlation coefficient value deals with the dyes, while the Langmuir equation did not exhibit good correlation of dyes.

Keywords: Environmental pollution, Chemical compounds, Textile dyes, Methylene blue, Crystal violet.

INTRODUCTION

Industrial wastewater is one of the most important sources of environmental pollutants so that sometimes the concentration of COD and BOD and other compounds in them reaches tens of thousands of milligrams per liter. Colored effluents, such as textile and dyeing industry effluents, are one of the most important industrial effluents that have created environmental problems in a wide range of environments. The textile and dyeing industry is one of the largest consumers of water. In these industries, different effluents will be produced due to the variety of production methods that are very different in terms of quantity and quality. Dyeing processes with the entry of dyes into wastewater are the main and most important cause of pollution in the wastewater of the textile and dyeing industries (Mahmood *et al.* 2005; Banimahd Keivani 2018).

In recent years, pollution of water as main universal trouble has been severely growing, because of the rise expansion of new industry separate assortment of wastewaters (Schwarzenbach *et al.* 2006; Shannon *et al.* 2010; Tao & Xin 2014; Abdulrazzak *et al.* 2016). Among the several pollutants, the contaminants dye tuff, because of their strong toxicity, unstable biodegradation, rise absorbance light, as well as suppression the aqueous organisms growth, show an important danger to humans, animal, and plants, over the earth (Khatri *et al.* 2015). To date, to remove dye injurious matters from wastewaters, a large amount of technology was employed such as adsorption

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(Yagub *et al*. 2014; Aljeboree 2016; Aljeboree & Alshirifi 2018), biological ways (Rai *et al.* 2005), precipitation of the chemical (Gupta *et al.* 2012), technology membrane (Salman *et al.* 2016), chemical oxidation /reduction (Alrobayi *et al.* 2017), and electrochemical system (Saravanan *et al.* 2015). Between them, absorption has been considered to be one of the utmost popular technologies and capable due to the respective operational ease and cost efficiency, specifically in the advanced treatments (Martínez-Huitle & Brillas 2009). TiO_2 stands out among the utmost impetuses of the encouraging because of its easy access, tall-term stability as well as non-poison (Abdulrazzak *et al*. 2016). Thus, recombination charge as a base encourages a low quantum output of TiO_2. To locate this problem, numerous methodologies have been proposed to get better photo-activity of TiO_2, such as loading with metal noble (Alkaim *et al*. 2013; Aljeboree & Alshirifi 2018) or examining the CNTs as material coupling with TiO_2 (Chai *et al.* 2013). According to several scientific field, carbon nanotubes (CNTs) have registered huge discernment because of the respective unrivaled chemical, structural, electrical properties, and thermal, are utilized in our nanoparticle design framework. Different studies explored the usefulness of coupling TiO_2 to C materials, such as CNTs, as one of the efficient methods for preventing the collecting of particles oxide, resulting in the greater photo-catalytic oxidation rate of contaminants. The present study aimed at assessing the fixed capacities of the adsorption textile CV and MB dyes via the prepared CNT/TiO_2. In addition, the effects of agitation time, primary concentration dye, weight dosage, temperature solution on the static adsorption of the dyes on CNT/TiO_2 were examined.

MATERIALS AND METHODS

Crystal violet (CV) [λmax = 580 nm] and methylene blue (MB) [λmax = 663 nm] were attained from Hilla Textile factory. The chemical structures of CV and MB are depicted in Fig. 1. The dyes concentrations were limited through understanding absorbance at wavelength feature utilizing (UV-visible spectrophotometer, Shimadzu 1650). We contrived the calibration curves among absorbance and the solution of concentration dyes.

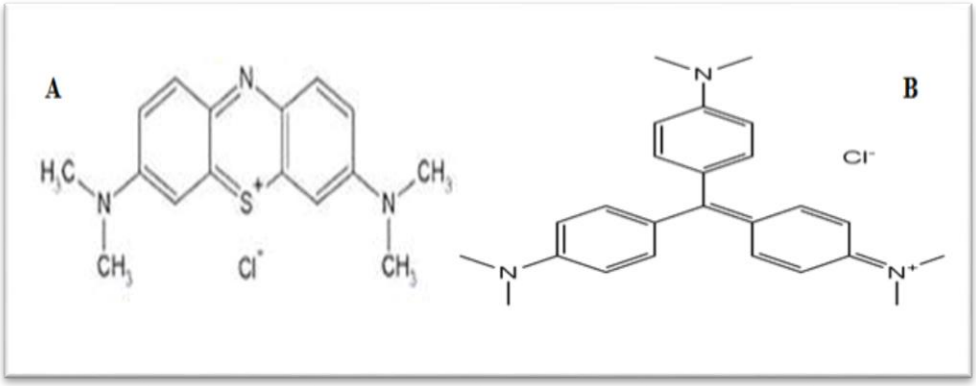

Fig. 1. Chemical structure of (A) MB and (B) CV.

Preparing the CNT-loaded TiO_2

In this stage, we procured the CNT-loaded TiO_2 via suspension of 0.5 g powder of TiO_2 in 100 mL DW via sonication, followed by adding desired amount of xg of CNT, separating the powders CNT-loaded via centrifuge, rinsing four times with the water deionized, and then drying in a furnace at 60 °C overnight.

Adsorption experiments

Removal from aqueous solution for MB and CV, kind of the adsorption experiments was implemented to estimate and relate the capacity of nanoparticles CNT/TiO_2. The MB and CV standard solutions (1000 mg L^{-1}) were prepared from DW, and all subsequent experiments were made via diluting these solutions. We performed the experimentations in a shaker water bath utilizing Erlenmeyer flasks, and the conditions data was determined through preliminary tests. All the data were considered at the volume of 100 mL-solution. Initially, the very adsorbent effect was studied on about 0.01 to 0.1 gm, the solution original pH (6.5), the temperature solution of 308 K, the speed at 2600 rpm, egestion time of 24 h, and primary conc. of 20 mg $L^{-1.}$ Then, equilibrium isotherms were attained at several temperatures' solution (288, 308, and 333 K) of two dyes, the range concentration of 5 to 50 mg L^{-1}, pH 6.5, and nanoparticles CNT/TiO_2 mass of 0.05 g L^{-1}. Finally, all data of the samples were collected,

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centrifuged at 2600 rpm for 10 min. To remove the two dyes (MB and CV), phase liquid was limited through spectrophotometry at the maximum wavelength (663 nm), (580nm) at the same order. The dyes removal percentage (E %) and adsorption efficiency (q_e) was determined via Eqs. 1 and 2 in the same order:

$$E\% = \frac{Co-Ce}{Co} * 100 \quad \textbf{(1)}$$

$$Qe = \frac{Co-Ce}{W} * V \quad \textbf{(2)}$$

Where C_o refers to primary concentration dyes in liquid (mg L^{-1}), C_e stands for the concentration of the equilibrium dyes in liquid (mg L^{-1}), W represents the active carbon weight (g). Finally, V implies the solution volume (L).

RESULTS AND DISCUSSION

Effects of adsorbent amount

Fig. 2 appears adsorption of crystal violet (CV) and methylene blue (MB) as one of the functions of mass absorbent. As shown in this Fig., through enhancing the weight of CNT/TiO_2 nanoparticles, the quantity of adsorbed two dyes enhances, while the density adsorption and quantity absorbed in each unit dosage decline. Therefore, the number of available absorption active sites enhances by elevating the mass absorbent. Thus, this condition causes an increased quantity of the absorption of two dyes (Hameed *et al*. 2017; Aljeboree & Alkaim 2019). The adsorption efficiency decreases by enhancing the mass adsorbent, which is largely caused by the absorption sites remained unsaturated through absorption reaction, while the amount of the active sites obtainable of the absorption enhances by elevating the mass adsorbent (Waleed *et al.* 2020).

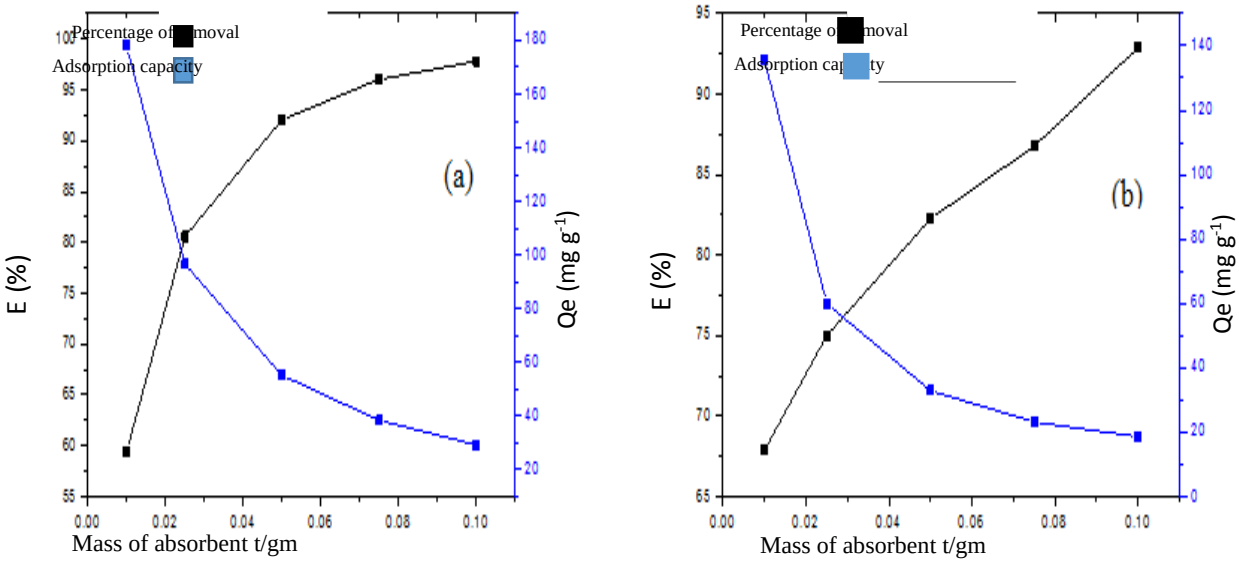

Fig. 2. Effects of the amount of absorbent on absorption (a) of MB, (b) CV: exp. condition: dye conc. 20 ppm, Temperature 35ºC, solution pH 6.5.

Effects of temperature and initial concentrations of CV and MB

The primary concentration supplies a significant leading requirement for overcoming each amount transferring the resistance of each molecule among the solid as well as the aqueous phases. Moreover, the primary concentrations of the two dyes were modified and intervals time were estimated until occurring any absorption on nanoparticles CNT/TiO_2. Figs. 3-5 depict impact of several primary equally the two dye concentrations on the removal percentage and adsorption efficiency of nanoparticles CNT/TiO_2. As shown in these Figs., the adsorption efficiency (Qe) increases by primary elevation in the concentrations of the two dyes (CV and MB), whereas the removal percentage declined by their primary concentrations. The removal percentage of the dyes via adsorption on nanoparticles CNT/TiO_2 originate to be rapidly low and subsequently to be down slower by raising in their concentrations. Such a condition happen through forces attractive among dyes molecules, and thus the mass absorbent such as Vander Waals forces and attractions electrostatic. Rapid diffusion on the external surface was followed by rapid pore dispersion into the intraparticle matrix, including chromophere groups like carbonylic,

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phenolic and alcoholic happening the adsorption, to reach the equilibrium fast (Aljebori & Alshirifi 2012). The removal percentage (%) of the two dyes, have been calculated at the solution temperature of equal to 283, 298, and 313 K. Results showed that temperature largely affect the absorption method by the alteration in solution temperature which leads to modifications in the equilibrium efficiency of the absorbent for adsorption of the certain absorbate. In addition, uptake of the two dyes declines by the elevated solution temperature, displaying the exo-thermic nature of reaction absorption, whereas elevated temperature showed the endo-thermic nature of reaction absorption. In addition, temperature elevates bonding physical among the compound-organic (containing dyes), weakens the active sites, followed by a decrease in obtainable adsorption active sites and a decrease in the freedom degree of type adsorbed (Alkaim *et al*. 2016).

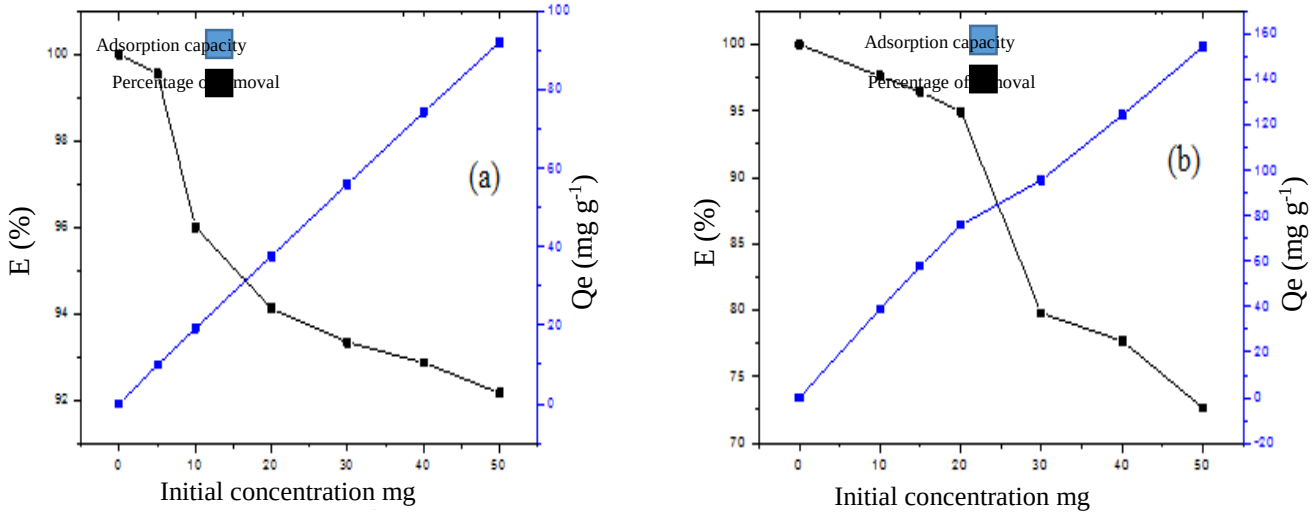

Fig. 3. Impact of primary concentrations on the percentage removal and quantity of the absorbed (a) MB (b) CV dyes onto CNT/TiO_2 nanoparticles.

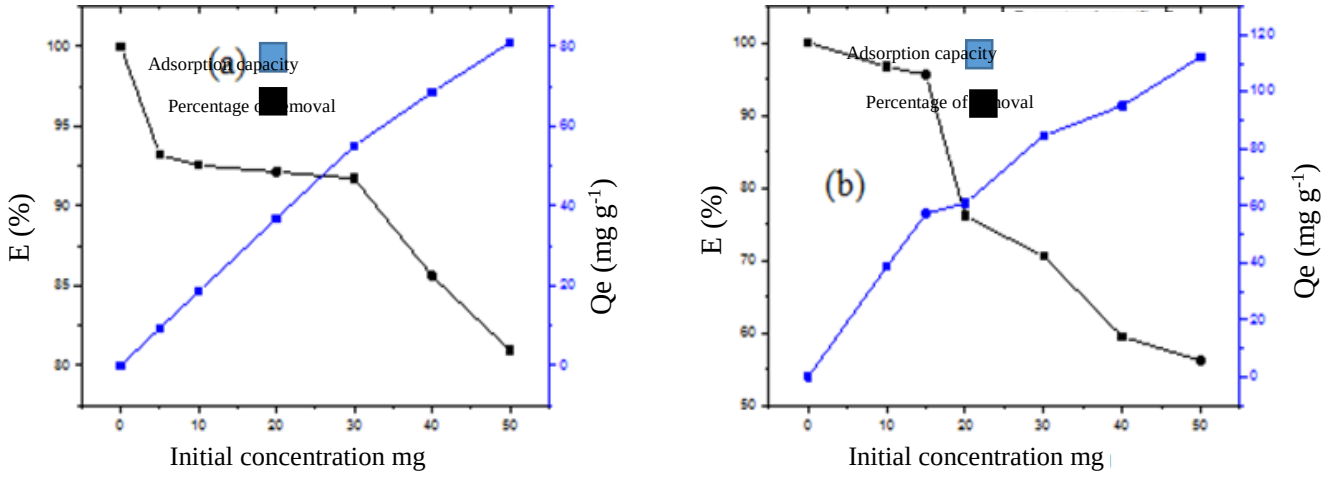

Fig. 4. Impact of the primary concentrations on the percentage removal as well as the quantity of the absorbed (a) MB (b) CV dyes onto CNT/TiO_2 nanoparticles.

Langmuir Isotherm

It was utilized to absorb contaminants from liquid solutions (Langmuir 1918). Langmuir derived another equation on the definite case of the adsorption route nature of the solution. The Langmuir model was developed based on that all accessible sites fixed number has the same energy, and it is available on the adsorbent surface, and there were reversible adsorption and no lateral interactions between the adsorbates. Thus, the Langmuir model is clarified in Eq. 3 (Langmuir 1918):

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

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Where qe: adsorbed amount per unit weight at equilibrium (mg g^{-1}), Ce: concentration of adsorbent equilibrium in the solution following the absorption (mg L^{-1}), qm: the constant of Empirical Langmuir represents the greatest absorption efficiency (mg g^{-1}), K_L: Empirical Langmuir constant (L mg^{-1}). Figs. 6 and 7 represent the model outputs, and Tables 1 - 2 present Langmuir constants. The Freundlich isotherm is known as Eq. 4 (Ho *et al*. 2002).

$$q_e = K_f C_e^{1/n} \qquad (4)$$

Where K_f is the capacity factor (L g^{-1}) and n stands for a deviation measured from the adsorption linearity. This value represents the non-linearity degrees among adsorption and concentration solution as follows: Adsorption route is chemical if the value under to unity or it is linear if the n value equal to unity, finally the favorable physical route when the value was more than the unity adsorption.
The qe plot versus Ce (Figs. 6 and 7), where K_F values 1/n has attained from the slope as well as the intercept of linear regressions (Tables 1-2).

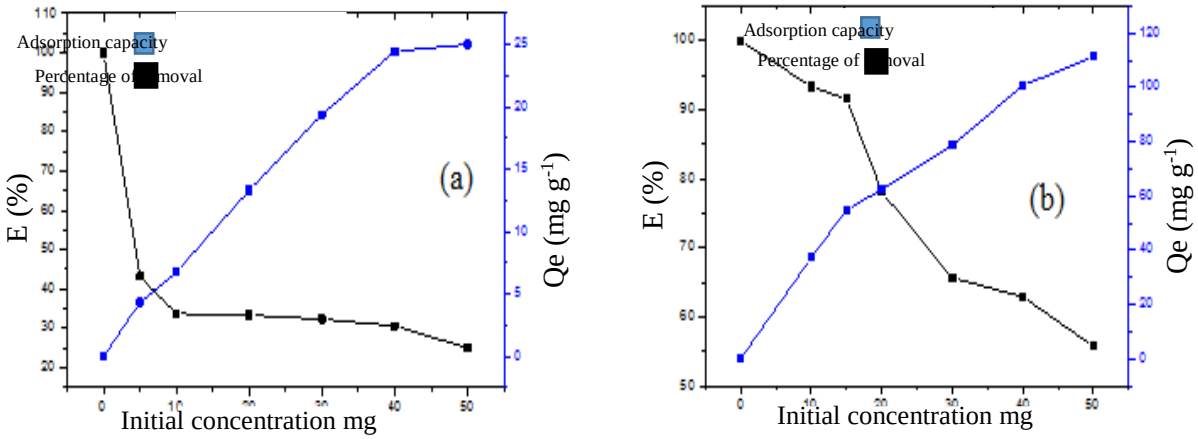

Fig. 5. Impact of the primary concentrations on the percentage removal as well as the quantity of the absorbed (a) MB (b) CV dyes onto CNT/TiO_2 nanoparticles.

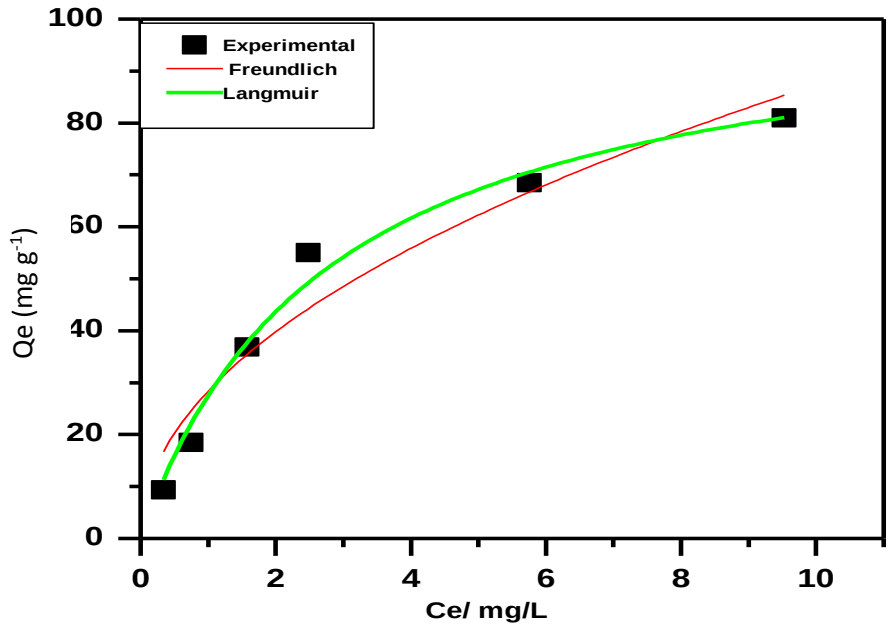

Fig. 6. MB dye absorption model of distinct absorption isotherm non-linear fit on CNT/TiO_2 nanoparticles at pH 6, the mass dosage equal to 0.05 gm, 35 °C initial concentration of 20 mg L^{-1}.

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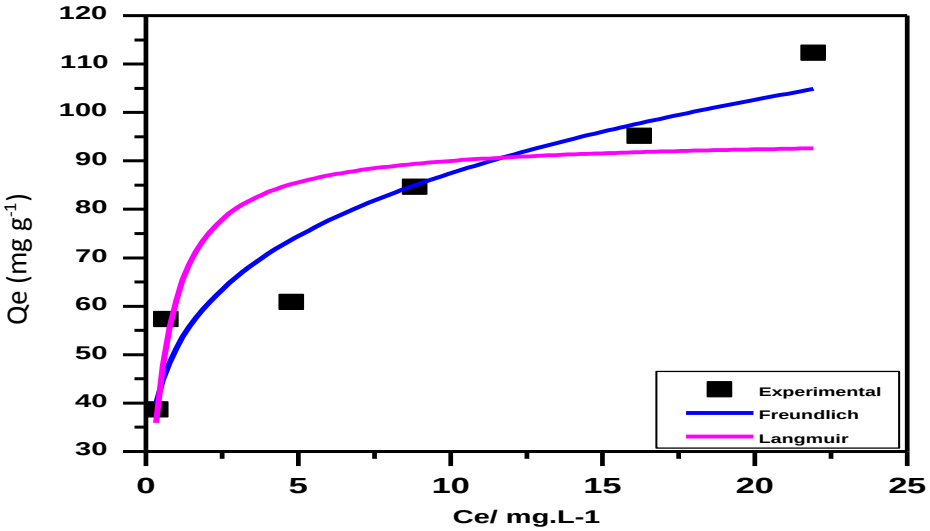

Fig. 7. CV dye absorption model of distinct absorption isotherm non-linear fit on CNT/TiO_2 nanoparticles at a pH 6, the mass dosage equal to 0.05 gm, 35 ^oC initial concentration of 20 mg L^{-1}.

Table 1. The model of Freundlich, Langmuir isotherms variables for MB dye absorbed on the CNT/TiO_2 nanoparticles surface at a temperature of 35 ^oC.

models Isotherm	Parameters	MB dye
Langmuir	qm (mg g^{-1})	104.934 ± 7.45805
	K_L(L mg^{-1})	0.3506 ± 0.06344
	R^2	0.98293
Freundlich	K_F	28.36014 ± 3.96432
	1/n	0.48863 ± 0.07646
	R^2	0.92713

Table 2. The model of Freundlich, Langmuir isotherms variables for CV dye absorbed over CNT/TiO_2 nanoparticles surfaces at a temperature of 35 ^oC.

models Isotherm	Parameters	CV dye
Langmuir	qm (mg g^{-1})	94.90036 ± 9.82538
	K_L(L mg^{-1})	1.8363 ± 1.05978
	R^2	0.6479
Freundlich	K_F	51.3477 ± 5.4961
	1/n	0.23118 ± 0.04302
	R^2	0.88499

CONCLUSIONS

Adsorption technique was used to remove dyes from water using CNT/TiO_2 nanoparticles, and the TiO_2 was developed from CNT/TiO_2 nanoparticles. The experiments were carried out to dyes uptake by CNT/TiO_2 nanoparticles and effect of some experimental factors like temperature, initial dyes concentration, and adsorbent dosage in solution. In addition, the adsorption equilibrium was characterized utilizing Freundlich and Langmuir isotherm. The results have shown immense potential, and it was found that the correlation coefficient value deals with the dyes, while the Langmuir equation was found a not good correlation of dyes.

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رنگ‌های منسوجات به‌عنوان مسئله‌ی اصلی برای محیط‌زیست: مقایسه دفع رنگ‌های بسیار سمی از محلول‌های آبی

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چکیده

آلودگی زیست‌محیطی، یکی از مهم‌ترین و اصلی‌ترین مسائل پیش روی جهان در حال حاضر است. تولید و مصرف جهانی ترکیبات شیمیایی که بسیاری از آن‌ها به تجزیه زیستی مقاوم هستند، در دهه‌های اخیر به‌طور معنی‌داری افزایش‌یافته است. این ترکیبات بعد از مصرف وارد محیط‌زیست شده و بنابراین، لازم است تا راه‌حل‌هایی برای کاهش مصرف و حذف آلاینده‌ها از محیط‌زیست ارائه شود. حذف رنگ‌های منسوجات کریستال بنفش (CV) و متیلن آبی (MB) از محلول با سطح CNT/TiO_2 مطالعه شد. پارامترهای مختلف فیزیکی شیمیایی نظیر زمان تماس، غلظت اولیه رنگ ۵-۵۰ میلی‌گرم در لیتر، مقدار جاذب (۰,۱-۰,۰۱) gm و نیز دمای (۱۵، ۳۵ و ۵۰) درجه را در تکنیک جذب سطحی batch در نظر گرفته شد. جذب دورنگ مطلوب MB وCV در pH بازی مشاهده شد. به‌علاوه، مقدار حذف با افزایش مقدار جاذب افزایش، و با افزایش غلظت رنگ اولیه و زمان هم زنی دورنگ، کاهش یافت. ظرفیت جذب (Qe)، با افزایش زمان تماس و غلظت اولیه رنگ افزایش، و سپس با افزایش وزن جاذب و دما، کاهش یافت. داده‌های تعادل با استفاده از ایزوترم های لانگمیر و فروندلیخ برآورد شد. ایزوترم فروندلیخ، بهترین جذب رنگ‌های MB و CV را توصیف می‌کند. جذب CV و نیز رنگ MB در تحقیق فعلی به $CNT//TiO_2$، با چندلایه ناهمگن است. نتایج حاکی از پتانسیل قابل‌توجه این روش بوده و مشاهده شد که مقدار ضریب همبستگی با افزایش رنگ افزایش می‌یابد، درحالی‌که معادله لانگمیر، همبستگی خوب رنگ‌ها را نشان نداد.

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