



## Highly Fast and Efficient Removal of some Cationic Dyes from Aqueous Solutions Using Sulfonated-oxidized Activated Carbon

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(Received 26 August 2018, Accepted 7 October 2018)

In this study, the potentiality of Sulfonated-Oxidized activated carbon as an adsorbent for removing Methylene Blue (MB), Crystal Violet (CV), and Thionin Acetate (Th) from aqueous solutions was investigated. The characteristics of the synthesized adsorbent were examined by FTIR and SEM techniques. By changing experimental conditions like contact time and adsorbent dosage, initial pH was studied to find the optimum adsorption conditions. The results showed that the adsorption process is too fast and after about 1 min more than 95% of dyes ( $20 \text{ mg l}^{-1}$ ) were completely removed. The experimental data were well represented by Freundlich adsorption isotherm and the maximum adsorption capacities of MB, CV, Th dyes were calculated about 410.0, 405.5 and 395.4  $\text{mg g}^{-1}$ , respectively. These findings are considerably higher than the adsorption capacities of reported adsorbent in the literature. In addition, simultaneous removal of the reported dyes was also carried out and more than 98% removal efficiency was obtained, therefore Sulfonated-oxidized activated carbon appears as an economical and effectual adsorbent for removal of MB, CV, Th from industrial waste waters.

**Keywords:** Wastewater treatment, Dye removal, Activated carbon, Cationic dye, Sulfonation-oxidation

### INTRODUCTION

Organic Dyes have a consequential role as a water pollutant compound in industrial dyestuff and textile waste water. These pollutant substances have detrimental effects on plant life's and abundant number of ecosystems existing in environment. These aforementioned dyes, which generally have strong color solutions, influence the seaweeds' photosynthesis activity by reducing the transparency of environmental waters. Therefore, applying some pretreatments for eliminating these contaminants from water before discharging is needed [1-4].

Numerous approaches such as Biological treatment [5], coagulation/flocculation [6], chemical oxidation [7], membrane processes [8], ozone treatment [9], ion exchange [10], photocatalytic degradation [11] and adsorption [12]

have been performed to remove dyes from effluents. These methods can be then categorized into more general topics such as Physical [13], Chemical [14-15], Biological [5-16-17]. Among these methods, adsorption process has the most application in the organic dyes' removal process due to the economic feasibility and high-quality water production in the process [18]. Zeolites, agricultural wastes, industrial by-products, clay and polymeric materials are the most common adsorbent used in organic dyes' removal. Low adsorption capacity of these adsorbents usually limits their application in waste water treatment [19]. Among these adsorbents activated carbon is one of the best selections for dye removal process [20-21]. High production cost is the main disadvantage of using activated carbon. Therefore, modifying activated carbon surface in order to enhance its adsorption capacity and turning it to an economical adsorbent is of a great challenge [21].

Cationic dyes like methylene Blue (MB), Crystal Violet

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(CV), Thionin Acetate (Th) are some of the most remarkable divisions of synthetic dyes which have lots of industrial and scientific usages. By accessing the cationic dye's production, researches were carried out around the world to control their undesirable effect on human and environment [22-23]. As a result, variant kinds of activated carbon with variable adsorption capacities have been synthesized from different natural sources such as coconut coir, rice husk, *etc*. The majority of these adsorbents have low adsorption capacity and high production cost. Some approaches have been executed in order to ameliorate the adsorption properties of activated carbon. Modifying the activated carbon with different nanoparticles like magnetite is one of these attitudes which significantly impress its adsorption and separation features. Besides, the functionalization and different treatments of activated carbon with variant reagents can result to better interaction forces and subsequently better removal capability [24-25-26].

In this study the activated carbon was oxidized by nitric acid and sulfonated by concentrated sulfuric acid respectively and then its potentiality for removal of some cationic dyes (MB, CV, Th) was investigated. Separation of dye loaded sulfonated-oxidized activated carbon from the solution was achieved by a simple filtration process. The adsorption isotherm models of the aforementioned dye were also investigated. This modification found to be an effective and low cost process in which could improve activated carbon adsorption capacity noticeably.

## EXPERIMENTAL

### Reagents and Materials

All chemical was purchased from Merck Company (Darmstadt, Germany) with the highest available purity. All glassware were soaked in dilute nitric acid for 12 h and rinsed twice with deionized water respectively prior to use. Scheme 1 indicates the chemical structure and their visible spectra of investigated dyes. Stock solutions of dyes were prepared by dissolving 1000 mg of each MB, CV, Th in 1 liter of deionized water. A dye solution of different initial concentrations was prepared by diluting the stock solution in proper proportions.

### Apparatus

pH of the solutions was determined by a Metrohm model 713 pH-meter. Determination of dye concentration of the solutions was carried out using a single beam UV-mini-WPA spectrophotometer. Infrared spectra's of samples were collected by a Perkin-Elmer model spectrum GX, FT-IR spectrometer with spectral range of 4000-400 cm<sup>-1</sup>.

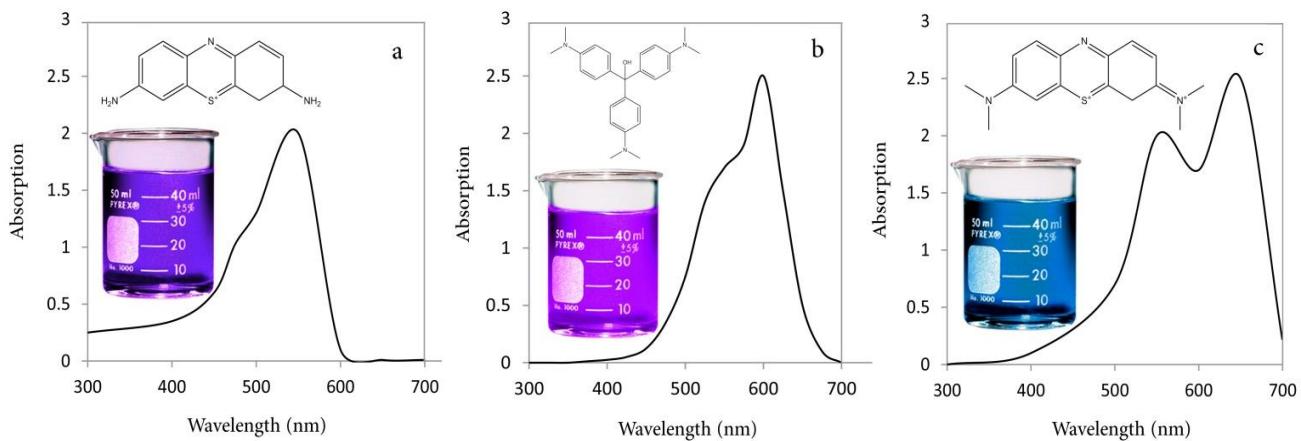
### Synthesis of Sulfonated-Oxidized Activated Carbon

Sulfonated-Oxidized activated carbon was synthesized according to the literature previously reported with some modification [27]. Typically, 2 grams of activated carbon was oxidized by refluxing it with 20 ml of 10% nitric acid at 90 °C for 4 and 8 h, respectively. Then the product was centrifuged at 4000 rpm for 30 min and the supernatant was decanted away and washed twice with 80 °C deionized water followed by yet another centrifugation at 4000 rpm for 30 min. The oxidized activated carbon dried in an air oven at 110 °C. For The sulfonation process 100 ml of concentrated sulfuric acid was added to the prepared oxidized activated carbon and stirred at 150 °C under nitrogen atmosphere for 15 h. The reaction mixture was cooled to room temperature and 300 ml of deionized water was added in portions under stirring. The product was centrifuged at 4000 rpm for 30 min and the supernatant was decanted away and washed twice with 80 °C deionized water followed by centrifugation (4000 rpm) for 30 min. the Sulfonated-Oxidized activated carbon dried in an air oven at 110 °C. The synthesis procedure is represented in Scheme 2.

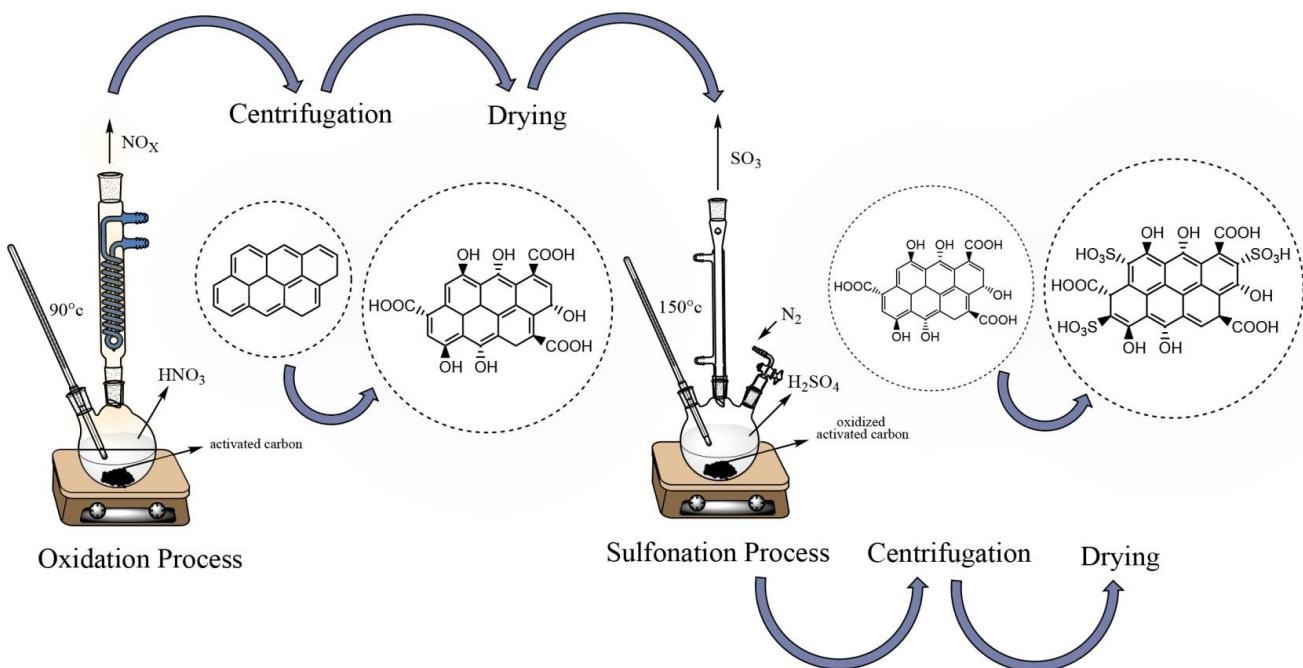
### Dye Removal Experiments

Fifteen mg of Sulfonated-Oxidized activated carbon was added to 30 ml of 20 mg L<sup>-1</sup> of each cationic dyes with predetermined concentration while the solution's pH was adjusted at 2.0 with 0.1 mole.L<sup>-1</sup> HCl and 0.1 mole.L<sup>-1</sup> NaOH solutions. In the next step, the solutions were stirred for 10 min at room temperature. Then the adsorbent was separated from the mixture via filtration throw a Whatman 42 filter paper. The residual concentration of dyes in the solution was determined spectrophotometrically at the maximum wavelength of each dyes. The removal efficiency of each dye was determined using the following expression:

$$R.E.\% = \frac{(C_0 - C_t)}{C_0} \times 100 \quad (1)$$



*Scheme 1.* Visible spectra and chemical structure of thionin (a), crystal violet (b) and methylene blue (c)



*Scheme 2.* Schematic illustration of sulfonated-oxidized activated carbon synthesis

Where  $C_0$  and  $C_t$  represent the initial and final concentration of dyes in  $\text{mg l}^{-1}$  in Eq. (1). The whole determinations were carried out in triplicate to insure the repeatability of the result. The mean of the three measurements was then reported. All the experiments were performed at room temperature.

## RESULTS AND DISCUSSION

### Characterization of the Adsorbent

SEM micrographs in Fig. 1 reveal details of activated carbon porosity development during the functionalization. Figure 1 suggests that most of activated carbon particles are

prepared by carbonizing vascular bundle which is observable as network of isolated cylindrical units. It is nonetheless noteworthy that according to Fig. 1a all pores are blocked, and the surface of the activated carbon is coated by foreign materials which indicates that the activation process was not complete. As a result of treatment by nitric acid, most of the foreign materials are cleared from the surface of the particles but nearly all pores are still remaining blocked. Eventually the sulfonation process serves the additional purpose of cleaning foreign materials from the carbon besides the creation of pores. Due to the thermal and oxidative stresses existing in the sulfonation process, the activated carbon particles have been broken to small pieces which contribute to the overall surface of the activated carbon.

As an evidence for the formation of expected functional groups on activated carbon, FT-IR spectra of the products were recorded in different parts of the synthesis process. Figure 2 illustrates FT-IR spectra of activated carbon, oxidized activate carbon and sulfonated-oxidized activated carbon. According to spectrum (a) which appertains to starting activated carbon, three peaks at around  $1160\text{ cm}^{-1}$ ,  $1567\text{ cm}^{-1}$  and  $1717\text{ cm}^{-1}$  relate to (C-O stretching) of tertiary alcohol, (C=C stretching) of cyclic alkene and (C=O stretching) of carboxylic acid functional groups, respectively. In addition, two peaks at around  $800$ - $900\text{ cm}^{-1}$  and  $3500\text{ cm}^{-1}$  in spectrum (b) relate to (C=C stretching) of substituted alkene and (O-H stretching) of alcohol, respectively, which is formed due to the oxidation process. Eventually, formation of two additional peaks at around  $1160\text{ cm}^{-1}$  and  $1370\text{ cm}^{-1}$  indicates the presence of sulfonic acid and sulfonate groups after sulfonation process, which relates to (S=O stretching) of sulfonic acid and sulfonate functional groups respectively.

### pH's Effect

pH of the solution plays an important and affecting role in adsorbing the dye molecules on the surface of the adsorbent. Binding sites of the adsorbent and chemical conditions of the solution can be affected by pH of the solution. The influence of dye solution's initial pH on the efficiency of adsorption removal process was studied in the range 1-10 for  $20\text{ mg l}^{-1}$  aqueous solutions of three investigated dyes with 10 min stirring. As shown in Fig. 3,

the removal efficiency of the adsorbent is remained nearly constant in pH 1 and 2 and then decreased mildly by increasing the pH the adsorption quantity. The calculated removal efficiencies for all pH values are more than 93%, which indicates the deficient dependence of the adsorption to the pH of the solution. However, in lower pH values like 1 and 2, most of the nitrogen, existed in groups of the dyes can be protonated and get positive charge. Although the sulfonate groups existing on the adsorbent's surface are the conjugated base of sulfonic acid, the adsorbent's surface is negative in a wide pH range. Consequently the electrostatic interaction between protonated dyes and the adsorbent is increased which results in maximum removal efficiency [28].

### Effect of the Amount of Adsorbent

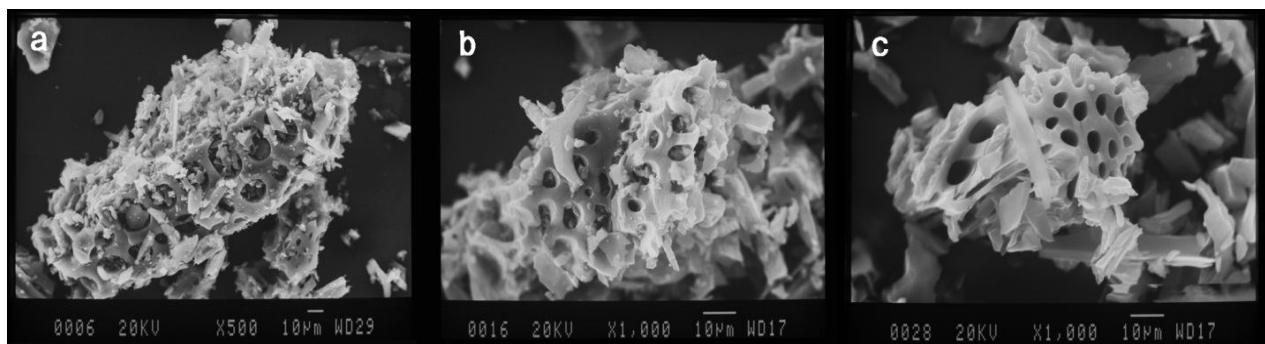
The affiliation of dye adsorption on amount of the adsorbent was investigated at room temperature and at pH = 2.0 by varying the amount of the adsorbent between the range of 0.005-0.015 g in contact with 30 ml solution of  $20\text{ mg l}^{-1}$  of each dye. The results are shown in Fig. 4. Evidently, by increasing the amount of the adsorbent, higher adsorption sites will exist in the solution and accession of the removal efficiency can be seen. According to Fig. 4 more than 98% removal efficiency can be achieved for each dye by adding 0.015 g of the adsorbent to the dye solution.

### Effect of Contact Time

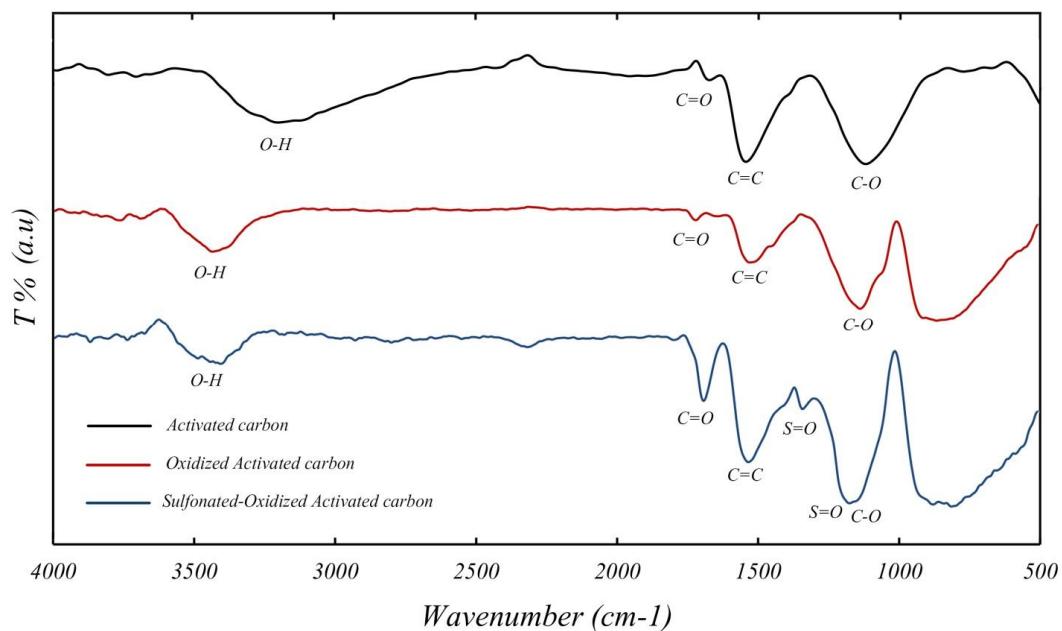
The extent of the adsorption was studied as a function of time for removing each dye from  $20\text{ mg l}^{-1}$  solution at pH = 2. For this purpose a 0.015 g of the adsorbent was added to the solution of each dye followed by stirring for predetermined periods of time. Eventually the absorbance of each solution was determined to monitor the dye concentration. As shown in Fig. 5, the adsorption speed of the investigated dyes on the surface is too fast and after about 1 min more than 95% of dyes were completely removed. However the maximum percentage of removal was accede after 10 min; hence agitation time of 10 min was adopted for further works.

### Adsorption Isotherms

The best approach for optimization the amount of the adsorbent is to find the appropriate adsorption model in



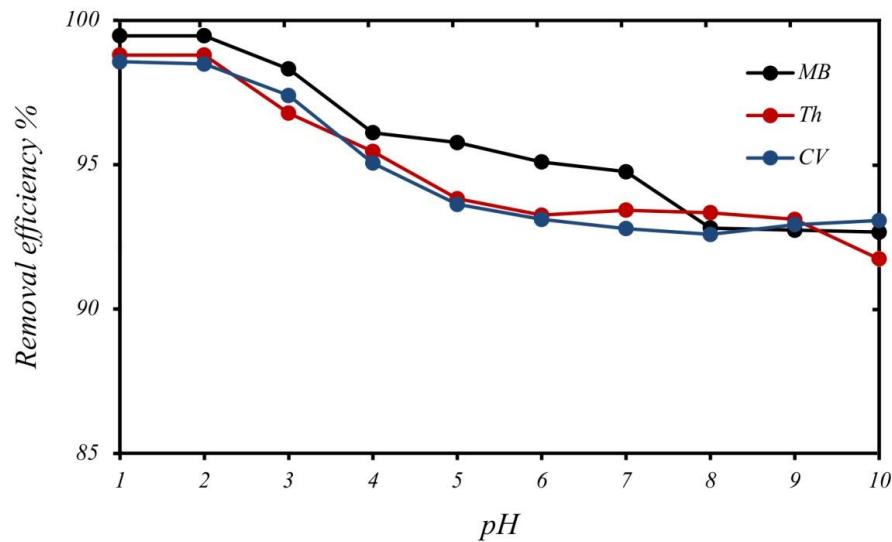
**Fig. 1.** SEM images of starting activated carbon (a), oxidized activated carbon (b), sulfonated-oxidized activated carbon (c).



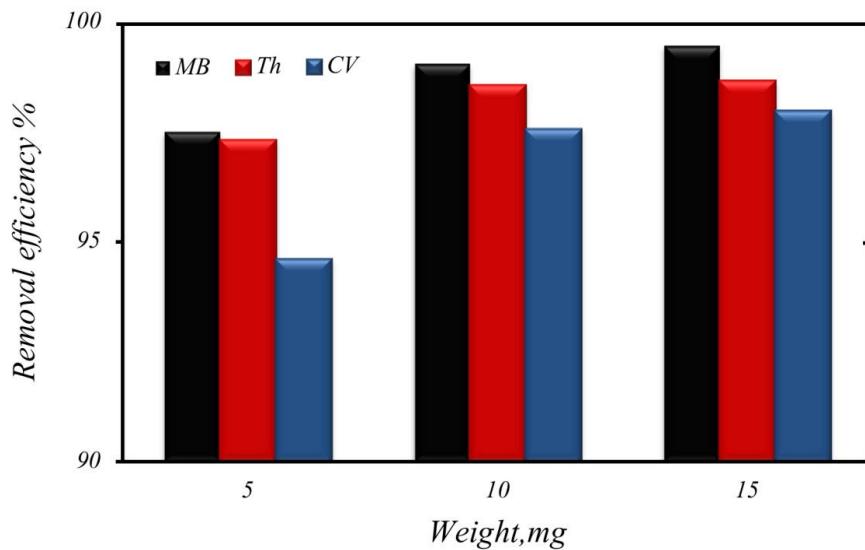
**Fig. 2.** FT-IR spectra of starting activated carbon (a), oxidized activated carbon (b), sulfonated-oxidized activated carbon (c).

which it has the most adaptation with experimental data. In this work Langmuir [29], Freundlich [30], Redlich-Peterson [31], Temkin [32] and Sip [33] models were employed to analysis the equilibrium data of adsorption of three investigated dyes onto sulfonated-oxidized activated carbon. The evaluation process for adsorption equilibrium curves was carried out by adding the weighted amount of

synthesized adsorbent to 30 ml solution of three dyes at pH = 2.0. The residual concentration of dyes was determined after equilibration. The Langmuir adsorption model assumes monolayer adsorption on a uniform surface with limited number of adsorption sites. The Langmuir isotherm shows that by approaching the concentration of the adsorbate to saturation point, an accession in amount of



**Fig. 3.** Percentage of dye removal at different pH values for MB, Th, CV .Conditions: 0.015 g of sulfonated-oxidized activated carbon, 30 ml of 20 mg l<sup>-1</sup> of dye, agitation time of 10 min.



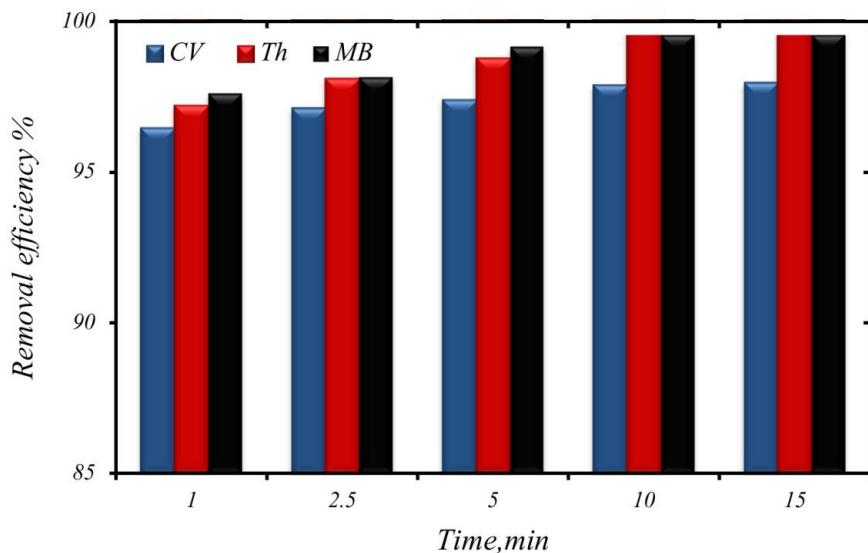
**Fig. 4.** Percentage of dye removal at different amount of sulfonated-oxidized activated carbon. Conditions: pH: 2.0, 30 ml of 20 mg l<sup>-1</sup> of dye, agitation time of 10 min.

solute adsorbed can be seen. As soon as all adsorption sites occupied, no further adsorbate can be adsorbed on the surface by increasing the solute's concentration.

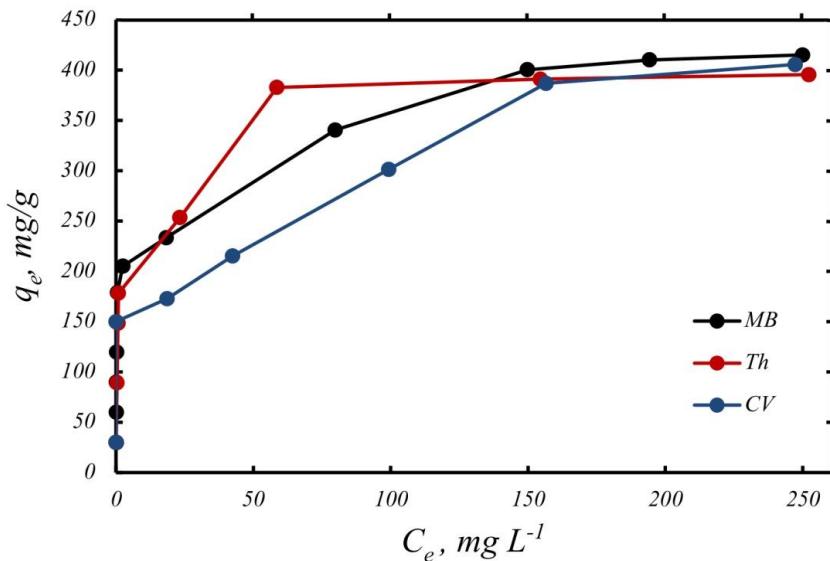
The following equation indicates the general form of the

Langmuir isotherm:

$$\frac{q_e}{q_m} = \frac{K_L C_e}{1 + K_L C_e} \quad (2)$$



**Fig. 5.** The effect of contact time on the adsorption of cationic dyes on sulfonated-oxidized activated carbon.  
Conditions: pH: 2.0, 30 ml of 20 mg l<sup>-1</sup> of dye. Adsorbent amount: 0.015 g.



**Fig. 6.** Langmuir adsorption isotherm of MB, Th, CV for sulfonated-oxidized activated carbon.

Where q<sub>e</sub> refers to the amount of dye adsorbed on per gram of the adsorbent at equilibrium. K<sub>L</sub> is a constant and C<sub>e</sub> is the equilibrium concentration of dye (mg l<sup>-1</sup>). q<sub>m</sub> is the maximum amount of dye can be adsorbed on per gram of the adsorbent (mg g<sup>-1</sup>), which relates to the number of

adsorption sites directly (Fig. 6).

The above equation can be rearranged to the following linear form.

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

The Freundlich isotherm can be represented as:

$$q_e = K_F C_e^{1/n} \quad (4)$$

Where  $K_F$  ( $\text{mmol}^{1-1/n} \text{l}^{1/n} \text{g}^{-1}$ ) is a Freundlich constant and indicates the adsorption capacity and  $1/n$ . Is the other constant in which  $n$  is related to adsorption energy distribution. Both of these constants depend on the temperature and adsorbent-adsorbate couple.

The linearized form of the Freundlich isotherm is described below:

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

Redlich-Peterson isotherm model is a modification of Langmuir model which comprises the features of Langmuir and Freundlich models simultaneously:

$$q_e = \frac{K_{RP} C_e}{1 + a_{RP} C_e^\beta} \quad (6)$$

In this equation  $K_{RP}$  ( $1 \text{ g}^{-1}$ ) and  $a_{RP}$  ( $1 \text{ mg}^{-1}$ ) are Redlich-Peterson constants and  $\beta$  is a dimensionless exponent which can be in the range of 0-1. At low and high concentration of the adsorbate this equation approximates to Henry's law and Freundlich model, respectively.

Temkin isotherm describes that the interaction between the adsorbate and adsorbing species leads to a linear decline of adsorption energy. This equation is stated as follows:

$$q_e = B_T \ln(K_T C_e) \quad (7)$$

$B_T$  depends on the heat of the adsorption and called Temkin constant.  $K_T$  is another Temkin isotherm constant.

Sips isotherm is a nonlinear coalescence of Langmuir and Freundlich isotherms which describes as follows:

$$q_e = \frac{q_m K_S C_e^{\frac{1}{n}}}{1 + K_S C_e^{\frac{1}{n}}} \quad (8)$$

In this equation the Sips constant ( $K_S$ ) relates to the adsorption energy.

The whole parameters of the aforementioned Isotherms were calculated for the adsorption of three investigated dyes onto Sulfonated-oxidized activated carbon and are shown in Table 1. The maximum adsorption capacities of three investigated dyes onto starting activated carbon and sulfonated-oxidized activated carbon are shown in Table 2. As an evidence, the maximum adsorption capacities of three investigated dyes onto starting activated carbon and oxidized activated carbon were measured by performing the adsorption process for 30 ml solution containing 700 mg  $\text{l}^{-1}$  of each dyes under optimum conditions and the calculated values are shown in Table 2. The results declare that the adsorption equilibrium data of Sulfonated-oxidized activated carbon have better adaptation with Freundlich Isotherm for all dyes. According to Fig. 6 the maximum adsorption capacities of MB, CV, Th are almost equal and is about 410.0, 405.5, 395.4  $\text{mg g}^{-1}$ , respectively. This indicates that sulfonation and oxidation processes can considerably enhance the capacity of activated carbon for adsorption of cationic dyes.

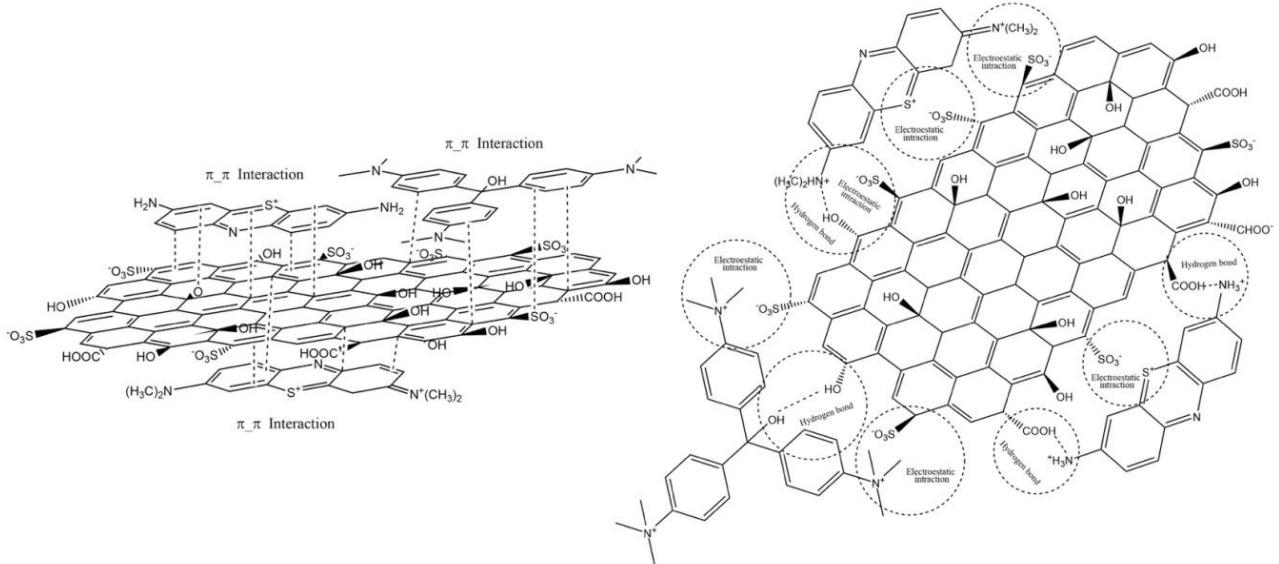
This is while the adsorption kinetic of three aforementioned dyes on starting activated carbon was too much slower and after about 15 min 95% removal was achieved. The obtained results shed light on the mechanism of cationic dye adsorption on sulfonated-oxidized activated carbon, additionally revealing that the synergistic interplay of electrostatic,  $\pi-\pi$ , and hydrogen bonding interactions results in too fast and highly efficient adsorption of organic dyes. The adsorption was observed to mainly proceed via similar mechanisms for all sulfonated-carboxylated adsorbents featuring free  $\text{SO}_3^-$  and  $\text{COOH}$  functional groups with electrostatic interactions. Moreover, the strong  $\pi-\pi$  interactions between the benzene rings of activated carbon structure and the aromatic backbone of three investigated cationic dyes increased the adsorption uptake of these dyes. In addition, hydrogen bonding interactions can occur between the hydroxyl and carboxylic acid functional groups (H-donors) existing on the surface of the sorbent and nitrogen atoms (H-acceptors) of three aforementioned dyes. Scheme 3 illustrates the different interactions existing between the investigated dyes and sulfonated-oxidized activated carbon.

**Table 1.** Adsorption Isotherms Parameters of Dyes onto Sulfonated-oxidized Activated Carbon

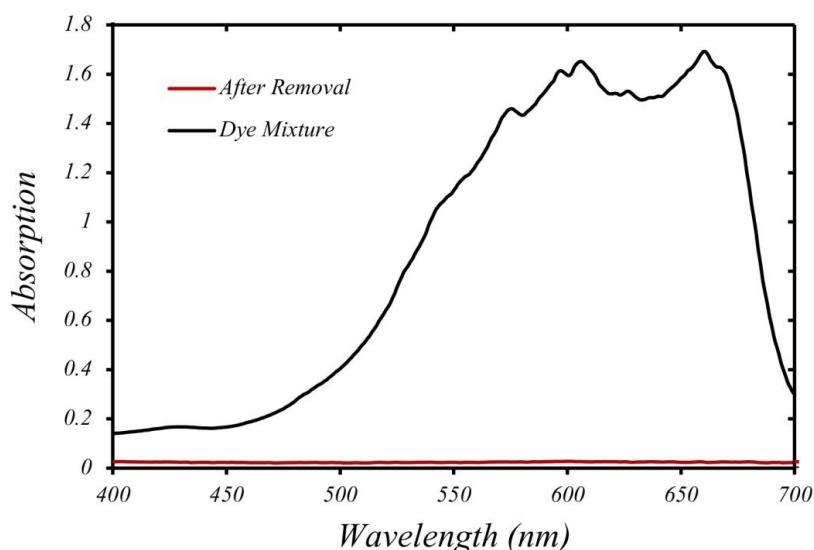
Isotherm Models	Parameters	Dyes		
		MB	Th	CV
Langmuir	$q_m$ (mg g <sup>-1</sup> )	338.37	262.63	321.78
	$K_L$ (l mg <sup>-1</sup> )	2.02	-33885.59	4.98
	$K_L q_{max}^{-1}$	$5.9 \times 10^{-3}$	-129.02	$5.24 \times 10^{-5}$
	$R^2$	0.8354	0.3177	0.6793
Freundlich	$K_F$ (mmol <sup>1-1/n</sup> l <sup>1/n</sup> g <sup>-1</sup> )	133.14	122.48	63.31
	1/n	0.20	0.22	0.34
	$R^2$	0.9958	0.9908	0.9952
Redlich-Peterson	$K_R$ (l g <sup>-1</sup> )	1911.51	710.84	2.73
	$a_R$ (l mg <sup>-1</sup> )	10.41	3.45	$9.27 \times 10^{-9}$
	$\beta$	0.852	0.875	3.28
	$R^2$	0.9479	0.9580	0.718
Temkin	$B_T$	41.81	45.71	39.56
	$K_{Te}$	58.39	29.80	29.80
	$R^2$	0.9392	0.946327	0.9463
Sips	$q_m$ (mg g <sup>-1</sup> )	1261.81	400.06	410.6
	$K_S$	0.13	0.53	0.31
	1/n	0.23	0.86	1.56
	$R^2$	0.9345	0.9118	0.6728

**Table 2.** Comparison of Maximum Adsorption Capacities of Dyes onto Starting Activated Carbon and Oxidized Activated Carbon and Sulfonated-oxidized Activated Carbon

Adsorbent	$q_{max}$ (mg g <sup>-1</sup> )		
	MB	CV	Th
Starting activated carbon	247.5	231.8	264.1
Oxidized activated carbon	327.4	321.8	290.2
Sulfonated-oxidized activated carbon	410.0	405.5	395.4



**Scheme 3.** Schematic illustration of different interactions between MB, CV, Th dyes and sulfonated-oxidized activated carbon



**Fig. 7.** Simultaneous removal of MB, Th, CV, from aqueous solution using sulfonated-oxidized activated carbon.  
Conditions: pH: 2.0, 30 ml of 20 mg l<sup>-1</sup> of each Dye. Adsorbent amount: 0.015g, agitation time of 10 min.

### Adsorption Kinetics

Adsorption process's rate can be examined by pseudo-first order and pseudo-second order kinetic models and furthermore the temperature can efficiently affect the

adsorption rate. Therefore, investigation of this matter is of vital importance. The adsorption process of three investigated dyes onto sulfonated-oxidized activated carbon was too fast and after about 1 min more than 95% of

**Table 3.** Comparison of Capacity Factor for some Reported Adsorbent with Proposed Method

Adsorbent	Capacity factor (mg g <sup>-1</sup> )			Ref.
	MB	Th	CV	
Activated carbon of <i>Ficus Carica</i>	47.62	-	-	[34]
Activated carbon of tea ( <i>Camellia sinensis L.</i> ) seed shells	324.7	-	-	[35]
Activated carbon of tobacco stems	123.45	-	-	[36]
Activated carbon of <i>Lantana camara</i> stem	19.84	-	-	[37]
strongly basic anion exchange resin	45	-	-	[38]
Graphene oxide	351	-	-	[39]
Activated carbon modified by Iron oxide nanoparticles	-	-	44.7	[40]
Activated carbon modified by magnetite nanoparticles	-	306.1	-	[25]
Poly(vinyl alcohol)-sodium alginate-chitosan montmorillonite hydrogel beads	137.2	-	-	[41]
Metal organic framework-235	252	-	-	[42]
Sulfonated-oxidized activated carbon	410.0	395.4	405.5	Proposed method

dyes was completely removed. This can be due to the presence of lots of anionic functional groups on the surface of the adsorbent which are produced by oxidation and sulfonation processes. Hence shortage of the adsorption time prohibits the consideration of adsorption kinetics for this adsorbent. Whereas the rate of the adsorption increases by accession of temperature, therefore effect of the temperature on the adsorption process couldn't also be investigated.

#### Simultaneous Removal of Dyes

Industrial dyestuff and textile waste water are a complex mixture of variant dyes; therefore the ability of the adsorbent for simultaneous removal of dyes is a consequential matter which allows its application in waste water treatments. For this purpose, the ability of the adsorbent for simultaneous removal of dyes was investigated by consideration of the dye concentration before and after adsorption process. The experiment was

carried out by stirring a mixture of 15 mg of adsorbent in 30 ml solution containing 20 mg l<sup>-1</sup> of each dye for 30 min. The removal efficiency of the adsorbent was calculated by recording the visible spectrum of the solution before and after the adsorption. As shown in Fig. 7, the complete removal of dyes has been completed successfully and more than 98% removal efficiency was achieved.

#### CONCLUSIONS

The present study disclosed two simple and scalable processes as Oxidation and sulfonation by two inexpensive materials (dilute nitric acid and concentrated sulfuric acid) that can effectually improve the adsorption capacity of activated carbon and make it a cost effective adsorbent for the removal process of some cationic dye pollutants from industrial waste water. According to Table 3, the adsorption capacity of the synthesized sulfonated-oxidized activated carbon for adsorption of MB, CV and Th is much higher

than reported adsorbents. Optimization of adsorption conditions was carried out at room temperature and the results indicated that under optimum conditions, adsorption process is too fast and after about one minute more than 95% of dyes is completely removed which is attributed to the both presence of sulfonic acid, carboxylic acid, and hydroxyl functional groups on the surface of the adsorbent and unique porous characteristic of activated carbon. Consequently, this advantage can present the synthesized adsorbent as a competent selection for dye removal in flow systems. In addition more than 98% efficiency of simultaneous removal of dyes proves its application in waste water treatments. Aforementioned advantages have significant industrial importance, as it will facilitate working equipment ensuring efficiency and economy.

## ACKNOWLEDGMENTS

The authors acknowledge the Bu-Ali Sina University Research Council and Center of Excellence in Development of Environmentally Friendly Methods for Chemical Synthesis (CEDEFMCS) and from Bu-Ali Sina University exceptional Talents development center for providing support to this work.

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