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The Application of Sio_2Nanoparticles for Anionic Dye Removal from Aqueous Solution

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Background & Aims of the Study: Dyes are one of the widespread chemicals which may have many adverse effects on life cycle. Thus, this study was conducted to survey the RB19 dye removal efficacy from aqueous solution, using the SiO_2 nanoparticles.

Materials and Methods: This study was an experimental-lab study. The batch adsorption system were utilized to evaluated the RB19 dye removal efficiency under different amount of effective parameters including contact time, pH, adsorbent dosages, initial RB19 dye concentration and temperature.

Results: the results of this study showed that the increasing of contact time, adsorbent dosage and temperature (up to 30) can positive effect on dye removal efficiency and can develop the dye removal efficiency, while the increasing of pH and RB19 dye concentration decrease the dye removal efficiency. The maximum dye removal efficiency was observed to be 95.72% at pH=5 and temperature of 30. Furthermore, the equilibrium studies of the RB19 dye adsorption depicted that the Langmuir isotherm and pseudo-second-kinetic models was successfully able to describe the equilibrium data.

Conclusion: It can be concluded that SiO_2 nanoparticles are excellent adsorbent to remove the dyes from aqueous solution.

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Background

Historically, the development of human life style has been associated with the need to large amount of energies and chemicals that might be destructive and hazardous for the environment (1-2). The dyes are of major class of the chemicals which have various target industries such as textiles, leather, paper, printing and cosmetics but the highest amount of dyes is used in textile industries (3-4). Dyes are drastically used in these industries and can generate the colorful wastewater (5). This produced wastewater which can simply release into environment and be considered as a major risk for various type of life in environments (6-7). Among various types of dyes, the reactive dyes are identified as widely used dyes to colorize the fibers (8). A large amount of these dyes (50%) is lost during dying and it is obvious that great volume of colorful wastewater is producing by this part (9). The toxicity and aesthetic risks of the dyes has been reported in various literatures. They are supposed to participate in some diseases such as dermatitis and cancers, etc (10-11). Thus, the colorful effluents should be considered as one major risk and best methods should be applied to alleviate the dangerous effects of these chemical to save the human and environment. Different approaches such as adsorption, membrane separation, electrochemical, flocculation-coagulation, reverse osmosis, ozone oxidation, and biological treatments have been used to remove the dyes. Although, these methods was able to successfully remove the dyes; however, the popularity of these methods are reducing due to their cost and complexity (12-13). The nanotechnology and use of the nano-material is one of the effective innovations to remove the pollutants which have gained many interesting among the societies. The nanoparticles have been used in various studies to remove a number of pollutants such as dyes (14-15), antibiotics (16), heavy metals (17-18), etc. Those studies showed that the nanoparticles has considerable efficacy in removal of mentioned pollutants. This substantial efficacy is due to the unique features of these materials including high adsorption capacity, simplicity in operation, rapid adsorption process, etc (19).

Aims of the study:

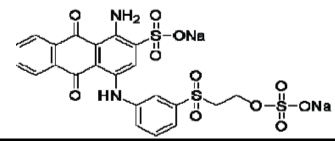
The aim of the present work was to study the SiO_2 nanoparticles ability in reactive blue 19 dye removal. Also, this study was conducted to determine the effect of the contact time, pH, temperature, dye concentration and SiO_2 nanoparticles dosage on dye removal efficiency.

Materials & Methods

Chemical material:

The Reactive Blue 19 (RB19) dye was provided from AlvanSabet Corporation, Hamadan, Iran. The chemical characteristics and structure of RB19 dye is represented in the Table 1.

Table 1) the chemical structure and properties of RB19 dye (20)

Chemical structure	
Molecular formula	$C_{22}H_{16}N_2Na_2O_{11}S_3$
Molecular weight (g/mol)	626.54
Maximum wavelength(nm)	592

Adsorbent properties:

The SiO_2 nanoparticles were purchased from Research Institute of Petroleum Industry (RIPI), Tehran, Iran which was used to study the adsorption characteristics of RB19 dye from aqueous solutions. The outer diameter of SiO_2 nanoparticles used in this study was in range of 10 to 20 nm. In addition, the specific surface area of SiO_2 nanoparticles was reported to be greater than $210 m^2/g$ and the mass ratio of the amorphous carbon of SiO_2 nanoparticles was lesser than 10%. The SEM images of this adsorbent were provided, using a Philips XL30 scanning electron microscope (SEM). This image (Fig.1) shows the porous surface of this adsorbent and reveals that it can be proper adsorbent for RB19 dye removal.

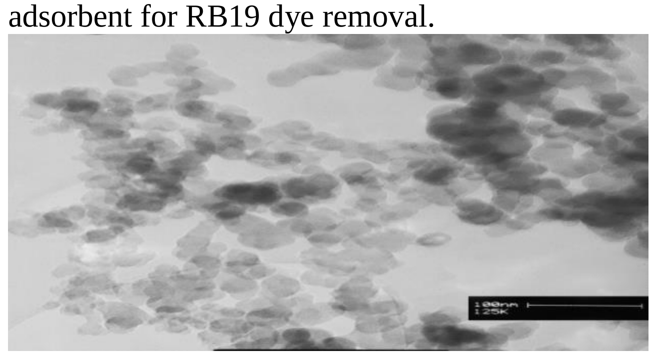


Figure 1) the SEM image of the SiO_2 nanoparticle

Batch adsorption studies:

At the first, a stock solution of RB19 dye (1000mg/L) was prepared and other desired solution with different initial dye concentrations were prepared by diluting of the stock solution. The batch adsorption system was utilized to study the influence of the studied factors on adsorption process including contact time (10-180min), pH (3-11), temperature (20-60 ℃), adsorbent dose (0.2-1.6g/L) and initial concentration of dye (10-500mg/L). All of the experiments were performed in 250mL flask Erlenmayer. 100ml of dye solution with different concentration was poured into the flask and SiO_2 nanoparticles with certain dose were added. The pH of solutions was regulated, using 0.1N HC1 or 0.1N NaOH. Then, prepared samples were mixed, using magnetic stirrer at 180rpm. The experiments were performed by varying of target parameter and in constant values of other parameters to obtain the optimum amount of each parameter. After mixing the samples, they were filtered and the final RB19 dye concentration in solution was determine by the UV-Visible spectrophotometer (DR-4000) at a wavelength of 592nm (20). The equilibrium experiments of adsorption process were conducted after determination of equilibrium time to survey the effect of adsorbent mass on dye removal for obtaining the adsorption isotherms. The following equation is used to calculate the amount of adsorption at equilibrium, q_e (mg/g):

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

Where; C_0 and C_e (mg/L) are the initial and equilibrium RB19 dye concentrations in solution, respectively. V (L) is the volume of the solution and W (g) is the mass of dry sorbent used.

The dye removal percentage is calculated as follows:

$$R = \frac{(C_0 - C_e)100}{C_0} \qquad (2)$$

Results

Effect of SiO_2 nanoparticles dosage:

In this work, the effect of the SiO_2 nanoparticles was studied by varying of this parameter in range of 0.2 to 1.6g/L. the results is represented in figure 2. This figure indicated that the increasing of the adsorbent dose can develop the RB19 dye removal efficiency. It is clear from the results that the RB19 dye removal efficiency is increased from 44.65% to 95.63% by the increasing of SiO_2 nanoparticles dose from 0.2 to 1.6g/L but the dye adsorption capacity is decreased from 74.41 to 14.94mg/g.

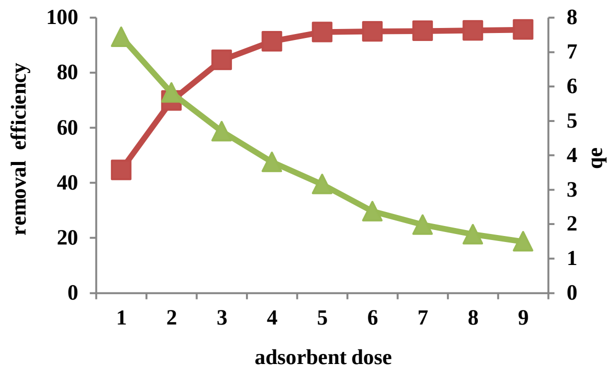


Figure 2)The effect of adsorbent dosage on RB19 dye removal efficiency (contact time=90min, pH=5, initial dye concentration= 25mg/L, temperature=30℃)

Effect of pH:

The effect of pH on RB19 dye removal efficiency was studied in different pH values of solution in range of 3to 11 and the results are represented in figure 3. As it can be seen, the highest RB19 dye removal efficiency was observed at the pH in range of 3 to 6 and then it is gradually decreased.

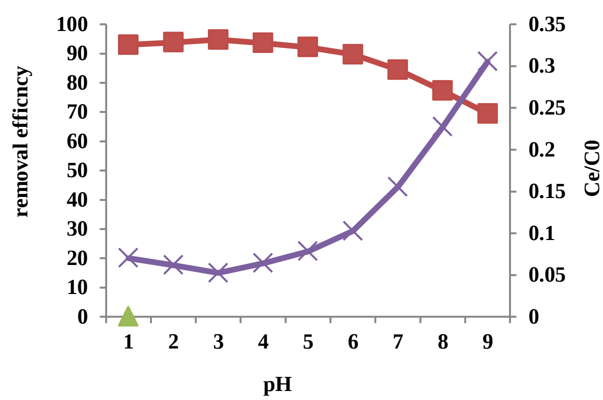


Figure 3)The effect of pH on RB19 dye removal efficiency (contact time=90min, adsorbent dosage=0.75g/L, initial dye concentration=25mg/L, temperature=30℃)

Effect of contact time and temperature:

The contact time and temperature are other effective parameters on dye removal efficiency.

The effect of these parameters was evaluated by variation of the contact time (10-180min) and temperature (20-60℃). The results of this study are shown in figure 4. As it can be seen, the dye removal efficiency increased by increasing the contact time and temperature. The best dye removal efficiency was 95.72% at the 30. In addition, the dye removal efficiency is significant at the initial time of the adsorption process and it remains constant after 90 min.

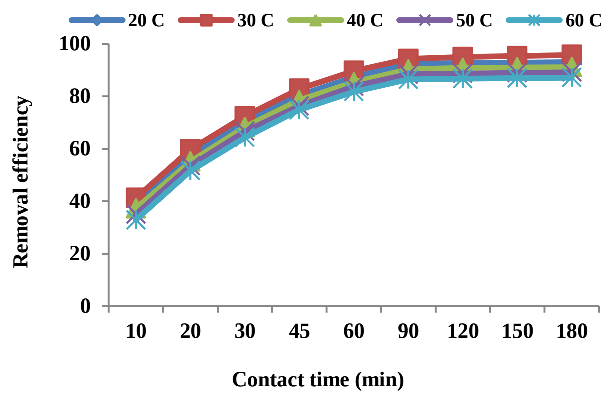


Figure 4) The effect of contact time and temperature on RB19 dye removal efficiency (adsorbent dosage=0.75g/L, initial dye concentration=25mg/L, pH=5)

The effect of initial RB19 dye concentration

The initial dye concentration is another parameter which can influence the adsorption process. The effect of this parameter was investigated by the variation of initial concentration of RB19 dye (10-500mg/L), using the constant amount of other parameters. The results of this study were brought in figure 5. This figure demonstrates that increasing the dye concentration has negative effect on dye removal efficiency.

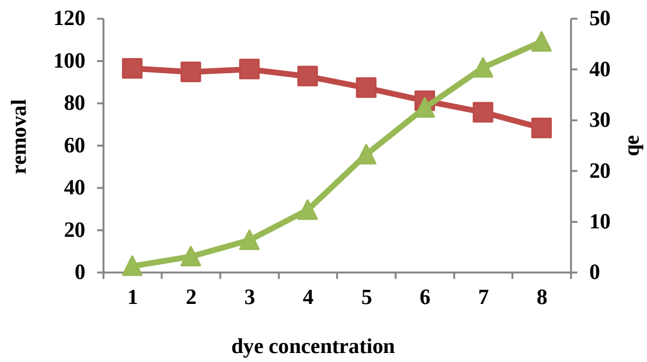


Figure 5)The effect of initial dye concentration on RB19 dye removal efficiency (contact time=90min, adsorbent dosage=0.75g/L, pH=5, initial RB19 dye concentration=50mg/L, temperature=30℃)

Isotherm studies

The equilibrium data of the RB19 dye adsorption were fitted on the Langmuir and Freundlich models to understand the best models to best design of the process and to optimize the use of nanoparticle.

Langmuir Isotherm:

The Langmuir isotherm is resulting on the theory of mono-layer adsorption on a homogenous surface. This equation of this isotherm can express by following equation (21-22):

$C_e/q_e=1/q_mK_L+C_e/q_m$ (3)

Where, C_e (mg/L) and q_e (mg/g) are the concentration of RB19 and the amount of RB19 dye adsorbed onto SiO_2 nanoparticles at equilibrium, respectively. q_m is indicates the maximum monolayer adsorption capacity (mg/g) and K_L is the Langmuir constant (L/mg).

Freundlich isotherm:

The Freundlich isotherm describes the adsorption on heterogeneous surfaces. The following equation is related to the Freundlich model isotherm (23-24):

$\text{Log } q_e=\log C_e+\log K_f$ (4)

Where q_e is the RB19 dye concentration on the SiO_2 nanoparticles (mg/g) at equilibrium, C_e is the concentration of RB19 dye in solution at equilibrium (mg/L) and K_f is the Freundlich constant. The results of the isotherm studies are presented in Table 2.

Kinetic studies:

In this study, the pseudo-first-kinetic and pseudo-second-kinetic models were applied. These models are useful to determine the rate of the adsorption process. The pseudo-first-order rate equation is expressed as following (25-26):

$$\log(q_e - q) = \log q_e - k_1/2.3 \quad (5)$$

Where; q_e and q are related to RB19 dye adsorption capacity (mg/g) onto the SiO_2 nanoparticle (mg/g) at equilibrium and at time t (min), respectively; and k_1 is the constant rate of the pseudo-first-order kinetic model (min^{-1}).

The pseudo-second-order model equation is calculated by below equation (27):

$$dq/dt = k_2 (q_e - q)^2 \quad (6)$$

Where; K_2 is the constant rate related to the second order model (g/mg/min). The plots of log (q_e–q) versus (t) are used to calculate the k_1 while the k_2 is calculated from the plots of t/q_t against (t).

Table 2) Isotherm parameters related to the reactive blue 19 dye adsorption on to SiO_2 nanoparticles

	Langmuir			Freundlich		
temperature	q_m(mg/g)	K_L(L/mg)	R^2	n	K_f	R^2
20℃	13.4	0.626	0.945	3.065	39.38	0.8597
30℃	14.37	0.816	0.9466	3.82	34.97	0.8304
40℃	12.48	0.516	0.9407	2.63	43.54	0.8729
50℃	11.5	0.432	0.9349	2.28	48.65	0.8803
60℃	10.61	0.368	0.9297	2.01	55.03	0.8877

Table 3) Kinetic parameters related to the adsorption of reactive blue 19 dye on to SiO_2 nanoparticles

Pseudo-first kinetic			Pseudo-second-kinetic		
q_e(mg/g)	K_1(min^{-1})	R^2	**q_e(mg/g)**	K_2(mg/g.min)	R^2
104.9	0.17733	0.763	**3.46**	0.023	0.9985

Discussion

Effect of SiO_2 nanoparticles dosage:

The adsorbent dosage has identified as most effective parameters on the adsorption of pollutants. As it was detected, although the dye removal efficiency increases by increasing the adsorbent dosage however the adsorption capacity decreases. It is expressed that the increasing of the adsorption surface site can be the logical reason for the higher dye removal efficiency. In contrast, the higher unsaturated adsorption sites on adsorbent surface are attributed to decreasing the adsorption capacity (28). Mahmoodi et al conducted a study to investigate the nanoparticles ability in direct dyes efficiency. They have found that the dye removal efficiency is developed by increasing the adsorbent dosage while the adsorption capacity decreased. They declared that the higher dye removal efficiency by increasing the adsorbent dosage can be due to increase the adsorbent surface and the accessibility of more adsorption sites. They have also reported that the declining of the adsorption capacity is due to overlapping or aggregation of adsorption sites which is led to the decrease the available total adsorbent surface area and to increase in diffusion path length (29).

Effect of pH:

This parameter plays an important role in adsorption processes. The above results showed that the RB19 dye removal efficiency is higher in acidic pH and the increasing of pH is led to decline the dye removal efficiency. Since the RB19 dye belongs to anionic dyes group; therefore, its molecules is negatively charged. As it is known, the surface of the adsorbent is positively charged in acidic pH; thus, the electrostatic attraction occurs between the negatively charged molecules of dye and the surface of the adsorbent which can increase the removal efficiency. The similar results for dye removal by other adsorbents were observed in other studies (30-31). The adsorption of reactive dye was evaluated by Asadi et al; they observed that the pH has an important role in the dye removal efficiency. they has also observed that the higher dye removal efficiency is obtained at pH=3 which is in consistent with our study (32).

Effect of contact time and temperature:

The contact time and temperature are other parameters which are influencing the dye adsorption process. The obtained results revealed that both two parameters have direct effects on RB19 dye removal and they can enhance the dye removal that they are similar to other studies (33-34). The increasing of dye removal efficiency with temperature indicates that the RB19 dye adsorption is endothermic naturally (35). The higher dye removal efficiency at initial time of the process is related to the large number of the adsorption surface site at the start of this process. As it was observed that the dye adsorption process reached to equilibrium within 90 min (36). Ahuja et al has conducted a study to evaluate the dye from aqueous solution, using zero valent iron nanoparticles and observed that the contact time has a significant effect on dye removal efficiency. They found that dye removal efficiency, which enhanced by increasing the contact time, is agree with the results of this study (37).

The effect of initial RB19 dye concentration:

The study of the effect of concentration of RB19 dye indicated that the removal efficiency decreases by increasing the initial dye concentration. The dye removal efficiency is higher at the lower dye concentration but it decrease by increasing the dye concentration. The highest RB19 dye removal was observed to be 96.45% for 50mg/L dye but it is clear that the lowest dye adsorbed per adsorbent mass is 12.86mg/g. The initial dye concentration of 50mg/L was selected as an optimum concentration. the lowest dye removal and highest dye adsorbed per adsorbent mass was observed to be 68.27% and 455.133mg/g, respectively which obtained for dye concentration of 500mg/L. these results are in consistent with the results of other studies (38). Behnajady et al has surveyed the ability of TiO_2-P_{25} nanoparticles for the adsorption of AR27 dye and they found that the dye elimination efficacy decreased from 97% to 39% with increasing of initial concentration of AR27 from 10 to 80mg/L. they reported that this event can be due to the saturation of adsorption sites onto TiO_2-P_{25} nanoparticles (39).

Isotherm studies:

As it was observed in above, Langmuir and Freundlich models were applied to study the RB19 dye adsorption isotherm onto SiO_2 nanoparticles. The results of isotherm studies were presented in table 2. The q_{max} and k_L are determined from the plot of (C_e/q_e) versus C_e and the K_f and n are calculated from the plot of log q_e versus log C_e. The higher amount of regression coefficient (R^2) is indicative of the better model to describe the equilibrium data. As it can be seen in table 2, the equilibrium data were studied at 20, 30, 40, 50 and 60 ℃. The R^2 values achieved from the Langmuir model were higher than its values for Freundlich in all temperatures which indicated that the equilibrium data are best fitted on the Langmuir model. It is also observed that the amount of R^2 for the Langmuir model at 30 ℃ is greater than other R^2 values of this model which indicates that the highest monolayer adsorption capacity obtained from the Langmuir model in this temperature (14.37mg/g). These results are in consistent with the results of other studies (40-41).

Kinetic studies:

As it was observed, the studying of the kinetic of RB19 dye adsorption was performed pseudo-first-kinetic and pseudo-second-kinetic models and the results obtained from these studies were represented in table 3. As it can be seen, the R^2 obtained from the pseudo second-order kinetic model is higher than this value for the pseudo-first-kinetic which means that the uptake process follows the pseudo-second-order (R^2>0.998) and the data are best fitted on the pseudo second-kinetic models. These results are agreed with the results of other studies (42-43).

Conclusion

In this work, RB19 dye removal efficiency by SiO_2 nanoparticles were evaluated in batch adsorption system. The effect of parameters including contact time, pH, adsorbent dosages, initial RB19 dye concentration and temperature were investigated and observed to be effective on the adsorption process. It was distinguished that the increasing of contact time, adsorbent dosage and temperature (up to 30) had positive effects on dye removal efficiency and can increase the dye removal efficiency while the increasing of pH and dye concentration decrease the dye removal efficiency. The optimum amount of contact time, pH, adsorbent dosage, dye concentration and temperature was found to be 90 min, 5, 0.75g/L, 50mg/L and 30, respectively. The highest dye concentration was observed to be 95.72%. The equilibrium data were best fitted on pseudo-second kinetic and Langmuir isotherm model. The highest monolayer adsorption capacity (14.37mg/g) was obtained at 30 ℃. It can be concluded that SiO_2 nanoparticles have a significant potential for dye removal from aqueous solution.

Footnotes

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Conflict of Interest:

The authors declared no conflict of interest.

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