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Study of the effectiveness of the third generation polyamide-amine and polypropylene imine dendrimers in removal of reactive blue 19 dye from aqueous solutions

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Abstract

Background: Dye and colored materials cause health risks in water and therefore, must be removed from water supplies and wastewater. The aim of this study was to evaluate the effectiveness of the third generation poly(amidoamine) (PAMAM) and poly (propylene imine) dendrimers (PPI-G3) in the removal of reactive blue 19 (RB19) dye from aqueous solutions and determine the optimum conditions for the removal.

Methods: This study was performed in a laboratory and batch scale. In this study, synthetic wastewater was examined with three different concentrations of RB19 (25, 50, and 100 mg/L), different pHs (3, 7, and 10), various amounts of dendrimer (0.4, 0.8, 1.2, and 1.6 g/L), and at different times (15, 30, and 60 minutes) during the adsorption process. The remaining amount of dye was measured by spectrophotometer at 592 nm wavelength. Langmuir and Freundlich isotherms were also tested.

Results: The results showed that by increasing the reaction time and adsorbent dosage, the rate of dye removal increased while by increasing the initial dye concentration and pH, the dye removal efficiency was significantly decreased. In this study, with increase of pH from 3 to 10, dye removal efficiency at a concentration of 25 mg/L, decreased from 72% to 20% and 88% to 17% by PAMAM and PPI dendrimers, respectively. Excel software was used for data analysis.

Conclusion: Both adsorbents had a good dye removal efficiency, but PPI dendrimer was more effective in removing RB19. Adsorption data followed the Langmuir isotherm.

Keywords: Adsorption, Wastewater, Dendrimer, Polyamide amine, Polypropylene imine

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Introduction

The dye is one of the most important pollutants in industrial wastewater that is widely used in industrial sectors such as textile, leather, cosmetics, paper making, food processing and pharmaceutical industries (1,2). Reactive dyes are water-soluble and anionic that are largely used in the textile industry. Reactive blue 19 (RB19) with its special chemical structure is one of the anthraquinone dyes which are the most widely used in textile, after azo dyes. This dye has relatively low fixation efficiency on the fiber and thus, considerable amounts of waste can be imported to wastewater. Since this dye is stable in natural environments and highly resistant to light, chemical agents and biological degradation, discharging wastewater containing dye into the environment will cause irreversible and serious problems (3,4). Due to the low bio-decomposition of dyes, the conventional biological wastewater treatment processes are not effective in dye wastewater treatment. The colored wastewater is usually treated by physical or chemical methods such as coagulation and flocculation, precipitation, adsorption, membrane filtration, electrochemical techniques, ozonation and etc. The adsorption process is one of the physicochemical processes developed due to the low cost, ease of operation, flexibility, and simplicity for treatment of wastewater containing dye, organic materials and metals (5,6). One of the newest adsorbents is dendrimer. Dendrimer is a Greek term composed of two parts: "dendron" meaning "tree" and "meros" meaning "part" and implies the tree-like branched structure of this group of compounds (7). The structure of dendrimers is composed of 3 parts: An inner core, shell (generation) comprising repeating units, and terminal functional groups (8,9). Depending on the number of generations, dendrimers include a wide range of dimensions often in the nanometer range so that in the third generation (G3), their dimension is equal to insulin hormone (5). Dendrimer as a nanomaterial (7) and due to its large surface, has high and effective capability to remove the contaminants (10). The main features of dendrimers are having many active endgroups as well as the blank spaces between the branches of the internal connector that have the ability to attract and imprison foreign molecules such as water, dye and other materials by chemical bonding and encapsulation (9).

These polymers are of great importance in various fields such as medical and industrial applications and pharmaceuticals as a material to eliminate germs and in the field of water and wastewater treatment, to remove pollutants (11,12). Table 1 shows different types of dendrimers including polyamide amine-organosilicon (PAMAMOS), poly(amidoamine) (PAMAM), polypropylene imine (PPI), chiral, liquid crystal, Tecto, hybrid, multilingual and Micellar (9). Many studies have been done on the use of dendrimers to remove pollutants from the environment. Patel et al in a study examined the removal of metal ions Cu^{2+}, Ni^{2+} and Zn^{2+} of water using G1, G2, and G3 dendrimers with the number of hydroxyl groups 8, 32 and 128 as adsorbents (13). In another study, PPI and chitosan were evaluated as a biopolymer to remove reactive black 5 and reactive red 198 (5). The microparticles of polyamide imine-chitosan have been used as an adsorbent for the removal of reactive blue 21 (14). In this study, the G3 (PAMAM and PPI) dendrimers were used as the adsorbents for the removal of RB19 dye from wastewater.

Material and Methods

This study was conducted discontinuously (batch) on a laboratory scale. In this study, the third generation PAMAM and PPI dendrimers were purchased from the Institute of Tehran Paint, and their structural form is provided in Figure 1 (15). The dye used in this study, was purchased from Sigma-Aldrich and other chemicals used

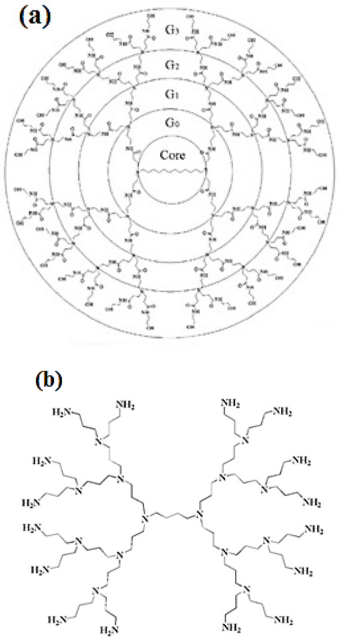

Figure 1. PAMAM (a) and PPI (b) dendrimers.

in the experiments, were obtained from Merck (Germany). Characteristics of RB19 dye are provided in Table 1 (16). In order to prepare the wastewater, the dye stock solution was created by solving the appropriate amount of RB19 powder in distilled water and then, different concentrations (25, 50, and 100 mg/L) were examined. In the present study, the influence of different adsorbent dosage (0.4, 0.8, 1.2, and 1.6 mg/L), pH (3, 7, and 10), initial dye concentration (25, 50, and 100 mg/L) and reaction time (15, 30, and 60 min) on dye removal was investigated. In order to mix and make a proper contact between the adsorbent and dye, a mechanical shaker at 250 rpm was used. The adsorbent was separated from the wastewater using membrane filter (0.45 micron in diameter). Sulfuric acid and sodium hydroxide 1N were used to adjust the pH. The remaining dye concentration was measured using a UV/Vis spectrophotometer (Optima SP-3000 Plus model, Japan) at 592 nm. The amount of dye adsorbed at equilibrium time (q_e) and dye removal efficiency (E) were respectively calculated using equations (1) and (2) (16).

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

$$E = \frac{C_0 - C}{C_0} \times 100 \quad (2)$$

In these equations, C_0 and Ce respectively show the initial and final concentration of the dye (mg/L), V, volume of the solution (L), M, adsorbent mass (g), q_e, adsorption capacity at equilibrium time (mg/g), and E, dye removal efficiency (%) (17). Langmuir and Freundlich adsorption isotherms were examined to describe the relationship between the adsorbent and the colored solution. Langmuir and Freundlich isotherm models are described using

Table 1. Characteristics of reactive blue 19 dye

Chemical Formula	$C_{22}H_{16}N_2Na_2O_{11}S_3$
Molecular Weight	626.5 g
Trade Name	Remazol Brilliant Blue R
Chemical Structure	

equations (3) and (4):

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

$$\log q_e = \log k_f + \frac{1}{n} \ln C_e \quad (4)$$

Where q_m and b are Langmuir constants that are related to the maximum adsorption capacity (mg/g) and energy (L/mg). K_f and n, are Freundlich constants which show adsorbent capacity and intensity. Microsoft Excel software was used for data analysis and drawing charts.

Results

Determining the effect of pH and contact time on dye removal

The effect of pH on the removal of RB19 in the presence of adsorbent dose (0.4, 0.8, 1.2, and 1.6 g/L) in desired dye concentrations (25, 50, and 100 mg/L) was studied and the results are shown in Figure 2. As shown in this figure, the maximum dye removal efficiency was observed at pH 3.

In the present study, the effect of the retention time (15, 30, and 60 minutes) as one of the independent variables in dye removal was also evaluated. The maximum adsorption for both PAMAM and PPI dendrimers took place 30 and 15 minutes respectively.

The effect of adsorbent dosage

Evaluating the effect of adsorbent dosage and determining the optimal dosage of adsorbent are the most important parameters that should be considered in the process of adsorption. The results of evaluation of the effect of adsorbent dosage on the removal of RB19 are shown in Figure 3. According to the findings of the study, by increasing adsorbent concentration from 0.4 to 1.6 g/L, dye removal efficiency by PAMAM and PPI dendrimers increased from 26% to 72% and from 35% to 88%, respectively. Thus, the optimal dosage of PAMAM and PPI dendrimers was 1.6 g/L because in the higher dosages, dye removal efficiency was constant or there was not a significant increase in dye removal.

The effect of dye concentration on the removal efficiency

To survey dye adsorption efficiency, the effect of dye concentration (25, 50, and 100 mg/L) on the removal efficiency was examined. Considering that adsorption at pH 3 had the highest efficiency, thus in this stage, adsorbent dosage 1.6 g/L and pH 3 were selected for dye adsorption. The results of this study are shown in Figure 4. As presented in this figure, by increasing concentrations from 25 to 100 mg/L, a significant reduction in the rate of dye removal was observed. So that by increasing dye concentration, dye removal efficiency decreased from 88% to 29% by PPI and also from 72% to 23.75% by PAMAM.

Evaluation of reactive blue 19 dye adsorption isotherm

Evaluation of experimental data with the Langmuir and Freundlich models and comparison of their correlation coefficients showed that RB19 dye adsorption obeyed the

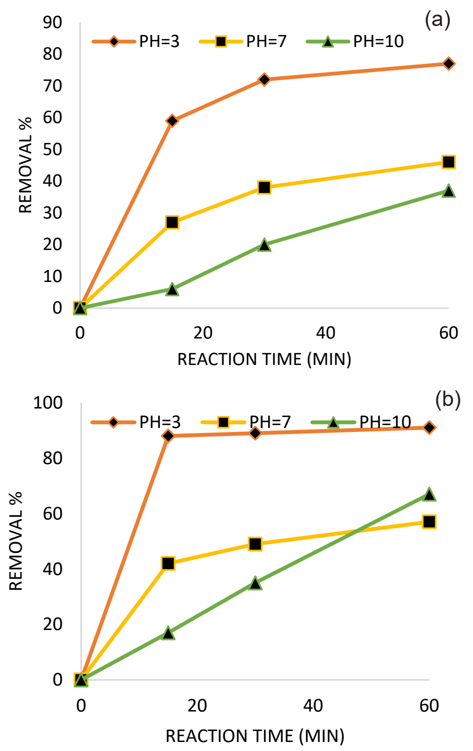

Figure 2. Effect of pH on dye removal efficiency by PAMAM (a) and PPI (b) dendrimers at different times. (dye concentration: 25 mg/L, dendrimer dosage: 1.6 g/L).

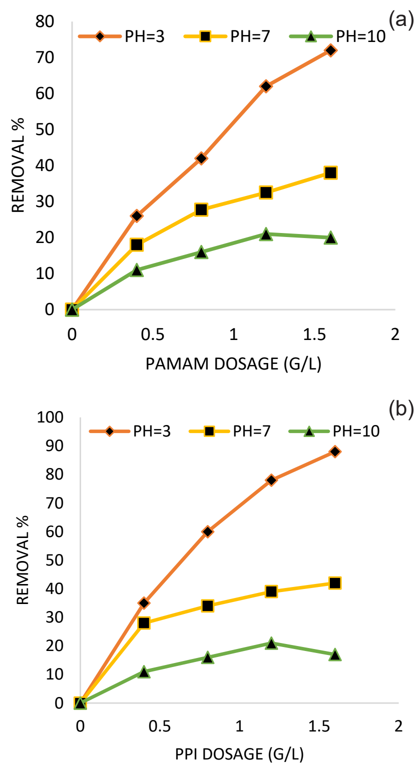

Figure 3. Effect of PAMAM (a) and PPI (b) dendrimers dosage on dye removal at different pHs (retention time: 30 min, dye concentration: 25 mg/L).

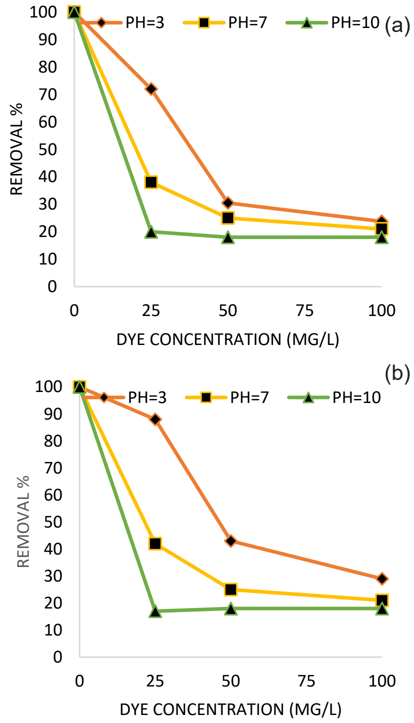

Figure 4. The effect of dye concentration on dye removal efficiency by PAMAM (a) and PPI (b) (retention time: 30 min, dendrimer dosage: 1.6 g/L).

Langmuir isotherm by the G3 PAMAM (R^2=0.94) and PPI (R^2=0.98) dendrimers (Figure 5). Maximum adsorption was 46.8 mg/g for PAMAM and 1029 mg/g for PPI. Values of parameters of Langmuir and Freundlich isotherms are presented in Table 2.

Table 2. Parameters of Langmuir and Freundlich models for RB19 dye adsorption by PPI and PAMAM dendrimers

Langmuir isotherm			Freundlich isotherm		
	PAMAM	PPI		PAMAM	PPI
Q_0 (mg/g)	46.8	1029	K_F	779.8	896.5
K_L	0.3049	0.233	n	3.18	4.29
R^2	0.9435	0.8965	R^2	0.84	0.89

Adsorption kinetics

To investigate the mechanism of adsorption of the RB19 on PPI and PAMAM, the pseudo-first- and second-order models were used. Table 3 shows the Kinetic parameters of the pseudo-first- and second-order models. The results showed that the R^2 values of the pseudo-second-order model (> 0.99) was higher than the pseudo-first-order (>0.90) for RB19 on PPI and PAMAM.

Discussion

Determining the effect of pH and contact time on dye removal

One of the most important effective factors in the adsorption process is pH that can affect adsorption capacity through affecting the degree of ionization and surface characteristics of adsorbent (18). The maximum dye removal efficiency was observed at acidic pH. In general, it can be stated that dye removal efficiency increases when the electrostatic attraction between the dendrimer with positively charged amino groups (NH_2) and anionic dye containing negatively charged sulfonic group, R-SO_3^- happens. These groups produce hydrogen at acidic pH. Generally, an increase in the pH leads to a

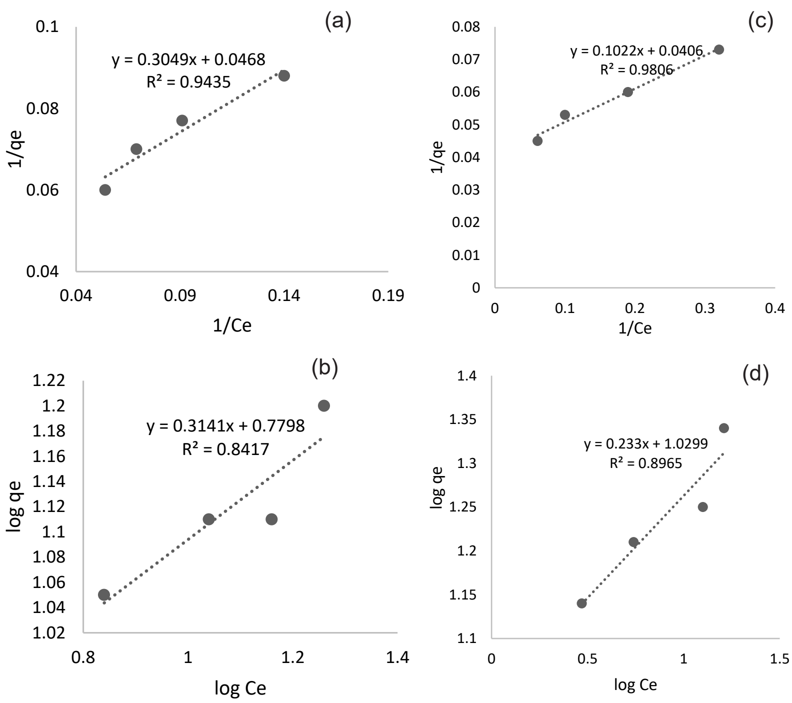

Figure 5. Adsorption isotherms of dye by PAMAM: Langmuir (a) and Freundlich (b) and by PPI: Langmuir (c) and Freundlich (d).

Table 3. The first-order and second-order adsorption rate constants, calculated adsorption capacity ($q_{e, cal}$) and experimental adsorption capacity (q_e) values of RB19 dye on the adsorbents

Adsorbent	$q_{e, exp}$ (mg/g)	First-order kinetic model			Second-order kinetic model		
		k_1 (1/min)	$q_{e, cal}$ (mg/g)	R^2	k_2 (g/mg/min)	$q_{e, cal}$ (mg/g)	R^2
PPI	15.31	0.0549	9.459	0.9062	0.045	15.037	0.9972
PAMAM	13.28	0.0457	9.351	0.9111	0.031	12.82	0.9918

decrease in the sites in the positively charged dendrimer and thus, a decrease in the dye removal efficiency (19,20). In general for anionic dyes, adsorption capacity increases by reducing the pH of the solution while it is reverse for cationic dyes (20). Almasian et al invetigated anionic dye removal by polyacrylonitrile and PAMAM. In this study, adsorption was higher at low pH and pH 2.1 was selected as the optimal pH (21). In the study of Salimpour Abkenar et al on anionic and cationic dye removal by PPI and flax, anionic dyes had better performance at pH 3 and cationic dyes, at alkaline pH (pH 11) (20). Yiyun and Jiepin used different generations of PAMAM dendrimers for removal of methane dyes, and the results showed that the removal of methylene blue and fuchsia acid was effective at high pH (22).

The maximum adsorption for both PAMAM and PPI dendrimers took place in the first 30 and 15 minutes, respectively. In fact, at the beginning of the adsorption reactions, vacant sites in the adsorbent effectively remove the dye, but after equilibrium, due to repulsive forces between molecules of adsorbent, removal efficiency reduces (23,24). In a study by Sadeghi-Kiakhani et al, G2 PPI dendrimer and chitosan were used in order to remove the reactive dye, in which by increasing the reaction time, the dye removal increased and the maximum dye removal efficiency was observed in the 30 minutes of the reaction (5). In a study by Amouei et al, removal of reactive orange 16 was evaluated by sunflower stalks. The results showed that by increasing the reaction time, color removal efficiency increased. In general, by increasing the contact time, the amount of adsorption to reach equilibrium time increased and then, remained almost constant (23).

The effect of adsorbent dosage

Evaluating the effect of adsorbent dosage and determining the optimal dosage of adsorbent are the most important parameters that should be considered in the process of adsorption. According to the findings of our study, by increasing adsorbent concentration until optimal dosage (1.6 g/L), dye removal efficiency by PAMAM and PPI dendrimers increased. It could be due to the increased surface area of the adsorbent and the number of available active sites for adsorption (14). In the study of Ghaneian et al on the effect of adsorption of the Russian knapweed flower powderon the removal of RB19, the results showed that by increasing the adsorbent dose from 0.2 to 0.6 g/100 mL, removal efficiency increased from 72% to 82% (16). In a study by Hayati et al, G2 PPI dendrimer was used for the removal of dyes from textile wastewater, and the results indicated that increasing concentrations of dendrimers leads to increased dye removal and the optimal dendrimer concentration was 1.8 g/L (19). In another study on the adsorption of reactive red 198 by titanium dioxide nanoparticles, by increasing the adsorbent concentration, the adsorption efficiency of dye increased and the optimal adsorbent dosage was reported 1 g/L (25), which are consistent with the results reported by Yiyun and Jiepin (22).

The effect of dye concentration on the removal efficiency

The PPI dendrimers can absorb dye molecules both the protonated functional groups ($-NH_3^+$, $>NH^+-$) and the cavities. In fact, due to saturation of adsorbent active sites by the dye molecules, an increase in the dye concentration can reduce adsorption capacity or dendrimer active surfaces (20). The results of Yiyun and Jiepin showed that in low dye concentrations, removal efficiency can be increased (22). In another study by Sadeghi-Kiakhani et al on removal of reactive black 5 and reactive red 198 dyes by polypropylene (PPI) dendrimers and chitosan as biopolymer, under the optimal amounts of process parameters, dye removal was reported 97% and 99% for reactive black 5 and reactive red 198, respectively (5). The effect of crap plant powder on the removal of RB19 dye showed that by increasing the initial concentration of dye, dye removal efficiency decreased in the research of Ghaneian et al (16).

Evaluation of reactive blue 19 dye adsorption isotherm

Adsorption isotherm studies have been carried out on 2 well-known Langmuir and Freundlich isotherm models. Langmuir isotherm model assumes monolayer adsorption onto a surface containing a finite number of adsorption sites of uniform strategies of adsorption with (with no interactions between the adsorbent molecules) no transmigration of adsorbate in the plane surface while Freundlich isotherm model assumes heterogeneous surface and multilayer adsorption (26). Evaluation of experimental data using the Langmuir and Freundlich models and comparison of their correlation coefficients showed that adsorption of RB19 dye obeys the Langmuir isotherm by the G3 PAMAM (R^2=0.94) and PPI (R^2=0.98) dendrimers (Figure 5).

Sayan et al examined thermodynamic adsorption in the removal of RB19 dye by activated carbon under ultrasonic irradiation. The results indicated that adsorption isotherm obeyed the Langmuir model and adsorption of RB19 was endothermic (26). In another study, nanocomposites including copper sulfide and PAMAM dendrimer were used to remove Isma acid fast yellow G dye, and the results indicated that adsorption obeyed Langmuir model and with pH increase, the rate of adsorption decreased (27).

Adsorption kinetics

The results showed that the R^2 values of the second-order kinetics (>0.99) was higher than the first-order kinetics (>0.90) for RB19 onto PPI and PAMAM. Moreover, the q_e value is calculated adsorption capacity for pseudo-second-order model was in agreement with the experimental q_e values than the pseudo-first-order model. Therefore, this study suggested that the second-order model well demonstrated the adsorption of RB19 kinetics. Similar result has been observed in the adsorption of reactive dye onto carbon nanotubes (28), nickel oxide (NiO) nanoparticles (29), palm shell powder, chitosan (30) and biomass (31).

Conclusion

Removal of RB19 dye from aqueous solutions by the third generation PAMAM and PPI dendrimers was investigated. Results of this study showed that by increasing pH, dye removal efficiency decreases and the highest dye removal efficiency was observed at pH 3. Also, by increasing the amount of adsorbent, dye removal efficiency increased. By increasing the initial dye concentration, dye removal efficiency decreased and by increasing the contact time, dye removal efficiency increased. In this study, the mechanism of adsorption of RB19 dye onto the third generation PAMAM and PPI dendrimers was fitted with Langmuir isotherm. According to the results, both adsorbents had good efficiency for dye removal, but PPI was more effective in removing RB19 dye. Adsorption data followed the Langmuir isotherm model. It seems that both dendrimers can be used for removal of different dyes in the textile industry as the environmental-friendly adsorbents.

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Ethical issues

It is confirmed that this manuscript is the original work of the authors. It has not been published, nor is it under review in another journal, and it is not being submitted for publication elsewhere.

Competing interests

The authors declare that they have no conflicts of interests.

Authors' contributions

All authors contributed equally to the study design, data collection, analysis, and interpretation. All authors critically reviewed, refined, and approved the manuscript.

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