

*Original Article***Removal of Reactive Blue 19 dye using a combined sonochemical and Modified Pistachio Shell adsorption processes from aqueous solutions**Somayeh Rahdar¹ Leila shikhe² **Shahin Ahmadi***¹

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(Received: 23 Jan. 2018; Revised: 3 Apr. 2018; Accepted: 27 Jun. 2018)

Abstract

Background and purpose: Reactive Blue 19 dye is one of the major dangers to human health and environment. Hence, the removal of these compounds from polluted water has been considered. This study was an attempt to investigate the removal of RB19 dye from aqueous solution in the presence of modified pistachio shell.

Materials and Methods: In this experimental study, the treatability of water polluted with Reactive Blue 19 dye was investigated by using sonochemical oxidation in a reactor. The sample with the desired concentrations of dye (10-80 mg/L) at PH 2-11 was prepared, then the required concentrations of modified pistachio shell (0.1– 1.2 g/L) was flowed into the reactor intermittently.

Results: The results of the current study showed that 40.26% of the dye was removed in the presence of modified pistachio shell after 20 min at pH 2, modified pistachio shell dose 0.7 g/L, and concentration of 20 mg/L.

Conclusion: The findings showed that the combined process of ultrasonic and adsorption in the presence of modified pistachio shell at optimal conditions of operation can be used as an acceptable option in the removal of dye from waters.

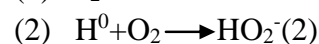
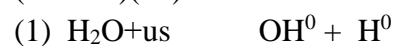
Key word: Reactive Blue 19; Aqueous Solution; Modified Pistachio Shell

Citation: Rahdar S, Shikh L, **Ahmadi Sh***. Removal of Reactive Blue 19 dye using a combined sonochemical and Modified Pistachio Shell adsorption processes from aqueous solutions. Iran J Health Sci 2018; 6 (3): 8-20.

1. Introduction

Dye containing wastewaters generated by industries is the major cause of environmental problems (1). Evacuation of wastewater containing colored materials into the environment and aquatic ecosystem prevents the penetration of light into the water depth. This can also cause disturbance in the process of photosynthesis and the destruction of aquatic plants (1, 2). The textile industry generates very large amounts of textile wastewater (3, 4). Reactive dyes are extensively used in the textile dyeing industry for cotton and polyamide fibers because of their luminous, various colors, high wet fastness, and ease of application (5). Most dyes and pigments are considered non-toxic, although some are not totally harmless (6). Interest in the environmental action of dyes is prompted primarily by concern over their possible toxicity and carcinogenicity (7). They are also aesthetically objectionable for drinking water and can cause allergy, dermatitis, skin irritation, and also agitate cancer and mutation in humans and animals (8-10). Azo dyes are the most widely used dyes in the textile industry, and the research shows that the highest amount of toxicity were found between basic and Diazo direct dyes (11, 12). The pollution is determined by biochemical oxygen demand, chemical oxygen demand, suspended solids, toxicity, high concentration of nutrients, presence of chlorinated phenolic compounds, sulfur and lignin derivatives, etc. (12, 13). Reactive blue 19 (RB 19) textile dye is easily available and commonly used. Researchers are concerned with mutagenic properties of RB 19 due to the

presence of electrophonic vinyl sulfone groups (14). Therefore, it is highly necessary to reduce dye concentration in the wastewater. The conventional methods for treating dye containing wastewaters are electrochemical treatment, coagulation and flocculation, liquid-liquid extraction, and adsorption (15, 16). Ultrasonic irradiation of aquatic environment can result in the increase and destruction of gas bubbles (Cavitations), so producing high transient temperature and pressure leads to the formation of free radicals (OH, OOH) via thermal breakup of water and oxygen. These radicals penetrate into water and oxidize dissolved organic compounds (17, 18). The mechanism proposed for the production of OH by ultrasonic (US) are given as Eq. (1 and 2)(19): \longrightarrow



Iran with the production of more than 44% of the world's total production is the largest producer and exporter of pistachios. So the waste generated from the processing and production section is high, and can be used as a cheap adsorbent (20). The main purpose of current research was to study the sonochemical degradation of reactive blue 19 dyes from aqueous solutions in the presence of MPS. The effects of pH, contact time, initial dye concentration and dosage on adsorption capacity and percentage of dye removal from aqueous solutions using ultrasonic process were investigated.

2. Materials and Methods

2.1. Materials

Reactive blue19 (RB19) is an anionic dye with molecular weight of 626.54g/mol and maximum absorption (λ_{max}) of 594 nm. The RB19 ($\text{C}_{22}\text{H}_{16}\text{N}_2\text{Na}_2\text{O}_{11}\text{S}_3$) used in this work was of analytical grade

(Sigma-Aldrich-US). The chemical structure of RB19 is shown in Figure 1.

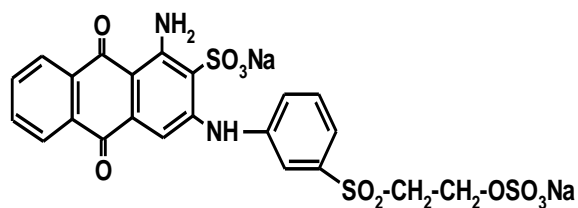


Figure 1. The chemical structure of RB19

2.2 Pilot ultrasonic

Reactor of determined surface included a digital ultrasonic appliance that was made of Plexiglas with volumes of 3.7 L, input energy per unit 2.5 w/cm^2 , and input power 500W including 50 mL samples in bath with ultrasonic waves. It is schematically shown in Figure 2 (B).

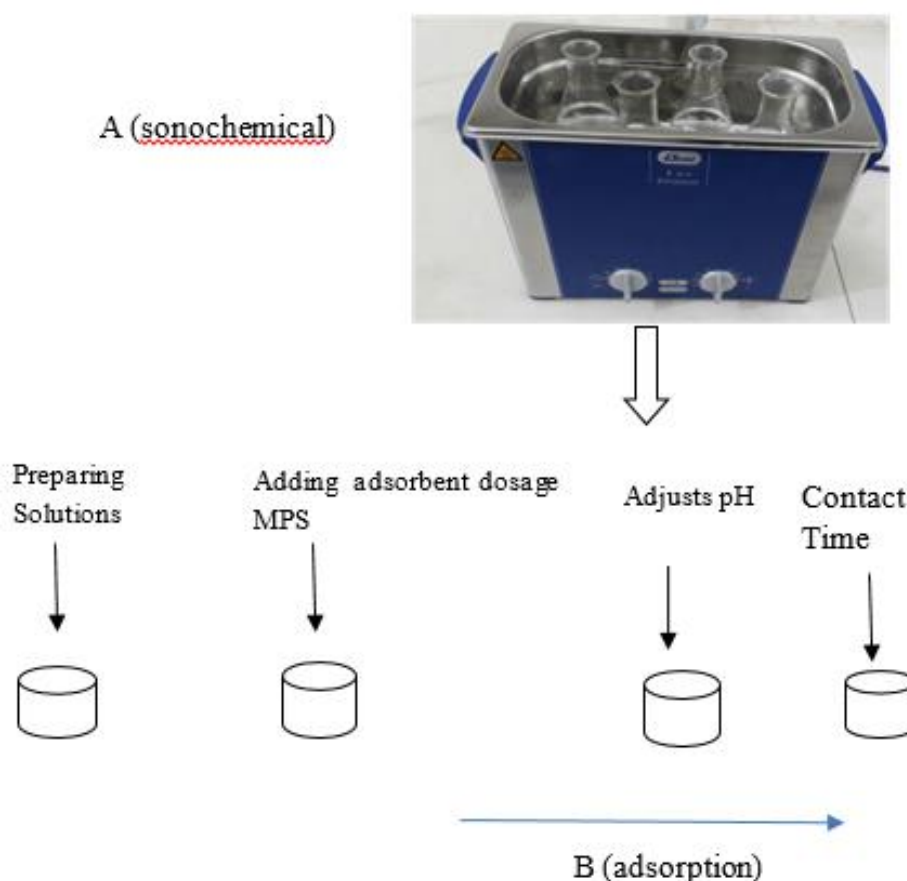


Figure 2. The process of preparing (A) and the schematic of sonochemical process (B)

2.2.Adsorbent Preparation

Pistachio shell (PS) was collected from Rafsanjan city, Iran. Samples were washed with distilled water and after drying, the PS was grinded and sieved by using a 100 mesh sieve. It was placed in a furnace at temperature of 550° for 60 min. The favorite diameter of ash particles was 0.15 to 0.85 mm. PS was then activated by using

20 ml of sodium hydroxide (30%). In the next step, samples were washed with distilled water and dried in oven in the temperature of 105°C for 24 hours (21).

2.3. Analysis

Fourier-transform infrared spectroscopy (FTIR) of MPS was done on a Perkin Elmer Spectrum ($4000\text{-}400 \text{ cm}^{-1}$) at room temperature.

2.4. Batch Adsorption Technique

Four process variables have been studied in this research which included the initial pH of solution (2- 11), adsorbent dosage (0.1- 1.2 g), contact time (10-60 min), and initial concentration of RB19 (10-80 mg/L). For this purpose, samples of 50 ml of different concentrations of RB19 were prepared. The final pH was measured using a MIT65 pH meter and poured into Erlenmeyer and added MPS and inside the ultrasonic apparatus under ultrasound waves (60Hz) according to the set different times was refined. The residual concentrations were also measured using UV visible spectrophotometer (Shimadzu Model: CE-1021-UK). To express the percentage of dye removal, the following Eq. (3) was used (22, 23):

$$\%R = \frac{(C_0 - C_f)}{C_0} 100$$

Where; C_0 and C_f represent the initial (before adsorption) and final (after adsorption) dye concentrations, respectively.

The amount of RB 19 adsorbed (mg/g) was calculated under different conditions using Eq. (4) (24):

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

In this equation, C_0 is the initial concentration; C_e denotes the liquid-phase dose of RB19 at equilibrium in mg/L, V denotes the solution volume (L), and M refers to the quantity of the adsorbent (g).

2.5. Adsorption Kinetics

Kinetic studies for process were performed at the point of dye removal from aqueous solutions under the optimum conditions at room temperature (298 K). Kinetic models are used to examine the rate of the adsorption process and the potential rate

controlling step. The pseudo-first-order rate equation is expressed as Eq. (5) (25):

$$\text{Log}(q_e - q_t) = \text{Log}(q_e) - \frac{k_1}{2.303} t$$

Where q_e and q_t are the amounts of RB19 adsorbed (mg/g) at equilibrium and at time t (min), respectively, and k_1 is the rate constant of adsorption (min^{-1}). Values of k_1 were calculated from the plots of $\log(q_e - q_t)$ against (t) at different concentrations. Also, the pseudo-second-order rate equation has been given by Eq. (6) (26, 27):

$$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{t}{q_e}$$

Where, K_2 is the second order rate constant ($\text{g mg}^{-1}\text{min}^{-1}$), q_t and q_e are the amount of the adsorbed on the adsorbent (mg/g) at equilibrium and at time t , respectively.

The kinetic studies were carried out by taking decolorisation into consideration under the optimum conditions for process. In this study, the kinetic model developed by Behnajady et al. (Eq.7) was used (28).

$$\frac{C}{C_0} = 1 - \left(\frac{t}{m + b.t} \right)$$

where C_0 is the initial concentration of RB19 (mg/L); C is the concentration at time (t) (mg/L); m and b are the two dimensionless characteristic constants of model relating to the initial removal rate and maximum oxidation capacities, respectively. To determine the characteristics of m and b , Eq.7 can be linearized as follows (Eq.8):

$$\frac{t}{1 - \left(\frac{C}{C_0} \right)} = m + b.t$$

A straight line was obtained by plotting $t / (1 - C/C_0)$ for dye removal contact time t , and m and b , the coefficients of Eq.6 were obtained from the intercept and the slope of the line, as reported, in Table 1.

3. Results

3.1. Adsorbent Characterization

The FTIR analysis of the modified pistachio shell (MPS) was done before and after adsorption to observe the functional groups participation in the process of adsorption. This was also done to evaluate the interactions between the adsorbate (dye) and adsorbent (MPS). The infrared transmittance was plotted against the wavelength (cm^{-1}). The FTIR analysis on the MPS before adsorption (Figure 3(a)) indicated the presence of C – H bending of alkyne (740 and 876 cm^{-1}), C – N stretch of aliphatic amines

(1194 cm^{-1}), C – O stretch of ether (1282 cm^{-1}), C = C stretch of alkenes (1566 cm^{-1}), and C = O stretch of carbonyl groups (1702). The intensities of C – H bending, C = C and C = O stretching vibrations, which are very strong bonds (adsorption sites), were shifted from 876 , 1566 , and 1702 to 872 , 1563 , and 1692 cm^{-1} , respectively, after adsorption (Figure 3(b)). Some of the absorption bands (C – O, and C – N stretching vibrations) disappeared, and new peaks were formed after adsorption which shows the involvement of the functional groups in the adsorption process.

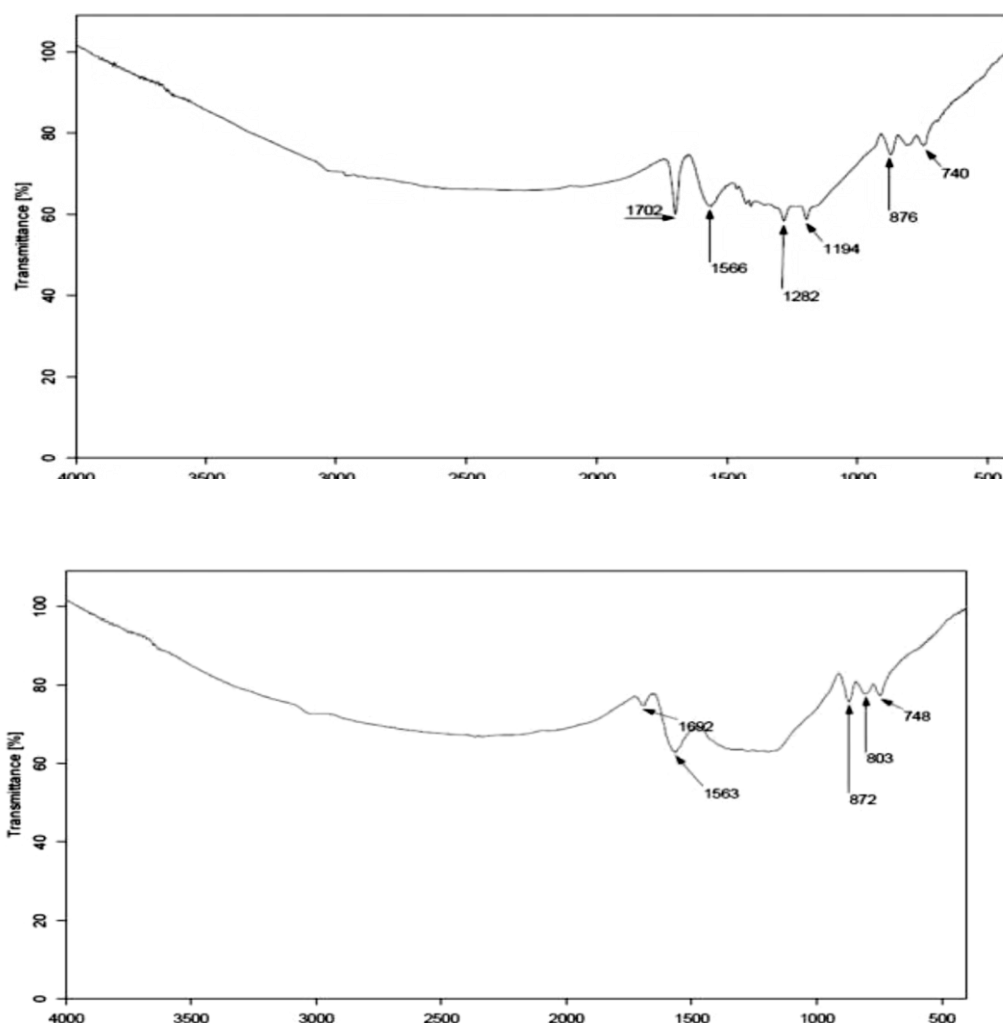


Figure 3. FTIR spectra of MPS (a) before and (b) after adsorption process

3.2. Effect of pH

To determine the effect of pH on RB19 removal, pH was altered from 1 to 11 for 50 ml of the dye solution at fixed concentration of 40 mg/L, contact time of 45 min and adsorbent dosage of

0.5g/L. The results are summarized at Figure 4. There was a decreasing trend in RB19 adsorption with an increase in pH from 2 to 11, with maximum efficiency of 45.65% at pH 2.

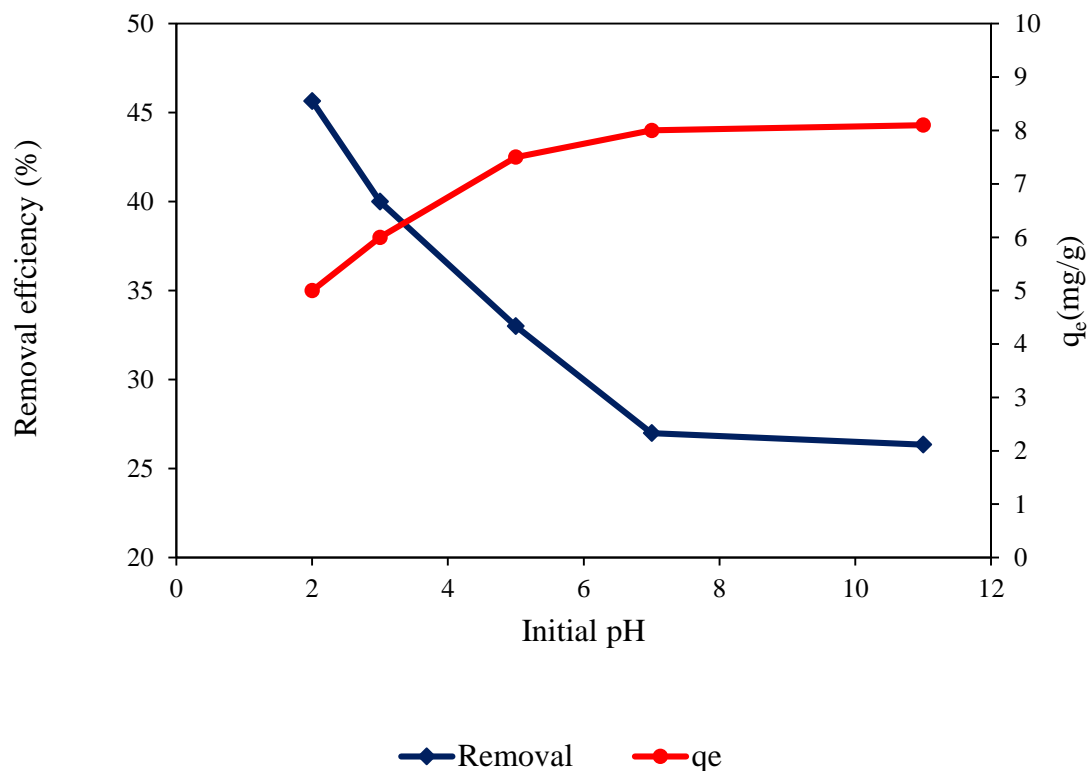


Figure 4. The effect of solution pH on RB19 adsorption by MPS (MPS dose = 0.5 g/L, contact time =45 min, and RB19 concentration = 40mg/L).

3.3 Effect of amount of adsorbent

The effect of amount of adsorbent on RB19 removal was investigated. To determine the effect of MPS dosage on RB19 removal, adsorbent dosage was changed from 0.1 to 1.2 g/L for 50 ml of

the dye solution at fixed concentration of 40 mg/L, contact time of 45 min, and pH2. Figure 5 shows that the RB19 elimination capacity increased with increasing adsorbent dose and the decline was continued with slope almost constant until dose of 1.2g/L.

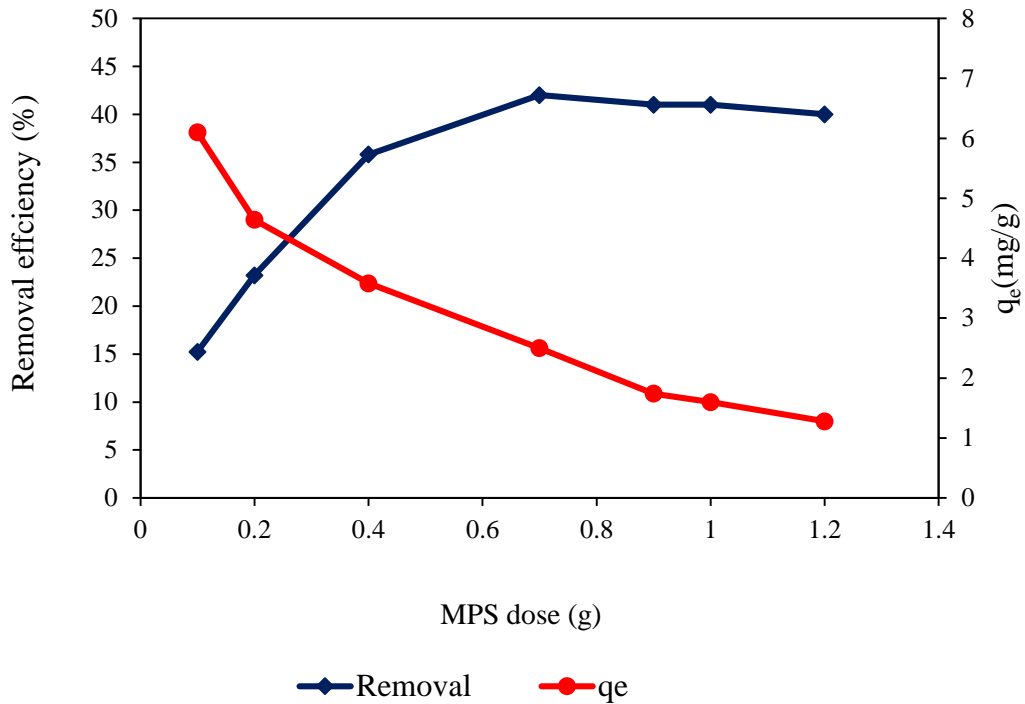


Figure 5. Effect of adsorbent dose on RB19 adsorption by MPS (RB19 concentration = 40mg/L, initial pH = 2 and contact time = 45 min)

3.4 Effect of initial dye concentration

Effect of different initial dye concentration (10–80 mg/L) on the adsorption of RB19 on

MPS (contact time: 45 min, RB19 concentration: 40 mg/L and dosage: 0.7 g/L) is shown in Figure 6.

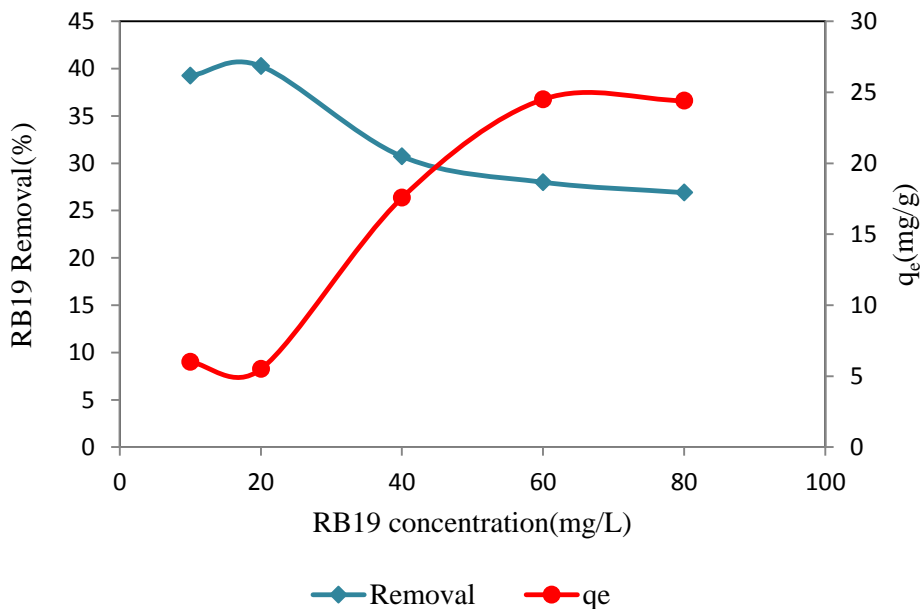


Figure 6. Effect of RB19 concentration on RB19 adsorption by MPS (Time = 45 min, initial pH= 2, and dosage: 0.7g/L).

3.5. Effect of time contact

The effect of contact time on the amount of RB 19 on MPS was also investigated at different contact times (10-60 min) at RB19 initial concentration of 60 mg/L, pH of 2, and

adsorbent dosage of 0.7 g/L at temperature of 298 K. Figure 7 shows the effect of contact time on RB19 removal. Maximum efficiency of RB19 removal was achieved in 45 min.

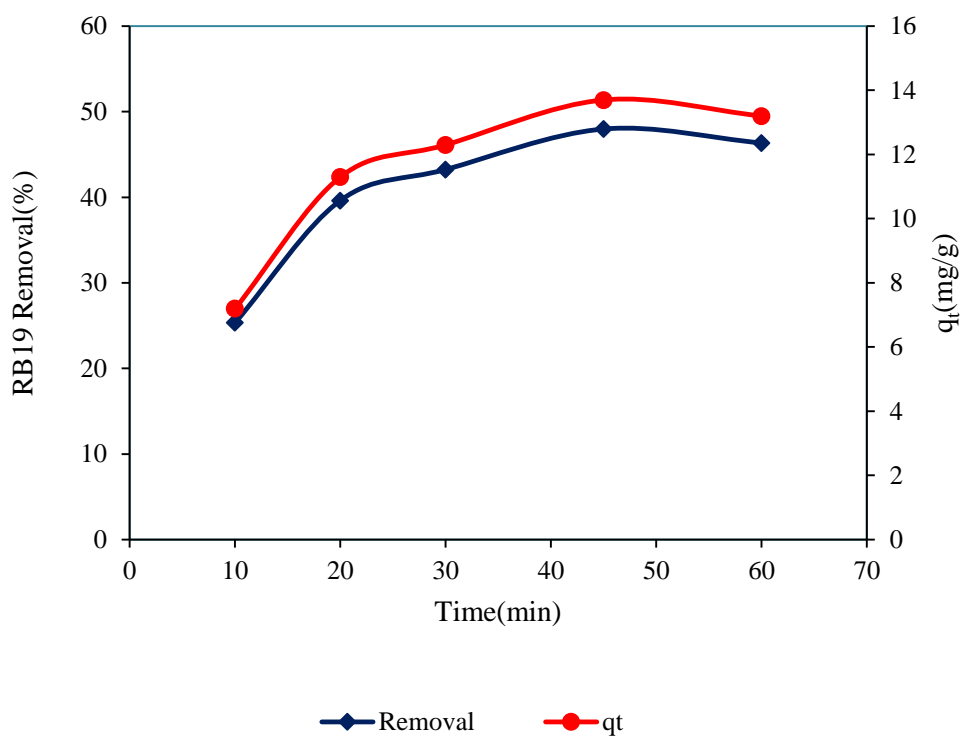


Figure 7. The effect of contact time on RB19 adsorption by MPS RB19 concentration = 20mg/L, initial pH= 2 and dosage: 0.7g/L).

The velocity constant of pseudo second-order, pseudo first-order and decolorisation is presented in Table 1.

Table 1. The coefficients of determination and characteristic constants of the kinetic model

C_0 (mg/L)	Pseudo second-order			Pseudo first-order			Decolorisation		
	K_2	q_e	R^2	K_L	q_e	R^2	b	m	R^2
40	0.005	16.7	0.99	0.13	36.2	0.9228	1.8016	17.076	0.987

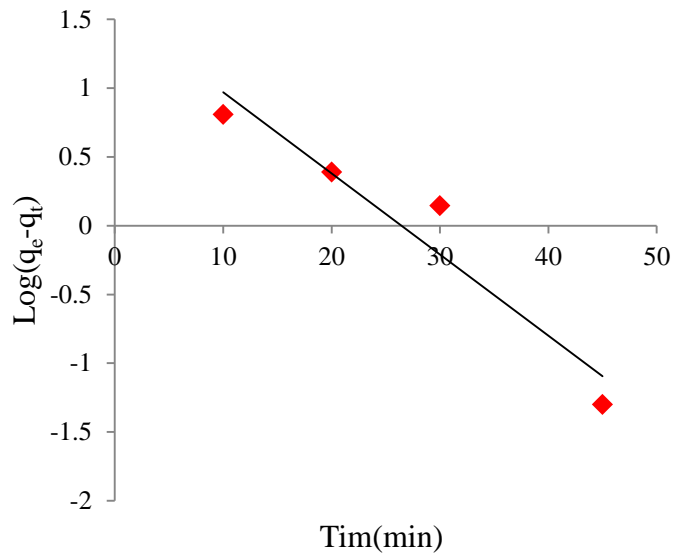


Figure 8. Kinetic isotherms pseudo-first-order

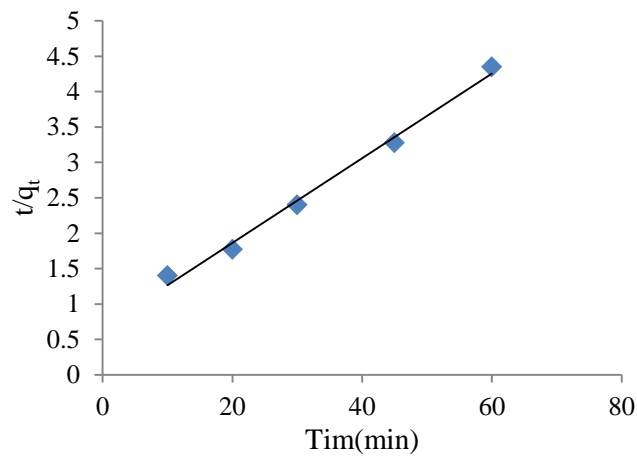


Figure 9. Kinetic pseudo-second-order

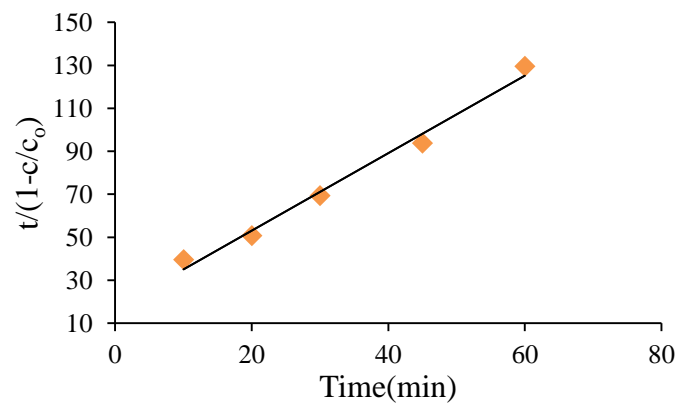


Figure 10. Relationship between $t / (1-C / C_0)$ and oxidation time t in optimum conditions for dye removal

4. Discussion

The initial pH of solution is one of the important parameters in chemical processes. The reaction occurs between the hydroxyl ions and the positive cavity where the positive cavities were formed at low pH (26). The removal of color significantly decreased with an increase in pH, because under low acid conditions, the dye encounters maximum ionization pH of molecular range that results from an increase in electrostatic gravity between anionic and cationic species (29). Also, in the high pH range, OH ions create water-soluble compounds, and delay the adsorption of paint. It should be noted that the production of OH radicals in the acid environment is slightly higher than the alkaline environment and their stability is higher, therefore, advanced oxidation processes in the acid environment are superior to the alkaline environment, especially in pH higher than 12, radical so OH are superior, since which, in alkaline conditions the oxidation potential of OH is reduced. This result was found to be consistent with the previous studies conducted (30, 31). The study of the removal of reactive dyes using sonochemical process by Eslami et al. 2015 showed that the maximum efficiency is related to pH 2 (32). Studies have shown that chemical sonochemical decomposition in the presence of adsorbent substances leads to the decomposition of organic compounds directly or indirectly, leading to the production of radical hydroxyl in the combination of the process of sonolysis and additives. The degradation of the additive particles reduces the particle size and increases the surface area, thus the efficiency of

the decomposition increases directly in the presence of additive substances after reaching the highest constant efficiency. At the same time, with the increase of adsorbent, the amount of production of hydroxyl oxidizing agents and thus the efficiency of color elimination increases (30, 31). In the present study, by increasing the initial dye concentration, the removal efficiency was reduced, because with increasing color concentration, the amount of adsorbed on the adsorbent increased in the adsorbent surfaces, which prevented the adsorption on surfaces by ultrasonic waves (33). The removal efficiency was also increased by increasing the concentration, but after a period of time, active sites of the adsorbent were easily accessed, and by further increase in concentration, these active sites were occupied, and the removal efficiency decreased (29). Similar behavior was also observed and reported by Muruganandham and Maleki et al. (33, 34). The removal efficiency was higher in the early times and after a while, the slope of diagram slowed down at a specific time placed in equilibrium. At the early stages, the active sites on the surface of the adsorbent were more accessible, but after a while, these empty sites were filled by RB19 molecules (35, 36). In this study, correlation coefficients (R^2) of pseudo first-order and decolorisation of RB19 were low. According to the obtained results (Table 1 and Figures 8–10), the current study was found to be more compatible with pseudo-second-order kinetic model. Therefore, pseudo second-order kinetic model showed suitable correlation for RB19 adsorption on MPS.

5. Conclusion

The results of this study indicated that the process of US/MPS in optimum conditions (RB19 concentration of 20 mg/L, pH of 2, MPS dose 0.7 g/L and contact time of 45 min) can remove a large impact on the concentration of RB19 in water. Among the linear models, pseudo second-order model showed the highest correlation coefficient. It was also documented that the pseudo-second-order model had the best fit with the experimental data. The high correlation coefficients also showed that the applied kinetic model demonstrated good concordance with the data, as the coefficient of determination (R^2) was equal to 0.99 for process. Thus, this method can be used as a novel method for treating wastewater contaminated with dye sources.

Acknowledgments

This article is derived from the BSc. Project of Ms. L. Shikhe, and all authors are grateful to the Zabol University of Medical Sciences for the financial support of this study (Project No. 1396.13).

Conflicts of Interest

The authors declare that there are no conflicts of interest regarding the publication of this manuscript.

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