Nano World Journal

https://doi.org/10.17756/nwj.2022-108

Research Article

Open Access

Utilization of Surfactants to Augment Decolorization Process by Biosorbent

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Received: November 03, 2022
Accepted: December 24, 2022
Published: December 26, 2022

Citation: Gül UD. 2022. Utilization of Surfactants to Augment Decolorization Process by Biosorbent. *NanoWorld J* 8(4): 107-112.

Published by United Scientific Group

Abstract

The development of the textile industry significantly impacts the country's economy, but also the untreated wastewater from this industry contributes to water pollution. Unfortunately, dyes used in the textile industry are accepted as the major pollutants for the aquatic environment. The inexpensive treatment technologies for textile effluents continuously save their importance to prevent water pollution. It is aimed to investigate the potential of biosorbents obtained from pistachio shells, which were wastes of agricultural products, modified with surfactants to treat the dye-containing solutions in this study. Biological waste material was modified with surfactants to enhance biosorption and the effects of parameters such as pH, contact time, and surfactant concentration were determined. The cationic surfactant Hexadecyltrimethylammonium bromide (CTAB) modified waste pistachio shells removed 95.92% of textile dye Remazol Black B (RBB) after 360 minutes, while the unmodified shells removed a maximum of 64.76% of the same dye after 1440 minutes. Also, the biosorption isotherm and kinetic models were used to clarify the biosorption mechanism. It was concluded that surfactants can be utilized to augment the decolorization capacity of easily available, low-cost, and eco-friendly waste biosorbents for the treatment of textile dye-contaminated water.

Keywords

Biosorption, Surfactant, Textile dye, Waste

Introduction

Among other industries, textile industry is considered the most polluting resource due to the discharge volume and effluent composition [1]. It has been estimated that approximately 10 - 15% of the dye passes to the wastewater during the dying process [2] and approximately 40 - 65 L of wastewater per kg of products comes out in the post-production stage of the textile industry [3]. Every year around 280.000 tons of dyed wastewater is realised to the receiving environment [4]. RBB is an anionic reactive azo dye, which has the largest market share in the polyamide dyeing sector in Turkey [5].

The surfactants are used as wetting, dispersing, and dyeing agents in the dying process because of their solvent and dye distribution regulating properties [6]. The uniform dyeing agents reduce the affinity of the dye to the fabric by entering the race with the dye in the textile product, and the dye increases the rate of movement on the textile product properly and provides better dyeing of the fabric [7]. Surfactants used in the dyeing process are also mixed with textile wastewater as well as underground and surface waters. In addition to the negative effects of the wastewater containing dye on the receiving environment; to reach the body of kidney function, reproductive system, liver, brain, and central nervous system can make hard damage in humans [8]. Industrial wastewater, especially the textile industry, should be treated with various methods before being released into the rivers, seas, and receiving environments. Wastewater treatment methods are divided into three physical, chemical, and biological treatment methods [9]. Biological treatment is preferred because it is more economical and eco-friendly than other treatment methods [10]. Particularly adsorption is recommended as a low-cost method due to the usage of economic adsorbents obtained from biological materials [11]. This study aims to examine the utilization of the surfactant-modified waste pistachio hard shells (PHS) in the removal of textile dye Remazol Black B (RBB) from the aqueous media. The secondary goal of this study is to examine the utility of solid wastes such as pistachio hard shells in wastewater treatment technologies as inexpensive biosorbents. In the literature, the adsorption capacity of different adsorbents modified with surfactant has been determined but there is no study on surfactant-modified waste PHS. In this context, the results of this study will contribute to the literature.

Materials and Methods

Preparation of biosorbent

The ground pistachio hard shells (PHS- 30 g) were ground (using Fore Tes weighed with analytical balance (TW423L model, Shimadzu) and washed with ethanol-distilled water solution (30:70) for three times. At last, PHS was washed with pure distilled water and filtrated with Whatman paper. The filtered ground PHS was dried in the oven (EN 932-5, Nüve) at 80 ˚C for 1440 minutes and used as a biosorbent.

Pre-modification operations

In a previous study, it was shown that the micropore volume of activated carbon used as an adsorbent increased as a result of NaOH application [12]. In the same study, it was reported that the nitrate adsorption capacity of activated carbon increased as a result of NaOH-CTAB modification. Therefore, in this study, pre-modification was made using NaOH before surfactant modification. For this purpose, the 1 g of biosorbent obtained from PHS was added into 100 ml distilled water solution containing 1M NaOH and stirred for 30 minutes at 50 ˚C at 150 rpm in the shaker. Then, the biosorbent in the solution was filtered, cleaned with distilled water, and dried at room temperature.

Modification with surfactant hexadecyltrimethylamonium-bromide (CTAB)

The NaOH-treated biosorbent was incubated in a distilled water medium containing 100 ml of CTAB at room temperature for 24 hours in a shaker (150 rpm). In order to examine the effect of surfactant concentration used in modification on dye removal 5 different biosorbents were prepared listed as;

(1) 0 mM CTAB- Pistachio Hard Shell: unmodified biosorbent for control (UB),

(2) 0.1 mM CTAB- Pistachio Hard Shell: modified biosorbent with 0.1 mM surfactant (0.1 MB),

(3) 0.5 mM CTAB- Pistachio Hard Shell: modified biosorbent with 0.5 mM surfactant (0.5 MB),

(4) 1 mM CTAB- Pistachio Hard Shell: modified biosorbent with 1 mM surfactant (1 MB),

(5) 2mM CTAB- Pistachio Hard Shell: modified biosorbent with 2 mM surfactant (2 MB).

Biosorbents (1 g) were added into 250 ml of distilled water containing surfactant CTAB with different concentrations (0.1, 0.5, 1, and 2 mM). The experiments were carried out at 150 rpm agitation rate for 24 hours at 25 ˚C. The adsorbent is then filtered and allowed to dry for 24 hours at 50 ˚ C in the oven.

Preparation of surfactant and dye solutions

The cationic surfactant called Hegzadecyltrimethylammonium bromide (CTAB), which was purchased from Sigma, was used in this study (Figure 1a). The stock surfactant solutions were prepared in distilled water with 1g/L surfactant and the desired amounts of solutions used in aqueous media for modification.

The anionic dye called Remazol Black B (Figure 1b) was taken from the Textile Factory (Bilecik) in powder form. To prepare the stock dye solution the powdered dye dissolved in distilled water with a concentration of 2% (w/v). Experimental arrangements were prepared by adding the determined amounts of this stock solution into the medium. The properties of the surfactant and dye used in this study were given in table 1.

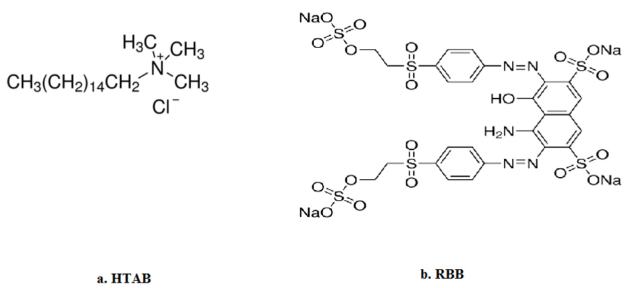

Figure 1: Chemical structure of **(a)** Hegzadecyltrimethylammonium bromide (HTAB) surfactant and **(b)** Remazol Black B (RBB) dye.

Table 1: The properties of surfactant and dye used in the study.

Name	Chemical Formula	Class	Molecular Weight
Hegzadecyltrimethylammoniumbromide (HTAB)	C19H42BrN	Cationic	364.46
Remazol Black B (RBB)	C26H21N5Na4O19S6	Anionic	991.82

Biosorption experiments

The biosorption experiments were investigated at batch scale level in 100 ml Erlenmeyer flasks having 50 ml dye containing distilled water. The effect of pH experiments was done at different pH levels such as pH 1, 3, and 5 using the H12211 pH meter, Hanna Ins. The effect of surfactant concentration for biosorbent modification was examined at varied surfactant concentrations such as 0.1, 0.5, 1, and 2 mM. The influence of contact time was examined at different time periods from 0 to 2880 minutes. The effect of temperature was also examined at 25 and 50 ºC. All experiments were done in triplicate and the average values were used in the results.

Dye removal analysis

After the incubation period, 2 ml samples were taken daily and centrifuged at 10000 rpm for 15 minutes. The supernatants were analyzed at a 600 nm wavelength spectrophotometer. During the spectrophotometric analysis, the flasks containing distilled water were used as controls. AgileSpec spectrophotometer and Daihan Scientific model centrifuge were used in experiments. The dye biosorption and dye uptake capacity of the adsorbent were calculated from Eq (1): Biosorption (%) = $(C_o - C_f) / C_o \times 100$ and Eq. (2): $q_m = (C_o - C_f) / X_m$, respectively. In equation 1 and 2; q_m, X_m, C_o, and C_f have expressed the maximum specific dye uptake (mg/g), the maximum dried biosorbent (g/L), the initial dye concentration (mg/L), and the final dye concentration (mg/L), respectively. All the experiments were carried out in triplicate. SPSS 17 and the ANOVA (one-way) are used for the statistical analysis of data obtained from experiments.

Results and Discussion

The effects of pH, surfactant concentration, temperature, and contact time on RBB biosorption capacity of surfactant-modified and unmodified waste PHS were examined in this study. Chaleshtori et al. [13] examined the effect of different pH values (pH 2-12) on the Acid Red 18 removal of the biosorbent obtained from the almond shell and determined that the best removal was at pH 2, which is the lowest pH value used. Mazarji et al. [12] investigated the pH effect (pH 3-11) of surfactant-modified activated carbon granules on nitrate removal and showed that the highest removal was at pH 5. Alhujaily et al. [8] studied the effect of pH values (pH 3-11) on the removal of anionic dyes Direct red 5B (DR5B), Direct blue 71 (DB71), and Reactive black (RB5) dyes by waste mushroom biosorbent modified with a cationic surfactant. They reported that the best dye removal was observed at pH ranging from pH 3 to 5. The experimental setup was designed in line with the data in the literature, and pH 1, 3 and 5 values were used in the experiments to determine the pH effect. In order to examine the effect of pH, all biosorbents (unmodified and modified PHS) were used in 100 ml flasks with 50 ml dye-containing solutions at variety of pH values as 1, 3, and 5 incubated for 24 h on the shaker (100 rpm) at 25 ˚C. As seen in figure 2, unmodified biosorbent (UB) performed a maximum dye biosorption rate at pH 5. The dye biosorption rate of modified biosorbents (MB) was increased by increasing the surfactant concentration used for biosorbent modification. Also, it was investigated that the biosorption rates decreased with the increasing pH value from 1 to 5. The modified biosorbents (MB) showed maximum dye removal at pH 1 as 34.33% and 33.02% with 1 and 2 mM CTAB application for modification, respectively. The UB decolorized 64.76% of the dye at pH 5. The pH of the environment significantly affected the biosorption process due to the changes in functional groups on the biosorbent surface.

A previous study demonstrated that surfactant-modified zeolite performed higher adsorption capacity than unmodified zeolite at low pH values [14]. Similarly, with this study, Gül and Dönmez [15] reported that the fungal surface charges were changed in the presence of a cationic surfactant and the maximum Remazol Blue (anionic dye) removal occurred at low pH values. Recently, Jimenez-Castaneda and Medina [16] reported that the modification of zeolite and clays with the cationic surfactant having long alkyl chains changed the surface charge from negative to positive by using cationic exchange and hydrophobic interactions. The cationic surfactant CTAB has a long alkyl chain (Figure 1). Also, Lin et al. [17] explained the sorption mechanism of arsenate on kaolinite modified with HTAB via the anion exchange mechanism dominantly. Chutia *et al.* [18] showed that after the cationic surfactant application of zeolite the new surface charge of zeolite induced the interaction with counter ions, therefore the sorption capacity of ionic pollutants was enhanced by the surfactant modification. Chutia *et al.* [18] showed that after the cationic surfactant application of zeolite, the new surface charge of zeolite could interact with counter ions, and the sorption capacity of ionic pollutants was enhanced by the surfactant modification. According to the results of pH experiments in this study, the dye removal behavior of the biosorbent was affected by the pH of the environment. The UB removed maximum removal at high pH values, but the MBs showed maximum decolorization at low pH values. While figure 2 examined pH 1 was the optimal pH for the biosorption of dye. For the surfactant concentration effect, it was observed that the raising surfactant concentration significantly affect the biosorption rate (Figure 2) and in the experiment when the surfactant concentration reached up to 1 mM, the biosorption rate increased.

The effects of temperature on the biosorption properties of biosorbents were tested. The dye removal was tested at 50 °C in the solutions with pH 1 and 5, which were optimal pH values for decolorization by MB and UB, respectively. The effect of low (25 °C) and high (50 °C) temperature values on biosorption rates were given in figures 2 and 3, respectively. In the optimal pH value (pH 5) for UB removing the dye, UB removed 64.76% and 18.39% of dye ions from solutions at 25 and 50 ˚C, respectively. The MBs showed maximum removal rates at 50 °C and the dye biosorption rates were increased by increasing the surfactant concentration for modification of biosorbents (Figure 3). The decolorization rate was 94.28% at 50 ˚C by modified with 2 mM CTAB, maximally (Figure 3). Recently, Melo et al. [19] indicated that increasing temperature caused decreasing in dye removal. Also, Zhao *et al.* [20] reported that anionic dye adsorption onto cationic surfactant-modified peanut husk was decreased by increasing temperature. On the other hand, Alhujaily et al. [8] reported that the anionic dye removal of cationic surfactant-modified

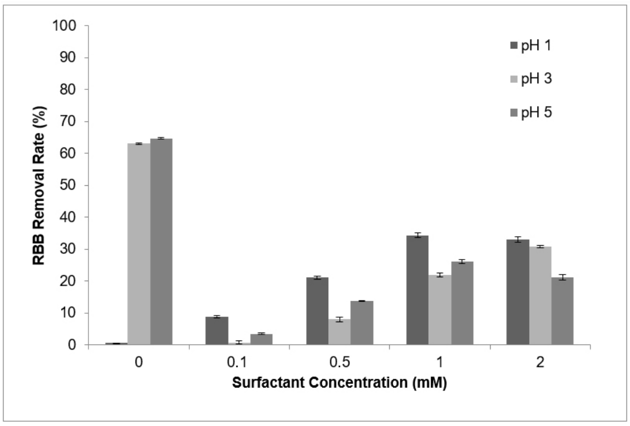

Figure 2: The effect of pH on dye biosorption with unmodified (0 mM) and modified with different surfactant concentrations (0.1, 0.5, 1, and 2 Mm), (p = .05, df = 2). Error bars represent the standard error of the mean.

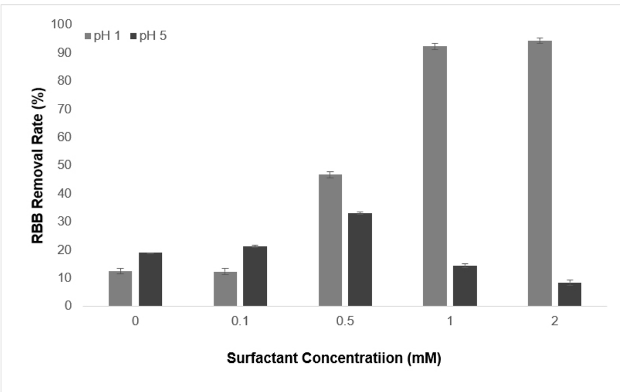

Figure 3: The effect of high temperature (50 ^{o}C) on dye biosorption with un-modified (0 mM) and modified with different surfactant concentrations (0.1, 0.5, 1 and 2 mM) at different pH (1 and 5) values, (p = .05, df = 2). Error bars represent the standard error of the mean.

mushroom was increased by the increase of the temperature from 20 to 50 ^{o}C due to the increment of dye molecules diffusion rate to adsorb on the pores of adsorbent. In addition to this, the increase in temperature may lead to the fluidity of dye molecules and encourage the formation of interactions between dye molecules and the surface of adsorbents by providing the necessary energy [8]. Also, Liu et al. [21] indicated that increasing temperature caused the expansion of the adsorbent internal structure and enhance the dye molecules to pass through. Similarly, in the results of this study, the decolorization rate was increased from 33.02% to 94.28% at the increasing temperature from 25 to 50 ^{o}C by modifying with 2 mM CTAB.

The effect of contact time experiments was carried out with optimal conditions as pH 1 and MB with 2 mM HTAB at 50 ^{O}C. Figure 4 shows that the maximum removal rate occurred at 360 minutes, and after 360 minutes there was no significant change in the removal rate. The equilibrium time was found as 360 minutes with a high removal efficiency.

The commonly used isotherm models as Freundlich, Langmuir, and Dubinin-Radushkevich (D-R) were calculated in the optimal conditions as pH 1 and MB with 2 mM CTAB at 50 ^{o}C. The isotherm equations were calculated from Equations (3, 4, 5)

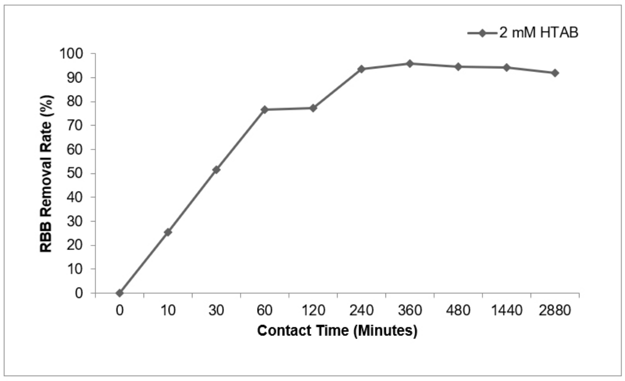

Figure 4: The effect of contact time on dye biosorption with 2 mM surfactant modified biosorbent at pH 1 and 50 ^{O}C, (p = .05, df = 2).

Eq. (3): *Langmuir Isotherm Equation*: $q = X_L K_L C_e / 1 + K_L C_e$

Eq. (4): *Freundlich Isotherm Equation*: $q = K_F C_e^{\,n}$

Eq. (5): *D-R Isotherm Equation:* $qe = q_{DR}\, e^{-K_{DR}\,\varepsilon 2}$

In these equations, X_L, K_L, K_F, X_{DR}, and C_e express the maximum adsorption capacity, the parameter for Langmuir isotherm, the equilibrium concentration (mol/L), the Freundlich constant, and the adsorbate concentration at equilibrium, respectively. And ε, K_{DR}, R, and T show the Polanyi potential coefficient ($mol^2 KJ^2$), the activity, the ideal gas constant (8.314 J/molK), and the absolute temperature (K), respectively. The Polanyi potential (ε) is expressed by the following Eq. 6: ε= $RTln(1+ 1/C_e)$. The adsorption energy (E) is expressed by the following Eq. 7: $E_{DR} = (2K_{DR})^{-0.5}$ If the adsorption energy is 8<E<16 kJ mol^{-1}, the adsorption is physically controlled and E<8 $kJmol^{-1}$ indicates that the adsorption proceeds physically (Dubinin et al. 1947; Ho 2006) [22, 23].

The kinetic models of pseudo-first-order and pseudo-second-order kinetic model equations were used to examine the RBB dye biosorption mechanism. Kinetic data were calculated with the equations: Eq. (8) $q_t = q_e[1- e^{-k_1 t}]$ and Eq. (9): $q_t = t / [1/k_2 Q_e^{2}] + [t/q_e]$ for Pseudo-first-order and Pseudo-second-order kinetic models, respectively.

In these equations, q_t (mol/kg), q_e (mol/kg) are the dye absorbed amount at time t (min) and the absorbed amount at equilibrium, respectively. k_1 and k_2 are the rate constant of the pseudo-first-order (/min) and the pseudo-second-order (/mol kg/min) models, respectively.

The isotherm and kinetic models were calculated from the data obtained from experiments carried out at optimal conditions. The correlation values of Langmuir and Freundlich isotherms were found as 0.999 and 0.827 for dye biosorption on biosorbent, respectively (Table 2). The biosorption of dye on MB was compatible with the Langmuir model because of the highest correlation value. Similarly, Alhujaily et al. [8] indicated that the anionic textile dye biosorption by a biosorbent obtained from mushroom waste modified with a cationic surfactant was fitted with the Langmuir model. The biosorption free energy calculated from the D-R isotherm was calculated as E_{DR}: 0.58 kJ/mol, which indicated that the dye biosorption process by the biosorbent was physically performed.

Table 2: Langmuir (L), Freundlich (F), and D-R isotherm parameters.

	Parameter	Value	R^2
Langmuir	X_L (mg/g)	61.1	0.999
	K_L (L/mg)	1.15	
Freundlich	K_F	2.05	0.827
	β	0.222	
D-R	X_{DR}	4.39	0.993
	$-K_{DR}x10^6$	1.49	
	E_{DR}/kJ/mol	0.58	

The biosorption kinetic calculations are given in table 3. The correlation value of the pseudo-second-order model was the highest (0.998), so the biosorption of dye on the MB was suited to the pseudo-second-order kinetic model. Similarly, Chaleshtori et al. [13] showed that the Acid Red 18 dye adsorption on activated charcoal originating from the almond shell was compatible with the pseudo-second-order kinetic model. Recently, it was shown that the dye biosorption process by surfactant-coated mushroom waste was fitted with the pseudo-second-order kinetic model [8].

Tablo 3: Pseudo-first-order and pseudo-second-order kinetic parameters.

Kinetic model	Parameter	Value	R^2
Pseudo first order	q_t/mgg	89.1	0.962
	q_e/mgg	73.8	
	k_1 x10^3/min	30.0	
	Hx10^3/mg/g/min	2190	
Pseudo second order	q_t/mg/g	89.1	0.998
	q_e/mg g^{-11}	85.8	
	k_2 x10^3/mgg min^{-1}	0.39	
	Hx10^3/mg/g/min	33.0	

Conclusion

In this study, the usability of the waste pistachio hard shells in dye removal was investigated and the waste shells were modified by the cationic surfactant to promote dye biosorption. Research findings showed that the biosorption efficiency of modified shells is higher than that of unmodified ones. According to the results of this research, the surfactant-modified waste pistachio hard shells removed 95.92% of reactive dye after 360 minutes, maximally. The results of this study supported that the solid wastes of pistachio shells can be used as low-cost biosorbents for textile dye-contaminated water.

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