

Biosorption of remazol orange RR from aqueous solution: kinetic, equilibrium and thermodynamic studies

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ABSTRACT

Remazol Orange RR (RORR) is one of the most frequently used textile dye which causes environmental pollution. In the present study, parameters affecting the RORR biosorption with dead *Saccharomyces cerevisiae* yeast were investigated. The maximum %RORR removal was found to be 84.9% at 2 g/L biosorbent dosage, 200 mg/L initial dye concentration, pH 3.0, and 25°C. RORR biosorption with *S. cerevisiae* was expalined by pseudo-second-order kinetics model and Langmuir adsorption isotherm with correlation coefficient (R^2) of 0.99 and 0.87, respectively. The enthalpy change (ΔH), entropy change (ΔS) and activation energy (E_a) were determined as 35.9 kJ/mol, 88.1 kJ/mol K and 3.36 kJ/mol, respectively. In addition, Gibbs free energy change was calculated as -10.04, -8.03 and -8.33 kJ/mol for 298, 308 and 318 K, respectively. It is concluded that the biosorption of RORR by *S. cerevisiae* occurs mainly by physical adsorption through a spontaneous process. The high dye removal and biosorbent capacity indicated that *S. cerevisiae* is a promising biosorbent for dye removal.

Keywords: Remazol orange; Biosorption; Saccharomyces cerevisiae; Adsorption isotherms; Kinetics; Thermodynamics

1. Introduction

Synthetic dyes are one of the major causes of water pollution. The annual amount of dye production is about 7×10^5 tons, and 10%–15% is released to waterbodies mainly by textile industry [1,2]. Besides, enormous amount of water usage in textile industry results with large quantity of waste water [3,4]. It is reported that dyes exhibit mutagenic, toxic [5] and carcinogenic properties for water creatures [6] and hinder their biological activities [7] as well as disturb the aesthetic appearance [8].

Reactive dyes generally consist of azo-based chromophore and reactive groups by which they can form covalent bond with functional groups of fiber [9]. Due to its harmony with cotton in terms of dyeing properties, reactive dyes are used more widely compared with other types and owing to their high solubility in water reactive dyes can easily pass through conventional water treatment systems. This situation clearly demonstrates the need for alternative methods for the removal of textile dyes from water and decolourization of reactive dyes is a crucial topic for a better environment [10]. Several methods namely electrochemical oxidation, reverse osmosis, coagulation, flocculation, flotation, chemical oxidation, ozonation, precipitation, sedimentation, ultrafiltration and color irradiation or combined usage of abovementioned methods were reported to reduce the dye content of wastewater [11,12] but most of these methods suffer from being expensive [13], requiring complex application protocols [14] and high energy consumption [15]. Among these methods, adsorption is one of the most popular techniques in terms of its advantages such as showing high efficiency [16], involving simple operation conditions [17], and being reasonably priced [18] while selectivity and affinity of the adsorbents to dyes constitutes the major consideration of this technique.

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Biosorption is a type of adsorption technique in which dead or alive biological materials are employed as the adsorbents [19]. Use of dead biomass in biosorption applications provides additional advantages, such as no necessity of aliment, reusability and resistance to wastewater toxicity [20]. Reactive dye biosorption onto several dead or living biosorbents such as Aspergillus versicolor [21], banana peel powder [22], barberry powder [23], Lentinus polychrous Lev. [24], Citrus sinensis [25], Corynebacterium glutamicum [26], sunflower seed hull [27], Metapenaeus monoceros shells [28], Pistacia vera [29], Trametes subectypus [30], peanut shell [31], eggshell [32], Nizamuddinia zanardinii [33] and Saccharomyces cerevisiae [34] have been researched recently. To our knowledge, removal of Remazol Orange RR (RORR) that is a reactive and azo dye, with S. cerevisiae has not yet been investigated. In this respect, the present study introduces a novelty to the related literature. It is known that the surface of S. cerevisiae contains carboxyl, phosphonate and amine groups [35]. Among these, carboxyl and phosphonate groups do not show affinity to anionic dyes due to electrostatic repulsion while amine groups are expected to interact with anionic dyes due to their tendency to protonate [36]. In this study, biosorption of Remazol Orange RR (RORR) onto S. cerevisiae and optimization of the operating parameters such as pH, initial dye concentration and biosorbent dosage were invesitigated. Biosorption mechanisms were explained by applying kinetic and equilibrium isotherms to the obtained data. Thermodynamic parameters of the biosorption process namely free energy (ΔG°), enthalpy (ΔH°), entropy (ΔS°) and activation energy (E_{a}) were calculated.

2. Materials and methods

2.1. Chemicals

All chemicals used in this study such as yeast extract, peptone, glucose, malt extract, potassium phosphate monobasic (KH_2PO_4) , ammonium sulfate $((NH_4)_2SO_4)$, magnesium sulfate heptahydrate $(MgSO_4 \cdot 7H_2O)$, calcium chloride $(CaCl_2)$, NaOH and H_2SO_4 were of analytical grade and purchased from Sigma-Aldrich (Germany). Remazol Orange RR (RORR) was obtanined from Dyestar (Germany). *S. cerevisiae* as lyophilized stock culture (code number: 251 TP (3–2)) was provided from Refik Saydam Hifzihsihha Presidency (Ankara, Turkey).

2.2. Instruments

Dye concentrations were determined by using UV/VIS Spectroscopy T 80+ (PG Instruments Ltd., UK). Sterilizer and incubator were used for yeast growth. Biosorpsion experiments were performed by using a shaking water bath.

2.3. Preparation of biosorbent and adsorbate

Agar slant, pre-activation and growth media were prepared to obtain *S. cerevisiae* yeast. The yeast was inoculated to agar slant which consists of 5 g yeast extract, 20 g peptone, 20 g glucose, 5 g malt extract and 20 g agar per liter of deionized water. Then, it was placed into the incubator at 30° C for 48 h. The yeast was collected from the agar surface and mixed with the pre-activation medium and incubated at 30° C for 48 h.The activated yeast was transferred to the growth medium which contains 50 g glucose, 12 g KH₂PO₄, 5 g (NH₄)₂SO₄, 1 g MgSO₄·7H₂O, 0.5 g CaCl₂ and 5 g yeast extract per liter. Then, it was mixed in a magnetic stirrer at 30° C for 24 h. Finally, the solution was centrifuged, sterilized, filtered and dried. The biosorbent used in this study was sieved (>1.5 mm) for biosorption experiments.

2.4. Biosorption studies

Dye concentrations were quantifed at wavelength of 492 nm where the highest absorbance was detected. The pH values of dye solutions were adjusted by adding 0.1 M HCl or 0.1 M NaOH solution dropwise. After, the biosorbent was added into this solution and placed into shaking water bath at 140 rpm at certain temperature. The percentage of RORR removal and biosorbent capacity were calculated from Eqs. (1) and (2):

$$R(\%) = \frac{C_0 - C_f}{C_0} \times 100$$
 (1)

$$q_e = \frac{V(C_0 - C_f)}{m} \tag{2}$$

where C_0 and C_f represent the initial and final dye concentration, *m* is the quantity of biosorbent and q_e is the biosorbent capacity at equilibrium.

The effect of pH, initial dye concentration and biosorbent dosage on RORR biosorption was investigated in the range of pH 2.0–5.0, 50–400 mg/L of initial dye concentration and 0.5–2 g/L of biosorbent dosage.

2.5. Kinetic and equilibrium models

The biosorption equilibrium experiments were performed in the range of 25°C–40°C and 50–400 mg/L initial dye concentration. The experimental data were evaluated by using Langmuir, Freundlich and Temkin isotherms and their linear forms are given in Eqs. (3)–(5), respectively [37]:

$$\frac{C_e}{q_e} = \frac{1}{bq_s} + \frac{C_e}{q_s}$$
(3)

$$\log q_e = \log K_f + \frac{1}{n} \log C_e \tag{4}$$

$$q_e = B \ln A_T + B \ln C_e \tag{5}$$

where C_e is the concentration of adsorbate at equilibrium, b is the Langmuir isotherm constant, q_s is the maximum amount of substance adsorbed per unit mass of adsorbent, K_f is the Freundlich isotherm constant, n is a constant associated with adsorption intensity, B is the constant about the heat sorption and, A_T is the equilibrium binding constant of Temkin isotherm [38–42].

RORR biosorption onto *S. cerevisiae* as a function of time was evaluated by pseudo-first-order and pseudo-second-order kinetic models as well as intraparticle diffusion and liquid film diffusion models which were formulized with Eqs. (6)–(9), respectively [43]:

$$\log(q_{e} - q) = \log(q_{e}) - \frac{k_{1}}{2,303}t$$
(6)

$$\frac{t}{q} = \frac{1}{k_2 q_e^2} + \frac{1}{q_e} t$$
(7)

$$q_t = k_t t^{\frac{1}{2}} + c \tag{8}$$

$$\ln\left(1 - \frac{q_t}{q_e}\right) = -R^t t \tag{9}$$

where k_1 , k_2 , k_i and R^i represent the rate constant of pseudo-first-order, pseudo-second-order, Weber–Morris diffusion constant, and liquid film diffusion constant, respectively [44–46].

2.6. Adsorption thermodynamics

The thermodynamic measurements were conducted at 25°C, 35°C and 45°C to determine ΔG° , ΔH° , ΔS° and E_a values of the biosorption process. Eqs. (10)–(12) were used for this purpose.

$$\Delta G = \Delta H - T \Delta S \tag{10}$$

$$\ln b = \frac{-\Delta H}{RT} + \frac{\Delta S}{R} \tag{11}$$

$$\ln k_2 = \ln k_0 - \frac{E_a}{RT} \tag{12}$$

where *R* is the universal gas constant (8.314 J/mol/K), *b* is Langmuir isotherm constant, k_2 and k_0 are pseudo-second-order kinetic model rate constants (g/mg min), respectively [47].

3. Results and discussion

3.1. Effect of contact time

Equilibrium duration of the biosorption was determined at constant parameters of 25°C, 100 mg/L dye concentration, 2 g/L biosorbent dosage, and 140 rpm shaking rate. Fig. 1 shows the change in RORR concentration as a function of time. It is observed from Fig. 1 that RORR concentration reduced sharply from 90 to 70 mg/L in 50 min, and became almost stable in 150 min. So, the experiment duration was determined as 150 min.

3.2. Effect of pH on biosorption

Favorable removal of reactive dyes at acid pH values has been reported in the literature [28]. Based on this information, the effect of dye solution pH on dye removal and biosorbent capacity were investigated between pH 2.0 and 5.0 while other parameters were kept constant; 50 mg/L initial dye concentration, 1 g/L biosorbent loading, 25°C and 140 rpm shaking rate. Fig. 2 presents RORR removal and biosorbent capacity at different pH values. It was observed that RORR concentration increased from 4.58 to 42.8 mg/L with the increasing pH from 2.0 to 5.0. The highest and



Fig. 1. Effect of contact time on RORR concentration.



Fig. 2. (a) RORR removal and biosorbent capacity at varying pH and (b) zero charge point of the biosorbent.

lowest RORR removal and biosorbent capacity values were obtained at pH 2.0 as 91.2% and 47.1 mg/g and at pH 5.0 as 17.3% and 8.93 mg/g, respectively. High RORR removal and biosorbent capacity at acidic pH values could be attributed to the electrostatic interaction between dye anions and positively charged cell surface [48-50]. This interpretation was also supported by the plot shown in Fig. 2b. The zero charge point (pH_{pzc}) of the S. cerevisiae biosorbent was determined to be 8.13 and it is known that the surface of biosorbent is positively charged at pH values below pH_{pzc} value [51]. It is clear from Fig. 2 that the percentage removal and biosorbent capacity at pH 2.0 are very close to the values obtained at pH 3.0. From this point of view, dye solution of pH 2.0 was considered to be more disadvantageous in terms of ease of application and economical feasibility. Therefore, optimum pH value was determined to be 3.0 for RORR biosorption on S. cerevisiae.

3.3. Effect of initial dye concentration on biosorption

The experiments were conducted in the range of 50–400 mg/L initial dye concentration while the other parameters were kept constant at 1 g/L biosorbent concentration, pH 3.0, 25°C and 140 rpm in order to observe the effect of initial dye concentration only. From Fig. 3, it can be clearly seen that percentage removal of RORR decreased from 80.3% to 28.1% and biosorption capacity increased from 54.7 to 100.7 mg/g with the increasing initial dye concentration from 50 to 400 mg/L. The highest RORR removal and biosorbent capacity values were found as 80.3% and 100.7 mg/g. The decreasing RORR removal with the increasing initial dye concentration of the sorption capacity is sorption sites on *S. cerevisiae* [52,53].

3.4. Effect of biosorbent dosage on biosorption

The experiments were conducted in the range of 0.5–2 g/L biosorbent dosage while the other parameters were kept constant at 200 mg/L initial dye concentration, pH 3.0 and 25°C. The effect of biosorbent dosage on the biosorption



Fig. 3. RORR removal and biosorbent capacity for varying initial dye concentration.

capacity and percentage removal is shown in Fig. 4 and an increament was observed in the percentage removal values from 22.5% to 84.9% with the increasing biosorbent dosage. The correlation between these two parameters was attributed to the increased number of the existing adsorption sites [54]. When the biosorbent dosage was increased from 0.5 to 1 g/L, the biosorbent capacity increased, while it was observed that the capacity values for dosages greater than 1 g/L decreased sharply. The decrease in biosorption capacity with the increasing biosorbent concentration over 1-2 g/L could be explained by the fact that increasing amount of adsorbent may increase the number of adsorption sites [13] and becomes more than required for the corresponding dye concentration while these sites remain unsaturated during adsorption [55] and the effective surface area decreases due to the formation of biosorbent aggregates at high biomass concentrations [56,57].

3.5. Kinetic studies

The pseudo-first-order kinetic model, pseudo-secondorder kinetic model, intra-particle diffusion model and liquid film diffusion model graphs at constant parameters of 100 mg/L initial dye concentration, pH 3.0, 1 g/L biosorbent concentration and 35°C are shown in Fig. 5. Kinetic and diffusion model parameters are given in Table 1.

In Table 1, it can be seen that the R^2 values for the pseudofirst and second-order kinetic models are 0.85 and 0.99, respectively. According to these values, it was believed that the pseudo-second-order kinetic model was more suitable for the biosorption of RORR with *S. cerevisiae* due to the high correlation coefficient. The main assumption of the pseudosecond-order kinetic model is that the binding rate of dye ions on the biosorbent is proportional to the square of number of free adsorption sites [58]. The R^2 values of employed intra-particle diffusion model and the liquid film diffusion model for determining the diffusion mechanism of the RORR biosorption were found to be 0.91 and 0.85, respectively. From Table 1, it was concluded that the intra-particle



Fig. 4. RORR removal and biosorbent capacity for different biosorbent concentration.



Fig. 5. Kinetic and diffusion models for the biosorption: (a) pseudo-first-order kinetic model, (b) pseudo-second-order kinetic model, (c) Weber–Morris intraparticle diffusion model, and (d) liquid film diffusion model.

Table 1 Kinetic and diffusion models parameters at 35°C

Model	Parameter (unit)	Experimental value	Model value
Pseudo-first-order	$q_1(mg/g)$	70.62	52.92
	$k_1(\min^{-1})$	-	0.0022
	R^2	-	0.85
Pseudo-second-order	$q_2(\mathrm{mg/g})$	70.62	68.93
	k_2 (g/mg/min)	-	0.0036
	R^2	-	0.99
Weber–Morris	$k_p (mg/g/min^{1/2})$	_	8.84
	R^2	-	0.91
Liquid film diffusion	R^{1} (min ⁻¹)	0.022	0.022
	R^2	-	0.85

diffusion model constitutes the major part of this biosorption process. According to the intra-particle diffusion model, adsorbate binding on the adsorbent varies in proportion to the square root of the contact time. The graph of q_i - $t^{1/2}$ is expected to cross through origin when the rate-limiting step

of biosorption process is intra-particle diffusion model. But, often this is not the case and the adsorption kinetics is controlled by both liquid film diffusion and intra-particle diffusion models [59]. It can be seen from Fig. 5c that q_t vs. $t^{1/2}$ plot does not cross through origin. Hence, it could be concluded

that the diffusion process cannot be explained only by the intra-particle diffusion model and liquid film diffusion also plays a part in this process.

3.6. Equilibrium studies

The Langmuir, Freundlich and Temkin isotherms obtained for RORR biosorption at constant parameters of 35°C, pH 3.0 and 1 g/L biosorbent concentration are shown in Fig. 6 and the parameters of the model values for these isotherms are given in Table 2. From Table 2, it is seen that the highest compatibility was obtained with the Langmuir model (0.87). R^2 value of the biosorption was found to be 0.99 at 50°C indicating that *R*² values increased with the increasing temperature for the RORR biosorption. Therefore, it was concluded that RORR biosorption onto S. cerevisiae is better explained by the Langmuir model. The Langmuir model indicates the constant adsorption energy and no interaction between the adsorbate and adsorbent molecules [60]. In addition, occurrence of single layer adsorption and presence of limited number of bonding sites are proposed by this model [61]. It is stated in the literature that the processes described by the pseudo-second-order kinetic model are compatible with the Langmuir model [61]. Herein,

RORR biosorption is consistent with the literature in terms of good fitted data both with pseudo-second-order kinetic model and Langmuir isotherm.

3.7. Thermodynamic studies

Thermodynamic parameters of the RORR biosorption, such as ΔG° , ΔS° , ΔH° and E_{a} were determined by Eqs. (13) and (14). The thermodynamic parameters, which are summarized in Table 3, calculated with the aid of Figs. 7 and 8. As can be seen from Table 3, ΔH° and ΔS° values for the RORR biosorption were found to be 35.9 kJ/mol and 88.1 J/ mol K, respectively, while the ΔG° values for 298, 308 and 318 K were determined as -10.04, -8.03 and -8.33 kJ/mol, respectively. A negative ΔG° value indicates that the RORR biosorption was a spontaneous process and the biosorption was suitable thermodynamically [62]. The positive ΔH° of RORR biosorption (35.9 kJ/mol) indicates that the biosorption was endothermic [63]. In addition, according to the literature, ΔH° values in the range of 4–40 kJ/mol display the physical adsorption [64]. In this line, it is considered that the RORR biosorption onto S. cerevisiae process could be explained by the physical adsorption. The positive ΔS° value for RORR biosorption shows that there is an affinity between the



Fig. 6. Adsorption isotherms of RORR biosorption: (a) Langmuir, (b) Freundlich, and (c) Temkin.

Table 2 Model parameters of the Langmuir, Freundlich and Temkin isotherms

Model	Parameter (unit)	Experimental value	Model value
Langmuir	$q_{\rm m} ({\rm mg/g})$	40.92	34.06
	K_{L} (L/mg)	-	0.017
	R^2	-	0.87
Freundlich	$q_{\rm m}$	40.92	47.31
	$K_{\rm F}$ (L/mg)	-	92.59
	N	-	2.99
	R^2	-	0.81
Temkin	$q_{\rm m}$	40.92	46.49
	A_t	-	-0.20
	В	-	-5.41
	<i>R</i> ²	-	0.78

Table 3

Thermodynamics parameters of the RORR biosorption onto *S. cerevisiae*

Temperature (K)	ΔG (kJ/mol)	∆H (kJ/mol)	ΔS (J/mol/K)	E _a (kJ/mol)
298	-10.04		. ,	
308	-8.03	35.9	88.1	3.36
318	-8.33			

adsorbent and adsorbate, and that the solid/solution interface has increased randomness during the adsorption [65].

The correlation coefficient was found to be 0.998 in Fig. 7 and E_a of RORR biosorption was determined as 3.36 kJ/mol. It was concluded that RORR biosorption onto *S. cerevisiae* was an endothermic process and the biosorption can be explained by the physical adsorption owing to the positive activation energy and low E_a value [66–68].

Table 4 summarizes the biosorbents used for the removal of various textile dyes in the literature. As seen from Table 4,



Fig. 7. Activation energy of the RORR biosorption onto *S. cerevisiae.*



Fig. 8. Van't Hoff plot of RORR biosorption onto S. cerevisiae.

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Compariso	on of litera	ature about re	moval of textile dye

Biosorbent	Dye	% Removal	Biosorbent	Reference
			capacity (mg/g)	
<i>S. cerevisiae</i> -MnO ₂ Composite	Malachite Green	ca. 90	86.7	[69]
Oyster Shells	Acid Green 25	95.4	33.3	[70]
S. cerevisiae	Basic Blue 41	94.0	23.5	[71]
Nostoc linckia	Reactive Red 198	94.0	93.5	[72]
Orange peels	Reactive Red	89.4	n.a.	[73]
Chitosan-gelatin-graphene composite	Orange II	84.3	72.2	[74]
Nitrogen-doped nanoporous carbon	Methyl Orange	98.5	222.2	[75]
ZIF-8	Acid Orange 7	80.0	80.4	[76]
S. cerevisiae	RORR	84.9	80.3	In this study

S. cerevisiae reported in this study has lower percentage removal value than those of other studies, but the biosorption capacity was higher than or close to these biosorbents. This low percentage removal value could be attributed to the use of less amount of biosorbents compared with other studies. These results show that *S. cerevisiae* is a promising biosorbent for the removal of reactive dyes.

4. Conclusion

In this study, the effects of contact time, pH, initial dye concentration and biosorbent dosage on RORR removal and biosorption capacity were investigated. Langmuir equilibrium model was found to be more suitable for RORR biosorption compared with Freundlich and Temkin isotherms. The biosorption obeys the pseudo-second-order kinetic model with R^2 of 0.99. The biosorption heat of this process is calculated as 35.9 kJ/mol and ΔG° values at 25°C, 35°C and 45°C are found to be –10.04, –8.03 and –8.33 kJ/mol, respectively. ΔS° and E_a is determined as 88.1 J/mol K and 3.36 kJ/mol. In this direction, the biosorption process take place through physical adsorption, and it is a spontaneous process.

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