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Application of response surface methodology for optimization of Remazol Brilliant Blue R removal onto a novel polymeric adsorbent

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ABSTRACT

The poly[2-hydroxy-3-(1-naphthyloxy)propyl] methacrylate [poly(NOPMA)] was used as a novel adsorbent for the removal of Remazol Brilliant Blue R (RBBR) known as a reactive dye. Physico-chemical properties of polymer were analyzed by FT-IR and SEM before and after dye adsorption. The Langmuir and Freundlich isotherms were used to explain the adsorption equilibrium, and when it was compared with each other it was seen that the adsorption process was well explained by the Langmuir isotherm model and showing a monolayer adsorption capacity for RBBR of 238.10 mg/g. Kinetic models known as pseudo-first-order, pseudo-second-order and intra-particle diffusion model were utilized to analyze the mechanism of adsorption, and results indicated that was an adsorption kinetic suitable with the pseudo-second-order. The thermodynamic parameters (ΔH^o, ΔS^o, ΔG^o) indicated that the adsorption process was spontaneous and endothermic. To optimize the study conditions, the effects of the concentration of RBBR, adsorbent amount and contact time on the adsorption were investigated by response surface methodology (RSM). According to RSM results, maximum RBBR removal was reached at the optimum initial dye concentration of 60.85 mg/L, adsorbent dosage of 0.04 g/50 mL and contact time of 59.91 min.

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1. Introduction

Remazol Brilliant Blue R (RBBR) is a water-soluble dye that has a good hydrophilic/hydrophobic balance with reactive β-sulfatoethylsulfonyl groups [1,2]. RBBR was widely used in the textile industry due to its favorable characteristics such as low energy consumption, simple application techniques and not easily biodegradable [3,4]. Also it is an anthracene derivative and is used to remove the organic pollutants in wastewater [5]. The development of the industry over the world has caused significant amounts of organic pollutants to interfere with nature. When these organic substances are thrown into the water, they may damage ecological balance and living organisms by the influence of the chemical groups in their structure. Scientists have used many methods such as biological treatment, filtration, oxidation, photodegradation and adsorption to reduce the environmental impact of this wastewater [6–10].

Among the above mentioned methods, liquid phase adsorption has been indicated as one of the most convenient and effective techniques since its lower cost, simplicity of design and availability around the world [11]. Adsorbent type is an important factor as well as adsorption amount, temperature, pH, solvent concentration in the adsorption process. Generally, materials which have wide surface area and porous structure are preferred because of their high adsorption capacity in the adsorption studies. In recent years polymers have been widely used as an adsorbent in wastewater treatment because of their wide surface area, stable chemical and mechanical properties, easy and cheap availability, reproducibility, high adsorption capacity, etc. [12,13]. The presence of different functional groups on the surface and interaction between adsorbent–adsorbate are effective for dye removal from aqueous solution.

Nowadays, thanks to the statistical programs prepared by the researchers, a large number of experiments can be carried out in a shorter period of time and more efficiently to obtain optimum conditions. The relationship between the parameters can be determined easily by using mathematical formulas formed at a low cost. Response surface methodology (RSM) is a statistical method to determine main effects and interaction of the two or more factors [14,15]. Central composite design (CCD) is a standard RSM design to analyze the relationship between the parameters easily by using mathematical formulas formed at a low cost. In this way, the

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Nomenclature

ANOVA	Analysis of variance
C_e	Equilibrium concentration of RBBR in solution (mg/L)
C_0	Initial RBBR concentration (mg/L)
C_t	RBBR concentration at time t (mg/L)
CCD	Central composite design
FT-IR	Fourier transform ınfrared spectrometer
K_F	Freundlich isotherm constant (mg/g)
K_L	Langmuir equilibrium constant (L/mg)
k_1	Rate constant for pseudo-first-order ($1\,min^{-1}$)
k_2	Rate constant for pseudo-second-order (g/mg/min)
k_i	Intraparticle diffusion rate constant ($mg/g/min^{0.5}$)
m	Mass of polymer (mg/g)
n	Heterogeneity factor of Freundlich isotherm
Poly(NOPMA)	Poly[2-hydroxy-3-(1-naphthyloxy)propyl] methacrylate
q_e	RBBR amount adsorbed at equilibrium (mg/g)
q_m	Maximum adsorption capacity (mg/g)
q_t	RBBR amount adsorbed at a time t (mg/g)
R	Assumed response
R^2	Correlation coefficient
R_L	Langmuir constant of separation
RBBR	Remazol Brilliant Blue R
RSM	Response surface methodology
SEM	Scanning electron microscopy
T	Temperature (K)
U_i	Real value of independent variable
U_i^o	Actual value at central point of independent variable
ΔU_i	Interval value
V	Volume of the solution (L)
x_i and x_j	Coded values independent variables
X_1	First factor, concentration (mg/L)
X_2	Second factor, adsorbent dosage (mg/g)
X_3	Third factor, contact time (min)
$\pm\alpha$	Axial points
β_0	Constant coefficient
β_i	Linear coefficient
β_{ii}	Quadratic coefficient
β_{ij}	Interaction coefficient
$\Delta G°$	Gibbs-free energy change (kJ/mol)
$\Delta H°$	Enthalpy change (kJ/mol)
$\Delta S°$	Entropy change (kJ/K/mol)

real optimal conditions can be found with respect to the tridimensional graphs [16–18].

So far no study has been reported on the investigation of the adsorption properties and optimization conditions of this original polymer using the RSM approach. We have investigated the influences of initial concentration of RBBR, contact time and adsorbent dosage on adsorption by the poly(NOPMA) using RSM. To learn about mechanism of adsorption, pseudo-first-order, pseudo-second-order and intra-particle diffusion kinetic models were used. Langmuir and Freundlich isotherm models were studied and coefficients were determined. Also thermodynamic properties ($\Delta H°$, $\Delta S°$, $\Delta G°$) of poly(NOPMA) were investigated. The results of this study support that poly(NOPMA) is an inexpensive and quite effective adsorbent for RBBR removal from aqueous solutions.

2. Materials and methods

2.1. Materials

The poly[2-hydroxy-3-(1-naphthyloxy)propyl] methacrylate [poly(NOPMA)] which was synthesized and characterized previously [19] was used as an adsorbent in adsorption process. Remazol Brilliant Blue R (RBBR) dye used as an adsorbate in this study was provided from Sigma-Aldrich. Chemical structure of RBBR and poly(NOPMA) was given in Scheme 1 (molecular formula of dye is $C_{22}H_{16}N_2Na_2O_{11}S_3$, and molar weight of 626.5 g/mol and λ_{max}: 595 nm).

2.2. Experimental design with response surface methodology

In present study, the effect of variables such as initial RBBR concentration (X_1), adsorbent dosage (X_2) and contact time (X_3) on RBBR removal was performed using 2^3-full factorial CCD which categorized in RSM (Design-Expert Software 7) (Table 1). Experimental design points and levels, containing the coded values of the variables used in experimental matrix ($-\alpha$, -1, 0, 1, $+\alpha$), including $2n$ axial points, n_c central points and 2^n factorial points, are given in Table 1.

A variable for statistical calculations was coded by the given formula [20]:

$$x_i = \frac{U_i - U_i^o}{\Delta U_i} \quad (1)$$

where x_i is the coded independent variable value, U_i is the real value of the independent variable, U_i^o is the actual value, at the central point of the independent variable and ΔU_i is the interval value.

The mathematical relation between the three independent variables and process response can be modeled by quadratic equality [21]:

$$R(\%) = \beta_0 + \sum_{i=1}^{n} \beta_i x_i + \sum_{i=1}^{n} \beta_{ii} x_i^2 + \sum_{i=1}^{n} \sum_{j=i+1}^{n} \beta_{ij} x_i x_j \quad (2)$$

where R is the response (RBBR removal efficiency), β_0 is a constant coefficient, β_i is a linear coefficient, β_{ii} is the quadratic coefficients, β_{ij} is an interaction coefficient, and x_i and x_j are the coded values of the adsorption variables.

Design Expert Software 7 was used for analysis of variance (ANOVA), regression and graphical analysis. The Design Expert Desirability Function is used to find the optimum values of the variables for maximum RBBR adsorption.

2.3. Adsorption studies

To realize the batch adsorption properties of polymer, the dye solution at different concentrations (25–100 mg/L) and constant pH (7.0) were shaked with the 0.04 g/50 mL polymer at the constant speed (250 rpm) for different periods at room temperature (20 °C) in a closed Erlenmeyer flask. Before the analyzed process, the dye solutions were centrifuged at 5000 rpm for 10 min and dye concentrations at supernatant were measured using a UV–vis spectrophotometer (UV-1800 Shimadzu) at wavelength of 595 nm.

The adsorption amount (mg/g) of RBBR at time t, q_t, was calculated by:

$$q_t = \frac{(C_0 - C_t)V}{m} \quad (3)$$

Also, adsorption amount at the equilibrium (q_e) was calculated by:

$$q_e = \frac{(C_0 - C_e)V}{m} \quad (4)$$

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