SCIENTIFIC PAPER

DECOLORIZATION OF MODEL WASTEWATER BY ADSORBENT OBTAINED FROM WASTE HEMP FIBERS

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Abstract

Water pollution has already become a significant worldwide problem especially in textile dyeing industry. This paper describes decolorization of dye water modelled by textile dye wastewater. Decolorization was performed on adsorbent made from physico-chemical modified waste hemp fibers, obtained as a by-product from production of ropes. Adsorbent is relatively dispersive and contains heterogeneous porous particles, with carbon as a dominant element. Obtained results have shown that positive effect of adsorption directly depends on contact time, pH, temperature and initial dye concentration. Dye concentration decreases in time, especially when the used concentration is the initial one. The effect of temperature below 40 °C is not significant, but adsorption gets more intensive when performed at 60 °C. The higher degree of decolorization is achieved at lower initial dye concentrations, although the highest initial dye concentration leads to higher dye adsorption. The experimental results of adsorption were described by using Langmuir model. The maximum adsorption capacity ranges from 1.98 to 2.13 mg g^{-1} for linear and 2.03 to 2.12 mg g^{-1} for nonlinear form.

Key word: hemp fibers, reactive dye, decolorization, adsorption, Langmuir isotherms.

Highlights:

- Decolorization of dye water modeled by textile dye wastewater
- Decolorization was performed on adsorbent made from physicochemical modified waste hemp fibers, obtained as a byproduct from production of ropes.
- The Langmuir model shows precise description of experimental data

Introduction

The amount of waste products created by textile industry makes them one of the biggest potential environmental polluters. The textile waste consists of different solid, liquid and gas materials. Therefore, textile industry has expanded its efforts to reduce or find an alternative method for disposal of post-producer textile waste in environment [1,2].

The textile dyeing and finishing industry has also become one of a largest polluter of clean water. Namely, the textile dyes could seriously damage the environment due to the presence of many contaminants, such as toxic organic residues, acids, bases and inorganic matter. Some of dyes are carcinogenic and mutagenic, since they emerged from hazardous chemicals such as benzidine, metals, etc. Outpouring off colored liquid waste in collective water not only affects their aesthetic nature, but also interferes with the transmission of sunlight reducing the photosynthetic activity of plants and disrupting the natural balance of life [3].

The last few years, several physical, biological and chemical techniques of decolorization have been adopted by textile industry [4]. Among them, the best results have been achieved with adsorption method, since it can be used for different kind of dyeing materials [5,6].

One of the basic methods for removal of water contaminants is adsorption technique with porous solid adsorbents. The main characteristics of porous adsorbents are high porosity and surface, as well as suitable physical and chemical nature of internal adsorptive surface. The commercial systems usually use activated carbon as adsorbent for dye removal from wastewater, due its significant adsorptive ability [5-7].

The paper presents a complex study of waste material (hemp fibers from production of ropes) possible application in dye removal from model wastewater made according to the colored wastewater obtained after textile dyeing without any additions other than dye. The waste material was physicochemical treated before application. The aim of this paper was to create the conditions for the potential practical application of the decolorization of colored wastewater from the textile industry based on the concrete knowledge obtained from the analysis of model wastewater.

Experimental

Substrate and chemicals

For modification and application, the decolorization process in textile industry, the hemp waste was used. The waste was obtained during ropes production from 100 % hemp fibres.

Water models were prepared by using a reactive dye, C.I. Reactive Blue 109 (Figure 1). This dye was chosen due its greater presence in practical application, i.e. in the factory dyeing of textiles.

Figure 1.

Preparation of adsorbent

After the waste fibers were collected in Erlenmeyer flask, they were washed up in hot distilled water at 40 ^{o}C, dried and treated with a sulfuric acid (1 g waste fibers: 5 g H_2SO_4) during 48 h at 20 °C. The waste fibers were then rinsed with distilled water and neutralized with solution of $NaHCO_3$. After drying at 250 °C for 2 h and air cooling, the waste fibers were milled and sieved until the particle size was bellow 0.5 mm. Thus, prepared adsorbent was used in the experiment - the decolorization of colored water.

Adsorption procedure

3 g of adsorbent and different concentrations of dye (10, 30, 50, 70 and 100 $mg\cdot dm^{-3}$) were added in Erlenmeyer flasks fixed on a rotary shaker at 140 rpm and thermostated for 5, 10, 20, 30, 45 and 60 min. This amount of adsorbent was selected based on a preliminary study where the amounts of 1, 3 and 5 g were tested. The application of 3 g adsorbent showed the most reliable results with the least variation. The applied dye concentrations are modelled based on dye concentrations lagging after the industrial dyeing of cotton textiles. Temperature of adsorption was varied between 20 °C and 60 °C. The pH of solution was adjusted with 1 M NaOH and 1 M H_2SO_4, and its influence on adsorption process was observed. The preliminary study pointed pH 3 of solution as the most effective for dye adsorption.

After the adsorption was completed, the suspension was filtered, and absorbance was measured at λ=575 nm, with Cary 100 Conc UV-VIS, Varian spectrophotometer.

Calculation of adsorption - models

The degree of dye removal (DR) by adsorption [8] was calculated according to equation:

$$DR = \frac{C_0 - C_t}{C_0} \cdot 100\ (\%) \qquad (1)$$

The amount [8] of adsorbate per mass of adsorbate at different time (q_t) and at equilibrium time (q_e) was expressed as:

$$q_t = \frac{(c_o - c_t)\cdot V}{w} \text{ and } q_e = \frac{(c_o - c_e)\cdot V}{w} \qquad (2)$$

where: C_o - initial dye concentration (mg·dm^{-3}), C_t - dye concentration in time t (mg·dm^{-3}), C_e - equilibrium dye concentration (mg·dm^{-3}), w - mass of adsorbent (g) and V - volume of adsorption solution (dm^3).

Modelling of dye adsorption [8]was done by using linear and nonlinear *Langmuir* adsorption isotherm:

$$\frac{c_e}{q_e} = \frac{1}{Q_{\max} \cdot b} + \frac{c_e}{Q_{\max}} \text{ and } q_e = \frac{Q_{\max} \cdot b \cdot c_e}{1 + b \cdot c_e} \quad (3)$$

where: q_e - amount of adsorbate per mass of adsorbate (mg g^{-1}); Q_{max} - maximum amount of adsorbate binds to adsorbent, (mg·g^{-1}); b - ratio of adsorption and desorption constant velocity (dm^3·mg^{-1}).

Adsorbent characterization methods

The morphology of adsorbate surface was observed with scanning electronic microscope JEOL JSM - 6610LV (Jeol, Japan). The samples were dried for 4 h at 110 °C; the surface was vaporized with gold and observed in instrument chamber with voltage 30 kV.

Qualitative and semi-qualitative elemental contents of sample` surface was X detected by using EDS (Energetic dispersive spectroscopy) X-ray detector.

Results and Discussion

Adsorbent characterization

The yield of waste hemp fibers modification into adsorbent was between 35-40 %. The preparations of the adsorbent were made 3 times, and the yield was presented in the mentioned range. Physico-chemical properties of adsorbent are as follows: bulk density, 0.290 g·cm^{-3}; porosity; 65 %, themoisture content 7.2 %. Those properties effect the surface and bulk morphology of the adsorbent, as well as, its subsequent behaviour when bonding between dye and the surface is forming. They also effect the interior of the powderparticles.

The appearance of adsorbent surface is shown in Figure 2. Used adsorbent is fine dispersive material with heterogeneous porous particles, with diverse shapes and forms. The depths of large particles have expressed cracks, cavities and canals as base of microporous materials. The large porosity is typical for similar adsorbents caused by irregular particles, high degree of amorphization and large number of gaps in structure. Besides shapes, porous particles vary in availability for adsorbing molecules, since they can be closed, open at one or both sides, isolated or bonded [8-10].

The assumption is that dye molecules penetrate through pores and canals into inner particles where they are adsorbed by inner active centres. The porous structure of particle surface facilitates dye diffusion in porous particles, which is already observed in previous investigations [8-10].

Figure 2.

The results of quantitative content of adsorbent obtained by EDS method are given in Table 1. The main component of adsorbent is carbon (61.95 %). Although, the content of aluminium and silicon is low, the high percentage of oxygen can be explained with its presence in a form of aluminium and silicon oxide, as well as alumosilicate. This is a result of adsorbent ability to react with oxygen form air. The similar adsorbents, like activated carbons when exposed to air adsorb oxygen from air increasing the number of superficial functional groups like phenolic, lactones, carboxyl, etc. The sulfur presence is expected since it appears even in activated carbons up to 5 % as organic and inorganic compounds. The complexes of carbon with sulfur are stable and can`t be removed even in temperatures above 1100 °C, unless the reducing atmosphere of hydrogen is present [11,12].

Table 1.

The influence of pH

The selected dye, Reactive Blue 109, is quite soluble considering the presence of five sulfonate groups. It is well known that solubility of those types of dyes depends upon chemical composition, especially on presence of sulfonate groups in molecules of dye. Although the higher number of sulfonate and other soluble groups increases the solubility of dye, it also decreases the dye substantively to adsorbent, since dye can have tendency to water phase.

The dependence of dye removal degree on pH of solution is shown in Figure 3. The maximum dye removal degree is achieved at pH 2-3 (82.5 %). The influence of pH is result of a complex aromatic compound with different functional groups and unsaturated bonds with appropriate ionization potential. It means that molecule of dye can lose or gain charge, depending on pH of solution. The small difference between removing dye from solution on adsorbent may result from impact of pH of solution on water chemistry and available bonds present on adsorbent surface. Also, possible dimerization in solution can also be included. Dimerization can be explained by the presence of intermolecular forces, as well as Van der Waals forces, ion-dipole and dipole-dipole interaction [6,8]. In general, some differences in dye adsorption at different pH of solution suggest the dependence between dye adsorption and surface characteristics of adsorbent and dye constitution.

Beside physical bonds, chemical adsorption of dye can`t be excluded, due to presence of polar functional groups. The functional groups of dye molecules can be bonded to adsorbent in different ways. Sulfonate groups of dye are usually bonded with adsorbent via covalent bond. Although all mechanisms of dye adsorption are not revealed, the ionic exchange, complexation, helatization, electrostatic interaction, hydrogen bonding, etc, can be regarded as fundamental interactions [6,8].

Figure 3.

The influence of contact time

The first contact between adsorbent and dye, or the first stage of dye adsorption depends on degree of mixing the dye solution. The maximum value is achieved with vigorous mixing. When increasing the solution flow rate, boundary layer thickness decreases and dye mass transfer rate increases, proportionally. The finish stage is achieved when dye mass transfer rate is no longer dependable of flow rate. After that, the adsorption rate depends on velocity of dye penetration through adsorbent. The last process implies the adsorbent permeation by dye solution. The dye diffuses through the structure a several times, which is conditioned by contribution of adsorbent bonding force. If this is seen as process of diffusion with adsorption, the velocity of dye penetration is determinate with dye range, as well as structure repulse to dye flow [10].

The correlation between amount of adsorbed reactive dye and different initial concentrations for the lowest and the highest temperature is shown at Figure 4. The results for medium temperature (t=40 °C) are not given because of the similarity, as there are numerous values between the numerous values at 20 and 60 °C. As it expected, the dye concentration in solution decreases with time, and it`s more intensive at higher initial concentrations. The amount of dye that remains after 60 min is higher at higher initial concentrations. It means that higher amount of dye is in the solution at higher initial concentrations, and at the same time higher amount of dye is adsorbed by adsorbent.

The similar work [13] shows the effect of contact time on the percentage removal of reactive orange dye at initial dye concentration (20, 30, 50 mg·dm^{-3}). The percentage removal of dye by different activated carbons was rapid in the beginning but it gradually decreased in time until it reached equilibrium. The pattern of graphs was almost the same for different dye concentrations. The plots reveal maximum percent removal of the dye after about 1 h of shaking. The rate of removal is higher in the beginning due to larger surface area available of adsorbent. After adsorption, the rate of dye uptake is controlled by the rate of dye transported from the exterior to the interior sites of the adsorbent particles.

Figure 4.

The effect of initial concentration

The Figure 5 shows the dependence between degree of dye removal and initial concentrations in different time at 20 and 60 °C. The higher degree of dye removal is achieved at lower initial concentrations. Realistically, the higher amount of dye is adsorbed by adsorbent at the highest initial concentration of dye. Higher temperature causes higher degree of dye removal due to availability of unoccupied sites. Higher concentrations of dye allow higher driving force to overcome the resistance of dye mass transfer from aqua to solid phase. Besides, an increase of the dye concentrations increases the number of collisions between the dye anions and the adsorbent which favours the adsorption process.

According to similar investigation, the amount of adsorbed dye had increased parallel with agitation time. The amount of Reactive red 120 adsorbed onto coconut tree flower carbon (CFC) was lower, compared with jute fibre carbon (JFC) at equilibrium. Also, dye adsorption increased from 2.8 to 6.7 mg g^{-1}, and 3.9 to 11.96 mg g^{-1} with the initial concentration of dye from 10 to 40 $mg \cdot dm^{-3}$ by CFC and JFC, respectively. Evidently, the removal of dye depended on the initial concentration of the dye. Curves were single, smooth and continuous leading to monolayer saturation of reactive dye on the carbon surface [14].

Figure 5.

Adsorption isotherms-models

The equilibrium values, known as adsorption isotherms, are necessary for projection of adsorption systems, as well as prediction of participant's behaviour during the adsorption. There is a different model which describes adsorption equilibrium, such as *Langmuir* adsorption model [10].

Figure 6 shows linear and nonlinear interpretation of Langmuir adsorption isotherm for dye adsorption at three different temperatures. Linear model was represented as dependence between (C_e/q_e) and equilibrium dye concentration (C_e), while nonlinear model was based on dependence between parameter (q) and equilibrium dye concentration (C_e). *Langmuir* constants, $Q_{\max}$ and b, (Figure6a) were determined from slope –intercept form of line (Figure

6a). The same parameters were determined from nonlinear chart (Figure 6b). The confirmation can be found in results quantitatively determine the Langmuir linear and nonlinear model, Table 2.

Nonlinear regression is a more general method that can be used to estimate models' parameters, and it can be applied even if the isotherm model cannot be linearized. Despite significant points covering with linear and nonlinear Langmuir isotherms at all tested temperatures, the priority is given to linear model due its slightly better results as confirmed in a similar study [15].

On the other hand, when it comes to the effect of temperature, (Figure 6b), as expected, adsorption is the most intense at the highest temperature. With initial concentrations of 10-30 mg·dm^{-3} there is as lightly effect of temperature. Above those concentrations (up to 100 mg·dm^{-3}), the curve at 60 °C shows significant increases of adsorbed dye.

The similar conclusion can be found in adsorption of reactive dyes. The adsorption of C.I. Reactive Blue 2 and C.I. Reactive Yellow 2 on activated carbon was studied at temperatures of 25, 35, 45 and 55 °C. The adsorption capacity of both dyes increased at higher temperatures, which indicates adsorption of dyes in this system as endothermic process. For both dyes, the surface coverage increased at higher temperatures, which could be attributed to increased penetration of reactive dyes inside micropores at higher temperatures or the creation of new active sites [16].

Figure 6.

The coefficient of determination (R^2) of Langmuir isotherms determined the model suitability, i.e. efficiency in the linear and nonlinear simulation of isothermal adsorption of reactive dyes on the adsorbent (Table 2). Linear Langmuir model for the adsorption of these at 40 and 60 °C have the highest values of this ratio (R^2=0.998 and 0.996, respectively), followed by the results for the nonlinear model with a slightly lower numerical value.

Parameter Q_{max} is slightly decreased with increasing temperature. It is known that the adsorption of the organic contaminants (including the dye molecules) by material is an exothermic process. Physical and possible chemical bond will lose the connection between organic parts and active site, with increase in temperature [10,17]. In addition, increase of the temperature increases the solubility of the dye and the interaction force between dye molecule and the solution (water) are stronger than those between the molecule and the adsorbent, which reduces the adsorption. The value of parameter b increases with increasing temperature, due to linear energy distribution between surface groups with different level of activation energy for appropriate reaction of adsorption. Also, it can be due to correlation between parameter b - energy (velocity) of adsorption and variations of surface and porosity of adsorbent. This parameter refers to the free energy of adsorption ΔG ($b \propto e^{-\Delta G/RT}$) showing an affinity of adsorbent for dye binding. Its value is reciprocal to the dye concentration at which a half of dye saturation was achieved, so the high value of *b* indicates the start of a desirable isotherm which reflects the high affinity of adsorbent for the adsorbate. Higher values of parameter b were obtained for adsorbent-dye system, which means stronger dye binding for adsorbent [17].

Table 2.

Like this study, low cost coconut shell based activated carbon (CSAC) was produced from agricultural wastes for the equilibrium studies of Reactive Blue 19 (RB19) dye adsorption from aqueous solution. The equilibrium data for the adsorption of RB19 onto the CSAC were fitted using the Langmuir and Freundlich adsorption isotherms. Based on the values of the isotherms model parameters with their Pearson's correlation coefficients and adj. R^2, coefficient of determination it can be observed that the equilibrium adsorption data for the three different process conditions are well fitted with the two isotherms. These results show that the experimental data agree with both isotherm models. In the case of Langmuir isotherm, there is homogeneity in the nature of CSAC surface, meaning that each molecule of the RB 19 dye/active site of the CSAC adsorption has equal activation energy and shows the formation of monolayer coverage of the dye molecule on the outer surface of the CSAC [18].

Table 3 shows Langmuir's constant data, the maximum adsorption capacity for sorbents based on agricultural and cellulosic materials used to purify colored water, i.e. for the decolorization of water. It is evident from the table that the value of the maximum absorption capacity data from this research is within the range of other results from published works.

Table 3.

Conclusion

The removal of reactive dye with adsorbent obtained from waste fibers depends upon contact time, initial dye concentration, temperature and pH of solution.

Obtained adsorbent is finely dispersed material with heterogeneous porous particles, while inner structure contains expressed cracks, holes and canals. Dominant element of adsorbent is carbon, while the higher amount of oxygen correlates with oxide of aluminium, silicon and aluminosilicate. During the adsorption, dye concentration reduces in time, and the maximum removal of dye is achieved at pH 2-3. The effect of temperature is visible only at the highest applied value. The lower initial concentrations of dye led to a higher degree of adsorbate removal. Although the removal of reactive dye with adsorbent from water solution have been good described by linear and nonlinear Langmuir model, the preference is given to a linear model that fits the experimental data best. The Langmuir constant, maximum adsorption capacity ranges from 1.98 to 2.13 $mg \cdot g^{-1}$ for linear form.

The question arises, what to do with the used adsorbent. In addition to regeneration, which is the first choice, there is also easier storage of solid adsorbent after adsorption compared to liquid waste, easy destruction in waste incineration plants, perhaps application in construction, etc.

The innovative aspect of the research relates to the use of hemp waste fibers for the production of a new sorbent, which can effectively remove textile dye from the water after the dyeing process. Also, based on the adsorption of textile reactive dye, it can be assumed that successful adsorption will also be realized in other groups of textile dyes that are used for dyeing natural as well as synthetic textiles.

The adsorbent created by waste hemp presents an efficient adsorbent for the removal of reactive dyes from the aqueous solution with possible application on industrial scale. In the future, the characterization of similar solid waste materials, as raw material to produce the adsorbent, can assist in the process of disposing a colored liquid waste.

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FIGURE CAPTIONS

Figure 1. Chemical structure of reactive dye, C.I. Reactive Blue 109.

Figure 2. Micrograph of adsorbent surface (Magnification 750×, left and 2000×, right).

Figure 3. The effect of pH on adsorption of reactive dye (C_o=100 mg dm^{-3}).

Figure 4. The change of reactive dye amount during adsorption at a) 20 ^{o}C and b) 60 ^{o}C

Figure 5. Percent of removed dyevs. initial concentration of dye at a) 20 ^{o}C and b) 60 ^{o}C

Figure 6. Linear a) and nonlinear b) Langmuir adsorption isotherms at different temperature for reactive dye-adsorbent system.

Table 1. Quantitative composition of adsorbent by EDS analysis.

Spectrum	**(%)**							
	C	**O**	**Na**	**Al**	**Si**	**S**	**Ca**	**Total**
Mean	61.95	29.82	4.70	0.03	0.06	3.44	0.00	100.00
Std. deviation	2.35	4.83	1.36	0.04	0.14	2.79	0.00	-
Max.	65.19	33.66	6.64	0.07	0.31	8.02	0.00	-
Min.	59.86	23.69	2.79	0.00	0.00	1.55	0.00	-

Table 2. Analytical expressions of linear and nonlinear Langmuir isotherms with coefficient for reactive dye-adsorbent system.

Model	**Analytical model expression**	**Model parameters**		***R*²**
Langmuir, 20 °C (linear model)	$C_e/q_e = 1.27 + 0.47 \cdot C_e$	Q_{max} (mg g^{-1})	2.05	0.985
		b (dm^3 mg^{-1})	0.08	
Langmuir, 40 °C (linear model)	$C_e/q_e = 3.46 + 0.50 \cdot C_e$	Q_{max} (mg g^{-1})	1.98	0.998
		b (dm^3 mg^{-1})	0.14	
Langmuir, 60 °C (linear model)	$C_e/q_e = 5.86 + 0.49 \cdot C_e$	Q_{max} (mg g^{-1})	2.13	0.996
		b (dm^3 mg^{-1})	0.37	
Langmuir, 20 °C (nonlinear model)	$q_e = \frac{0.17 \cdot C_e}{1 + 0.08 \cdot C_e}$	Q_{max} (mg g^{-1})	2.12	0.976
		b (dm^3 mg^{-1})	0.08	
Langmuir, 40 °C (nonlinear model)	$q_e = \frac{0.26 \cdot C_e}{1 + 0.13 \cdot C_e}$	Q_{max} (mg g^{-1})	2.04	0.994
		b (dm^3 mg^{-1})	0.13	
Langmuir, 60 °C (nonlinear model)	$q_e = \frac{0.85 \cdot C_e}{1 + 0.42 \cdot C_e}$	Q_{max} (mg g^{-1})	2.03	0.971
		b (dm^3 mg^{-1})	0.42	

Table 3. Comparison of the maximum monolayer capacities of textile dye on various agricultural and cellulosicadsorbents.

Adsorbent	**Adsorbate**	**Maximum adsorption capacity, mg·g^{-1}**	**References**
Waste hemp fibers (acid modification)	Reactive Blue	1.98-2.13	This study
Sugarcane bagasse pith (Physicalactivation by carbonization)	Reactive orange	3.48	[13]
Sugarcane bagasse pith (50 % $ZnCl_2$followed by pyrolysis)	Reactive orange	2.83	[13]
Sugarcane bagasse pith (28 % H_3PO_4followed by pyrolysis)	Reactive orange	1.80	[13]
Coconut shell based activated carbon	Reactive Blue	2.22-2.91	[18]
Coir pith	Congo red	6.72	[19]
Coir pith	Blue acid	5.87	[19]
Coir pith	Procion orange	2.60	[19]
Thermally activated coir pith carbon from coconut husk	Methylene blue	5.87	[20]
Raw *Posidonia oceanica* fibres	Methylene blue	5.56	[21]
Yellow passion fruit peel	Methylene blue	0.0068	[22]
Shells of Macore fruit	Methyl orange	2.96	[23]
Shells of Macore fruit	Methylene blue	6.85	[23]
Almond shell-activated carbon	Methylene blue	1.33	[24]
Walnut shell-activated carbon	Methylene blue	3.53	[24]
Apricot stones-activated carbon	Methylene blue	4.11	[24]

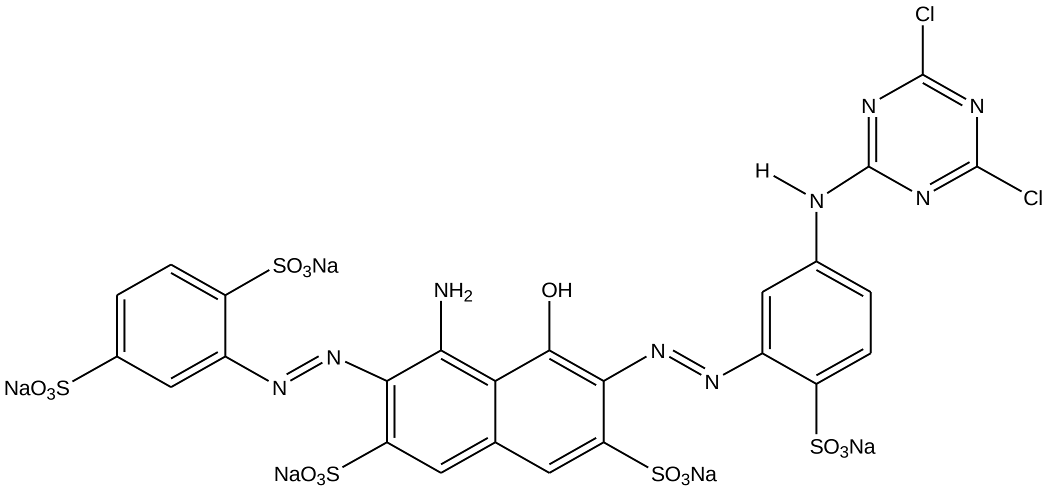


Figure 1.

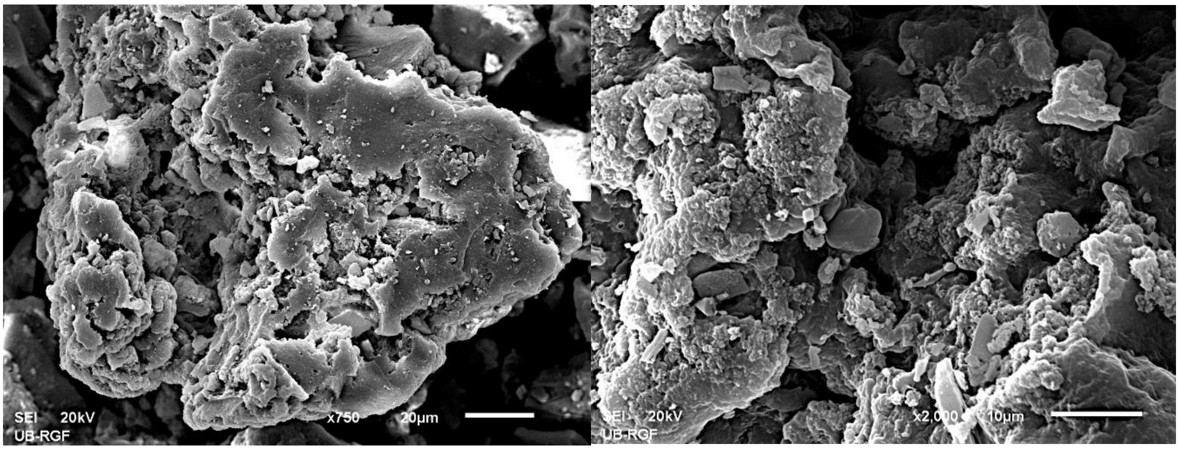


Figure 2.

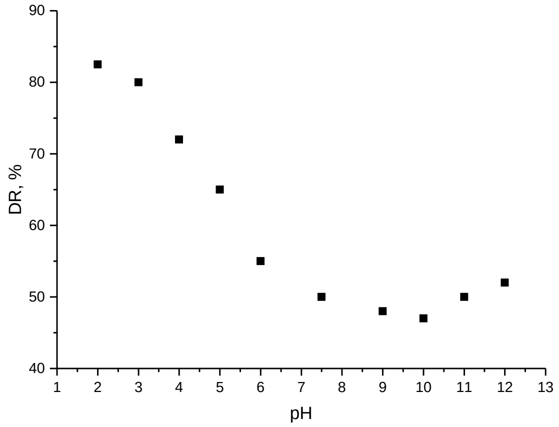
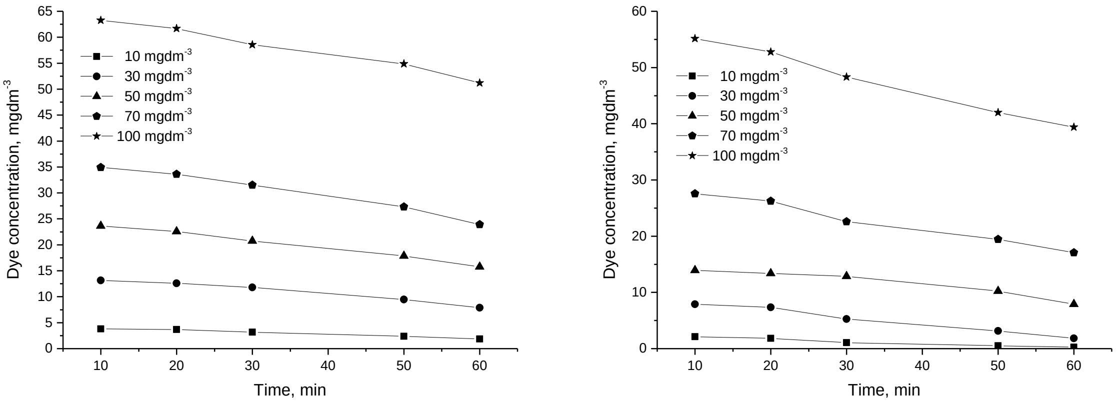


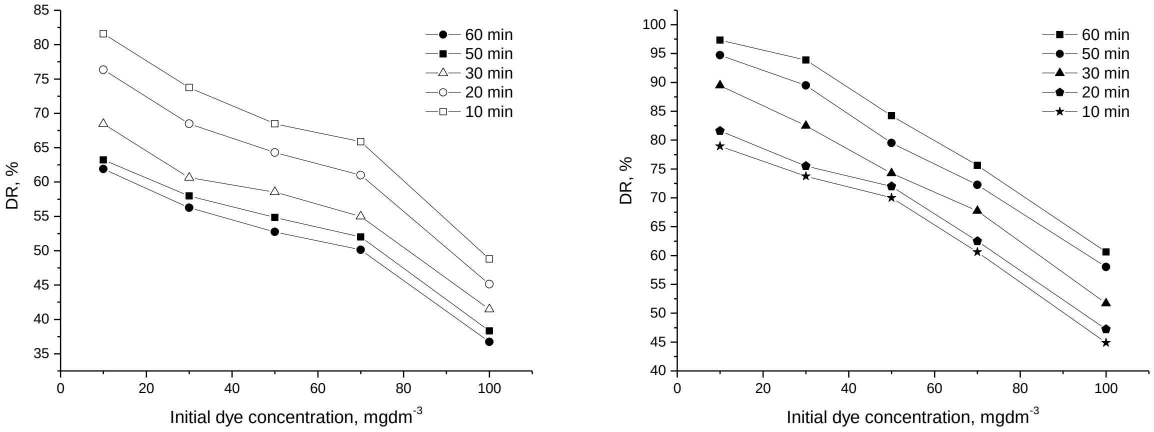
Figure 3.



Dye concentration, mgdm^{-3}
Time, min
10 mgdm^{-3}
30 mgdm^{-3}
50 mgdm^{-3}
70 mgdm^{-3}
100 mgdm^{-3}
0
5
10
15
20
25
30
35
40
45
50
55
60
65
10
20
30
40
50
60

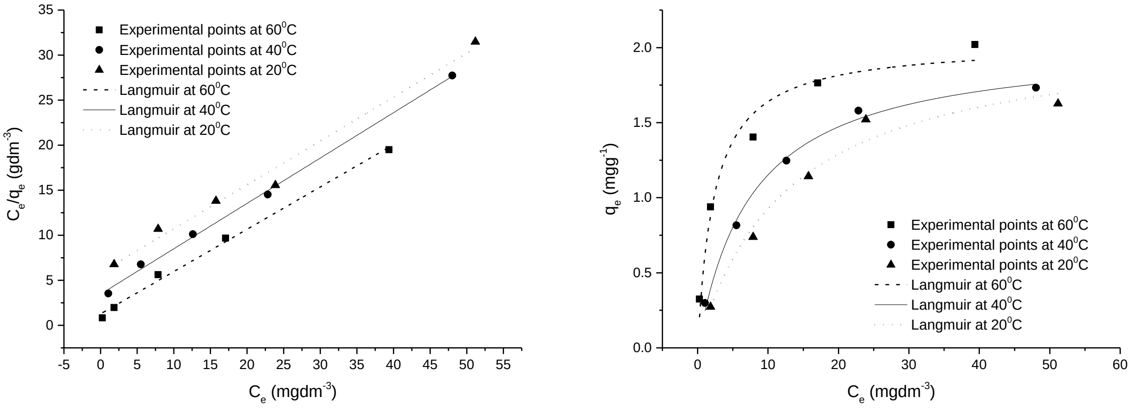
a) b)

Figure 4.


DR, %
85
80
75
70
65
60
55
50
45
40
35
0
20
40
60
80
100
Initial dye concentration, mgdm^{-3}
60 min
50 min
30 min
20 min
10 min
DR, %
100
95
90
85
80
75
70
65
60
55
50
45
40
0
20
40
60
80
100
Initial dye concentration, mgdm^{-3}
60 min
50 min
30 min
20 min
10 min

a) b)

Figure 5.


Experimental points at 60⁰C
Experimental points at 40⁰C
Experimental points at 20⁰C
Langmuir at 60⁰C
Langmuir at 40⁰C
Langmuir at 20⁰C
C_e/q_e (gdm^-3)
35
30
25
20
15
10
5
0
-5
0
5
10
15
20
25
30
35
40
45
50
55
C_e (mgdm^-3)
q_e (mgg^-1)
2.0
1.5
1.0
0.5
0.0
0
10
20
30
40
50
60
C_e (mgdm^-3)

a)

b)

Figure 6.