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Decolorization of ionic dyes from synthesized textile wastewater by nanofiltration using response surface methodology

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ABSTRACT

Decolorization of aqueous solutions containing ionic dyes (Reactive Blue 19 and Acid Black 172) by a TFC commercial polyamide nanofilter (NF) in a spiral wound configuration was studied. The effect of operating parameters including feed concentration (60-180 mg/l), pressure (0.5-1.1 MPa) and pH (6-10) on dye removal efficiency was evaluated. The response surface method (RSM) was utilized for the experimental design and statistical analysis to identify the impact of each factor. The results showed that an increase in the dye concentration and pH can significantly enhance the removal efficiency from 88% and 87% up to 95% and 93% for Reactive and Acid dye, respectively. Results showed that dye removal efficiency increased by an increase in pressure from 0.5 to 0.8 MPa, while further increase in pressure decreased the removal efficiency. The maximum dye removal efficiencies which were predicted at the optimum conditions by Design Expert software were 97 % and 94 % for Reactive Blue 19 and Acid Black 172, respectively. According to the results of this study, NF processes can be used at a significantly lower pressure and fouling issue for reuse applications as an alternative to the widely used RO process.

1. Introduction

The textile industry is known as one of the largest consumers of fresh water and color. According to Lucas et al. [1-3] between 25 and 250 m^3 water is consumed per each ton of product. Therefore, these industries produce a high volume of wastewater including complex structures such as wetting agents, dyes, fixing agents, softeners and many other additives [4, 5]. Color, high pH, high COD and low biodegradability are characteristics of textile wastewater [6-8] The annual production of synthetic dyes globally, exceeds over 700000 tons and around 10% to 15% of the used dyes in the dyeing process appear in the sewage [9,10] There are around 10000 different dyes which should be monitored in waste water streams [11]. Released textile wastewater containing dyes are the main sources of toxicity in environment, which imposes a significant risk to the living organisms. There are a few reports that have associated the textile chemicals to causing cancer and other mutagenic diseases [12]. Consideration and appropriate treatment should be taken into account to ensure that the final discharge will not cause any harm to society and the environment. A variety of methods such as, chemical precipitation, adsorption, ion exchange process, electro-coagulation, membrane systems, and even biological treatments have been used for dye removal from industrial effluent. Among them, chemical precipitation and reduction processes not only need separation stage and produce high amounts of sludge, but also require significant treatment chemicals [13]. Biologically assisted approaches have also been reported as an inefficient treatment due to the complex and stable structure of synthetic dyes [14]. The pressure driven membrane processes (MF, UF, NF and RO) have been found to be

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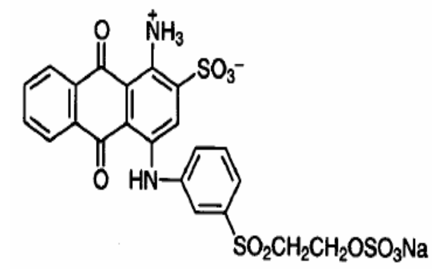
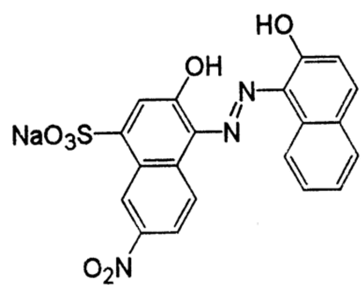
efficient and environmentally friendly. Although RO is known as the most efficient separation technique in terms of permeate quality and dye rejection, the required cost and high pressure as well as fouling phenomenon has limited its application. In comparison with RO, NF can be used in textile wastewater treatment to remove dyes at a significantly lower pressure and thus lesser fouling [15, 16]. Hassani et al. investigated dye removal rate of four different feed types of dyes (Acidic, Disperse, Reactive and Direct) by utilizing a spiral wound nanofiltration membrane (MWCO of 90 KDalton). Their results showed that increasing the dye concentration can lead to higher dye removal efficiency (98 %). Also, at different pressures the removal efficiency for Acidic and Reactive dyes reaches to 99.7 %. However, the effect of pH on color removal efficiency was not considered [17]. Sanchuan et al., found that the trans-membrane pressure and dye concentration can significantly affect the dye retention and water permeability. According to their results, the dye retention, water permeability and salt rejection rate of an aqueous solution containing 2000 mg/l Congo red and 10000 mg/l NaCl were 99.8%, 7.0 $l/m^2.h.bar$, and lower than 2.0%, respectively. It should be pointed out here that they did not study the optimization of operational parameters [18]. Sahinkaya et al., used a combination of activated sludge with NF to treat denim textile wastewater and to reach reuse standards quality. The COD removal efficiency in the activated sludge reactor was 91±2% and 84±4% based on the total and soluble feed COD, respectively. They managed to achieve 75% color removal efficiency through the adsorption of color on biomass or precipitation within the reactor. The effective parameters and optimum conditions had not been investigated [19]. In an investigation performed by Liu et al. reverse osmosis (RO) and nanofiltration (NF) were evaluated and compared for a textile effluent treatment in terms of COD removal, salinity reduction and permeate flux. However, color removal efficiency and the impressive parameters had not been studied [20]. Acid and Reactive dyes can escape from conventional treatments, because they show resistance against microbial, chemical and photolytic degradation [21]. As mentioned before, NF can be used efficiently for treating textile effluent at low pressure and less fouling, which significantly minimizes dyes escaping. The aim of the present work is to optimize the operational parameters of a NF process for removal of Reactive Blue 19 (RB 19) and Acid black 172 (AB 172) from an aqueous solution. The relationship between dye removal efficiency and three main parameters including pH, initial dye concentration and pressure, were evaluated by applying response surface methodology.

2. Materials

Reactive Blue 19 (with molecular formula of $C_{22}H_{16}N_2Na_2O_{11}S_3$, MW=626 $gmol^{-1}$, λ_{max}=592nm) and Acid Black 172 (with molecular formula of $C_{20}H_{12}N_3NaO_7S$ and MW=461$gmol^{-1}$, λ_{max}=572nm) were supplied by Alvan Sabet Co. and were used without further purification. The structures of Reactive Blue 19 and Acid Black 172 are presented in Fig. 1. To adjust pH, HCl (37%), NaOH were purchased from Merck Company of Germany.



(a)



(b)

Fig. 1. Reactive Blue 19 structure (a) and Acid Black 172 (b).

2.1. Experimental set-up

In this study as schematically presented in Fig. 2, a continuous co-current NF set up was used. A commercial polyamide thin film composite membrane in a spiral wound configuration was used for the NF process. The membrane specifications are presented in Table 1. The pumps used in this system are diaphragm-type. The pumps output flow and pressure are 0.8 l/min and 6 bar, respectively.

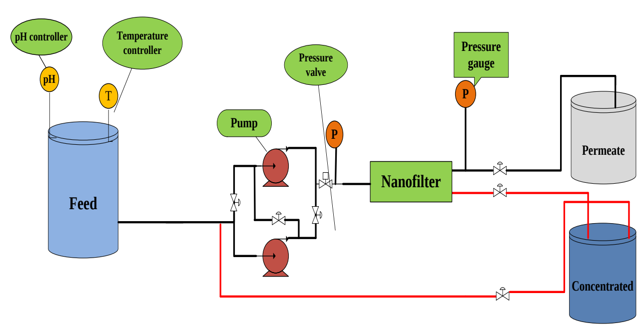


Fig. 2. Schematic represents the NF experimental apparatus.

2.2. Experimental procedure

The synthesized wastewater was prepared by mixing the dye powders (Reactive Blue 19, Acid Black 172) in three concentrations of 60, 120 and 180 mg/L in distilled water. The pH of solution was adjusted by 0.1M HCl and 0.1M NaOH. The temperature of the solution was kept constant at room temperature (25 C^0) with recovery percentage of 75±3% of the feed volume. The recovery percentage was kept constant by varying the feed and permeate fluxes. All measurements were performed according to the American Public Health Association water and wastewater examination methods [22]. The permeated dye concentrations were obtained by analyzing the absorbance at maximum wavelength of each dye (592 and 572nm for RB19 and AB172) using a V-570 spectrophotometer following the standard method No. 2120C (Spectrophotometric method – single wavelength method). The corresponding concentration was then calculated from a calibration curve with ten points (R^2 was 0.98 for reactive dye and 0.95 for acid dye). The dye removal percentage was calculated by using NF membrane rejection as shown below:

$$\mathrm{DR}(\%) = \left[1 - \frac{C_P}{C_f}\right] \times 100 \quad (1)$$

Where DR is the dye removal percentage, $\boldsymbol{C_p}$and $\boldsymbol{C_f}$ are permeate and feed concentration, respectively.

Table1. Commercial polyamide TFC membrane specifications.

Provider	CSM Company, Korea
Skin layer	Polyamide
Maximum tolerable pressure	20 bar
pH range	2-11
Isoelectric point	4.5
Surface electrical charge	Negative
Active surface (m^2)	0.35

2.3. Response surface methodology

The response surface methodology (RSM) is an effective method for the optimization of responses.The RSM method can be employed on the basis of different designs including Central Composite, Box-Behnken, One Factor, D-Optimal, etc. [23]. In current study, the Box-Behnken design was selected to optimize the responses for a three level factors design. A full factorial design for the three parameters of pH, pressure and concentration requires 27 runs while by using the design the total number of experiments was reduced to 15. The confidence level (CL) for randomly conducted experiments was 95% to avoid possible errors due to the systematic bias. In this study, the aim was to obtain maximum removal percentage of Reactive Blue 19 and Acid Black 172, which were considered as the responses. The contour plots and the analysis of variance (ANOVA) evaluation were used to analyze the results. The contributory factors and their selected levels are presented in Table 2.

Table 2. Factors and selected parameters.

Factors	Level 1	Level 2	Level 3
Dye	60.0±2.0	120±3.0	180.0±5.0
pH	6.00±0.10	0.80±0.1	10.00±0.1
Pressure (MPa)	0.50±0.10	0.80±0.1	1.100±0.1

3. Results and discussion

The experimental design and the responses of the experiments for both dyes are presented in Table 3 and 4. Results are the average values obtained by 3 parallel experiments. The ANOVA table for the dye removal efficiency of Reactive Blue 19 and Acid Black 172 is shown in Table 5 and 6, respectively.

Table 3. Box-Behnken design results for Reactive Blue 19.

Exp. No.	Dye Conc.	pH	Pressure (MPa)	Dye Removal
1	120.0±2	8.00±0.1	0.8±o.1	93.8
2	60.00±3	8.00±0.1	1.1±o.1	90.5
3	180.0±5	10.0±0.1	0.8±0.1	95.7
4	180.0±2	6.00±0.1	0.8±o.1	91.4
5	60.00±2	8.00±0.1	0.5±0.1	89.6
6	60.00±5	10.0±0.1	0.8±0.1	93.4
7	120.0±3	6.00±0.1	1.1±0.1	89.3
8	120.0±3	10.0±0.1	0.5±0.1	91.0
9	180.0±5	8.00±0.1	1.1±0.1	92.8
10	120.0±5	8.00±0.1	0.8±0.1	92.9
11	120.0±2	8.00±0.1	0.8±0.1	93.5
12	120.0±3	10.0±0.1	1.1±0.1	92.7
13	60.00±3	6.00±0.1	0.8±0.1	89.6
14	120.0±3	6.00±0.1	0.5±0.1	88.9
15	180.0±3	8.00±0.1	0.5±0.1	91.7

3.1. Effect of pH

The results in Figs. 3a and 4a indicate that an increase in pH from 6 to 10 has a positive effect on the color removal efficiency. This might be related to the two factors of electrostatic repulsive force and membrane swelling. From Table 1, the isoelectric point for applied commercial NF membranes is at pH of 4.5 above which the membrane surface is negatively charged. With an increase in pH, the electrical repulsive force between the membrane surface and the dye molecules increases and thus the dye removal efficiency rises to 95% and 93% for Reactive Blue and Acid Black, respectively. According to Mahmoodi et al. [24], the membranes will swell at higher pH values and cause the pores to shrink. Reduction in pore size will directly increase the membrane rejection and removal efficiency. However, as the experiment was conducted at constant flux and no significant pressure increment was observed when pH increased, the contribution of pore shrinkage on the removal efficiency enhancement is negligible.

Table 4. Box-Behnken design results for Acid Black 172.

Experiment No.	Dye Concentration ($^{1-}$mg L)	pH	Pressure (MPa)	Dye Removal Percentage
1	120.0±3	8.00±0.1	0.8±o.1	91.4
2	120.0±5	10.0±0.1	0.5±o.1	89.2
3	120.0±2	6.00±0.1	0.5±0.1	87.0
4	180.0±2	10.0±0.1	0.8±o.1	93.6
5	60.00±3	10.0±0.1	0.8±0.1	91.0
6	180.0±3	8.00±0.1	1.1±0.1	90.9
7	60.00±2	8.00±0.1	1.1±0.1	88.5
8	180.0±3	6.00±0.1	0.8±0.1	90.2
9	120.0±5	10.0±0.1	1.1±0.1	90.8
10	120.0±3	8.00±0.1	0.8±0.1	91.8
11	180.0±3	8.00±0.1	0.5±0.1	90.0
12	60.00±2	6.00±0.1	0.8±0.1	88.0
13	120.0±3	8.00±0.1	0.8±0.1	91.5
14	120.0±5	6.00±0.1	1.1±0.1	87.4
15	60.00±5	8.00±0.1	0.5±0.1	88.0

Table 5. Analysis of variance for Reactive Blue 19 removal percentage.

Model terms	Mean square error	Sum of the error squares	Degree of freedom	F-value	P-value	Status
Model	5.840	52.60	9	28.060	<0.0001	Significant
A:dye concentration	9.220	9.220	1	44.280	<0.0001	Significant
B: pH	23.94	23.94	1	114.95	<0.0001	Significant
C: P	2.050	2.050	1	9.8400	0.0257	Significant
A×B	0.065	0.065	1	0.3100	0.6004	Not significant
A×C	0.001	0.001	1	0.0480	0.8352	Not significant
B×C	0.440	0.440	1	2.1200	0.2049	Not significant
A×A	0.013	0.013	1	0.0620	0.8132	Not significant
B×B	1.870	1.870	1	8.9800	0.0302	Significant
C× C	15.66	15.66	1	75.170	0.0003	Significant
Lack of fit	0.270	0.810	3	2.2900	0.3184	Not significant

3.2. Effect of dye concentration

Increasing the dyes concentration resulted in higher removal efficiency due to the increase in size exclusion mechanism and space prevention [25]. As can be seen in Figs. 3a and 4a, Reactive Blue 19 removal efficiency is 6% more than that of Acid Black dye. As can be seen in Table 5 and 6, F-values for the concentration parameter are 293 and 44 for Acid Black 172 and Reactive Blue 19 respectively. Therefore, the concentration is the second important factor after pH for removal of both dyes. Dyes and the membrane surface charge at the NF operational pH are negative; as a result, by increasing the dyes concentrations the repulsive forces between the membrane surface and the dyes molecules increase.

3.3. Effect of pressure

In order to increase the pressure, the feed and permeate flow valves were adjusted to increase the feed side pressure while keeping recovery percentage constant. As shown in Figs. 3c and 4c, increasing the pressure from 0.5 to about 0.8 MPa led to an increase in removal efficiency for both dyes. However, further increase in pressure revealed a marginal reduction in the removal efficiency [26]. Operating NF processes at higher pressure increases the chance of concentration polarization which promotes fouling on the membrane surface and consequently increases the intrinsic membrane rejection and removal efficiency [26]. The maximum removal efficiency for dye removal from contaminated water was estimated 97.77% by Box-Behnken method, which was attained at the optimum conditions of 180 mg l^{-1} of dye, pressure of 0.825 MPa and pH of 9.9 for Reactive Blue 19. However, the maximum efficiency based on the experimental data for Reactive Blue 19 was 95.7% at the concentration of 180 mg l^{-1}, pH of 10 and pressure of 0.8 MPa. The maximum estimated removal efficiency at the concentration of 178 mg/l of dye, pressure about 0.87 MPa and pH of 10 for Acid Black 172 from Box-Behnken method was 93.63% which is close to the experimentally obtained removal efficiency (93.6%, see Table 4)

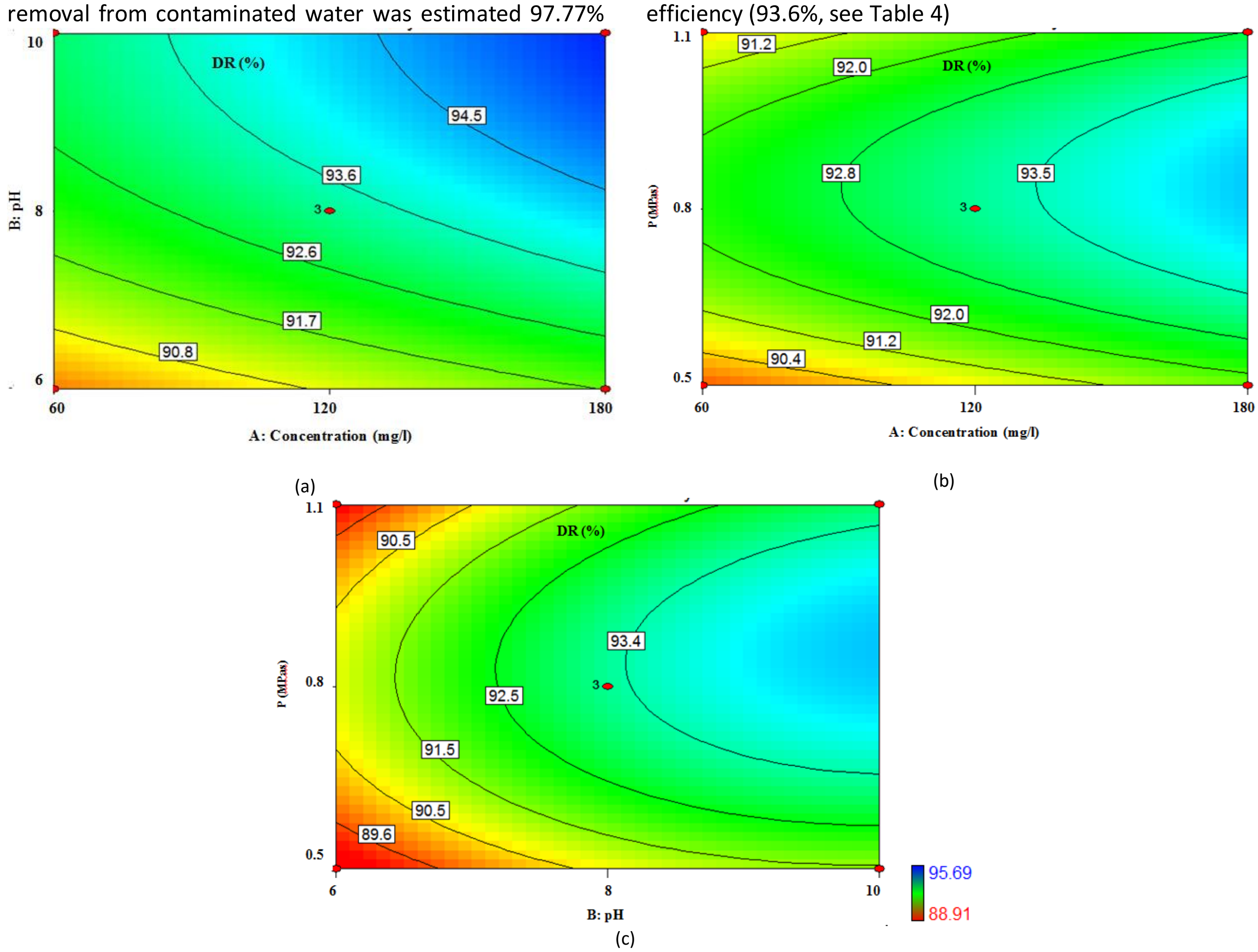


Fig. 3. Contour plots of the Reactive Blue 19 removal efficiency; (a): the effect of dye concentrations and pH on dye removal percentage (DR %) of Reactive Blue 19 at constant pressure. (b): the effect of pressure and dye concentration on the removal efficiency at constant pH (c): the effect of pH and pressure on the removal efficiency at constant dye concentration.

Table 6. Analysis of variance for Acid Blue 172 removal percentage.

Model terms	square Mean error	Sum of the error squares	Degree of freedom	F-value	P-value	Status
Model	5.360	48.22	9	156.95	<0.0001	Significant
A:dye concentration	10.10	10.10	1	293.30	<0.0001	Significant
B: pH	17.17	17.17	1	502.95	<0.0001	Significant
C: P	1.400	1.400	1	41.090	0.0014	Significant
A×B	0.130	0.130	1	3.8000	0.1089	Not significant
A×C	0.034	0.034	1	1.0000	0.3627	Not significant
B×C	0.420	0.420	1	12.380	0.0170	Not significant
A×A	0.013	0.013	1	0.1100	0.7522	Not significant
B×B	2.250	2.250	1	65.870	0.0005	Significant
C× C	17.43	17.43	1	510.67	<0.0001	Significant
Lack of fit	0.092	0.031	3	0.7800	0.6054	Not significant
Pure Error	0.079	0.039	2	-	-	-

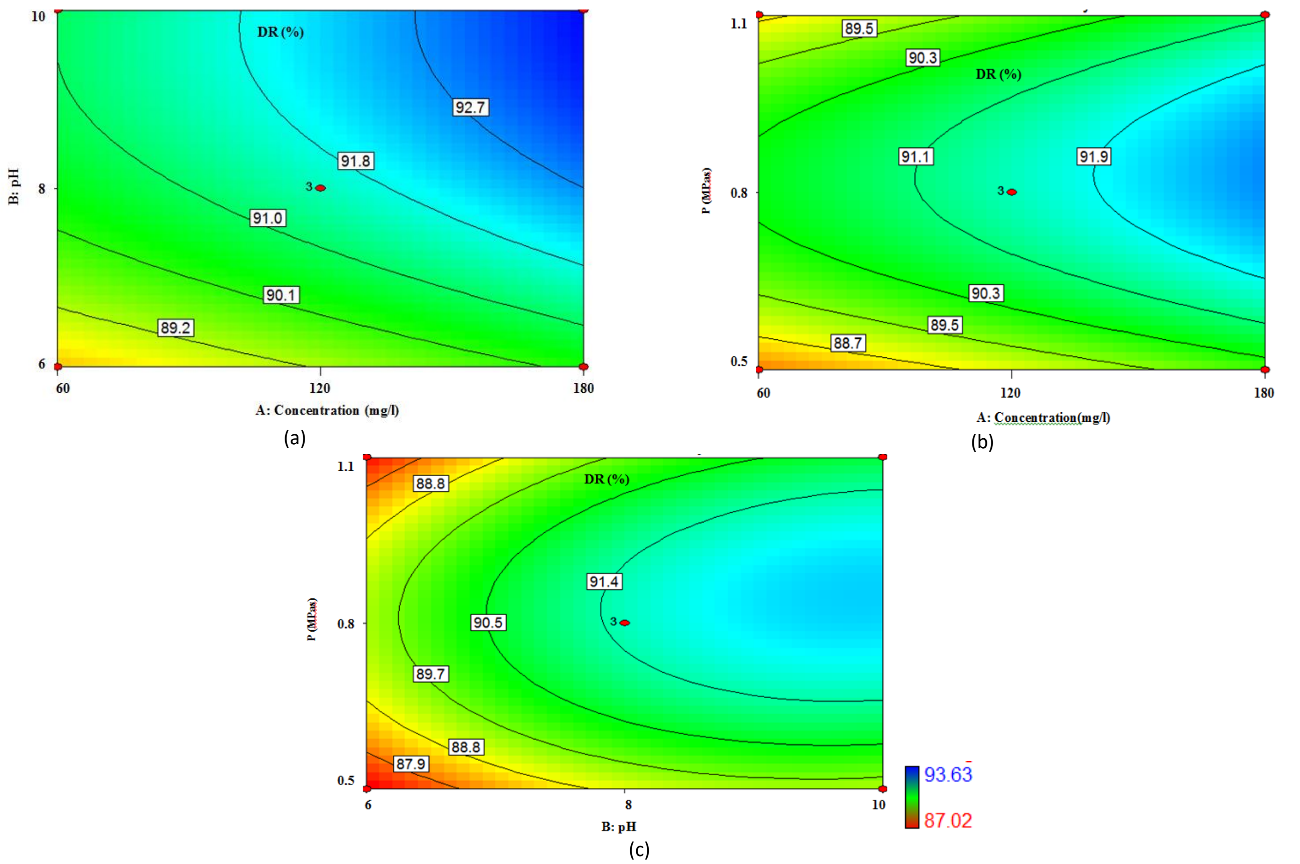


Fig. 4. Contour plots of the Acid Black 172 removal efficiency; (a): the effect of dye concentrations and pH on dye removal (DR %) efficiency of.Acid Black 172 at constant pressure. (b): the effect of P and dye concentration on the removal efficiency at constant pH (c): the effect of pH and pressure on the removal efficiency at constant dye concentration.

4. Conclusions

According to the obtained results, the commercial spiral wound polyamide nanofilter (TFC) was remarkably efficient for removing dyes from textile wastewater. pH had the most significant effect among the other factors (having the highest F-value) on the removal of both applied dyes (Reactive Blue 19 and Acid Black 172). The results indicated that with an increase in pH and dye concentration, the removal efficiency of both colors increased. Also, our experimental data revealed that under optimum condition, operating pressure had a significant influence on the dye removal efficiency. The agreement between the estimated and obtained removal efficiency showed that the design of experiments using response surface method not only can be considered as a good choice for the experimental design and statistical analysis but also for the optimization of process parameters.

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