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Optimization of separation phases of activated carbon by hydrocyclone process on using response surface methodology

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ABSTRACT

The aim of this study is to build an efficient hybrid treatment process for the depollution of mixed and highly polluted industrial textile wastewater. The proposed system consists of an effective coupling between adsorption by activated carbon and a phase separation by a hydrocyclone. The response surface methodology using the Box–Behnken experimental design was applied and explored in order to optimize the most influencing factors. The experimental results showed that the decolorization efficiency (CR%) by activated carbon for Novacron Blue 4R (NB4R) dye was 87.15% under optimal treatment conditions with a pH of 11, a concentration of carbon of 12.42 g/L and an initial dye concentration of 62.50 mg/L. The optimization study of separation of activated carbon by the hydrocyclone allowed a separation efficiency (ES%) about 88.74%. The cited efficiency was ensured in the optimal conditions which were a volume flow rate of 81.83 L/min, a carbon concentration of 12.42 g/L with a size of 0.5 m. The proposed hybrid process is shown to be more efficient in terms of treatment efficiency and recovery of the carbon particles. In addition, big improvements were reached especially in the term of the final wastewater quality after adsorption/hydrocyclone combination when compared with the current conventional treatment method in the concerned industry.

Keywords: Hydrocyclone; Activated carbon; Optimization; Response surface methodology; Industrial wastewater

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1. Introduction

The treatment of industrial textile wastewaters remains a difficult task as it exhibits large variations of flow rate and composition, high residual concentrations of dyes, auxiliaries, and salts in addition to big fluctuations on physical parameters such as pH and temperature [1–3]. The persistence of the current manufactured dyes or auxiliaries makes additional difficulties given that they are selected for their good resistance and stability even after a long exposure to light, sun, oxidants, high temperatures, or with microorganisms [4–6].

Facing such limits, textile wastewater treatments peculiarly still unchanged and typical biological methods remain an integral part of any wastewater treatment plant in this field [7,8]. The most adopted process still the activated sludge treatment [8]. Some aerated lagoons are also employed but sequential batch reactors represent the more recent biological systems adopted by textile factories. All of these treatments are based on the use of bacteria or

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microorganisms. In the aerobic degradation process, the involved microorganisms (also called aerobes), use molecular/free oxygen to assimilate organic impurities and convert them into carbon dioxide, water, and biomass. In the anaerobic degradation process, anaerobes do not require air (molecular/free oxygen) and the assimilated organic are transformed in this case to methane, carbon dioxide gas, and biomass [8–10]. Prior to each biological application, the effluent must be neutralized and temperature must be adjusted in order to avoid chocking the microorganisms [10]. Definitely, the selection of the neutralization reagent must satisfy some economical, technical, and ecological characters. Such a balance could be ensured by the use of carbon dioxide. The generation, in situ, of the carbonic acid ensures a better compromise between economic and ecological benefits. It also prevents problems related to transport and stockage and the excessive accumulation of chlorides, sulfates, and other salts [11,12].

In spite of all the cited advantages, and regarding the relevant environmental impact of textile effluents and instability problems (chock risk), basic ecological indicators remain unsatisfactory after biological treatments [1,9,10,13,14]. In fact, viable biomass that effectively removes both color and chemical oxygen demand from wastewater is not successfully ensured until yet. In addition, all the implanted processes do not prove efficient in a long-term study [10,15]. The stochastic behavior makes additional complications to the monitoring of industrial discharges and related depollution. The neutralisation step, either by the generation of the ecological carbonic acid, was also criticised because it does not conduct to a satisfactory acidification (pH could not be lowered down to 8 with this acid). This led to the formation of isolated and dispersed biomass making harder the microorganism activity and lower the separation rate [16].

Effective and economic treatments of effluents containing such recalcitrant pollutants have become an obligation to respect reglementary requirements which became more difficult to reach. Until now, there is no single and economically attractive treatment method that can effectively treat textile wastewaters, either the proposed traditional and advanced chemical methods which are costly and not always reliable in justifying an interest in depollution [1,2,17]. That is why new technologies including electrical, sonic, magnetic, optical, and thermal methods have been investigated by scientists as complementary treatments to the biological degradation process [17,18].

Hydrocyclone technology is a new alternative to separate phases which has several advantages as suitability for continuous operations, low capital costs of acquisition and installation, easy and rapid in particles separation, low maintenance, high separation efficiency, as well as high operational reliability and minimal space occupied [19–21].

The classic hydrocyclones consist of two main parts, a cylindrical part which is connected to a conical part (Fig. 1). For the first main part, on its side face an inlet through which the feed enters tangentially. In this part, an outlet is located vertically on top of the cylinder which extends within the cylinder and called a vortex finder. The second part is connected to the cylindrical section on the top and the underflow from the bottom end is called spigot [22].

In the hydrocyclone, the phases separation is due to centrifugation forces exerted by vortex carry. Consequently, the fine particles move to the central axis of the cyclone and will be carried out by the overflow stream. The larger particles addressed to the cyclone wall, they are discharged by underflow orifice [23]. In fact, the efficiency of separation in the hydrocyclone depends on various factors such as feed parameters such as particle size distribution, density fraction, and slurry density or geometrical parameters of model [24,25]. Thus, the hydrocyclone uses the static energy of the fluid (i.e. water flow or pressure) to create a vortex that is essential in the separation process [26]. According to the above principles, the hydrocyclone can separate, classify, and concentrate particles efficiently in the number of applications [27]. In other terms, the use of hydrocyclone could avoid all problems related to the phase separation either after a biological treatment or an adsorption process.

Adsorption treatment systems using granular activated carbon have also been tested for the treatment of textile wastewater, especially those issued from dyeing factories. In fact, adsorption is efficient to detoxify, deodorize, purify, remove color and to recover the harmful products from liquid solutions [28–30]. For this reason, many industry fields use the adsorption technique to treat wastewaters. This is the case of food and pharmaceutical industries, drinking water, and industrial wastewater. Adsorption applications on a large scale were seen as having more advantages than other separation methods because of their convenience, easy operation, simplicity of design, high efficiency, and also for their wider applicability in colourful wastewaters [31–36].

The efficiency of adsorption processes mainly depends on physical and chemical factors such as the physical and chemical characteristics of the adsorbate (molecular weight and size, functional groups, polarity, solubility, surface area, pore size distribution, and surface chemistry) and the condition of the background solution (pH, temperature, presence of competitive solutes, and ionic strength) [29,33,36–38].

Several researches have shown that activated carbon adsorption is one of the best technologies used in wastewater treatment [39]. In fact, it has been efficiently used in several pollution control systems due to its high adsorption capacity. Activated carbon can be found in the form of granular or powdered forms. The pore size of the activated carbon defines their moval efficiency [40]. The particles of powdered activated carbon, they are a pulverized form with a size less than 0.15 mm and for granular activated carbon have irregular shapes with the available commercial sizes ranging from 0.5 to 2.5 mm, [41–44]. Although all the cited advantages, few industrial applications of adsorption treatment in the textile field were noted. The adsorption process was highly limited by the further formation of sludges and other difficulties related to the phase separation after treatment, making necessary the combination of activated carbon adsorption with an efficient separation phase treatment.

In this present study, the advantage of the coupling between the phase separation by hydrocyclone and adsorption of dye by activated carbon was studied. Such coupling could ensure an enhancement of the depollution efficiency by making more effective both the adsorption and the

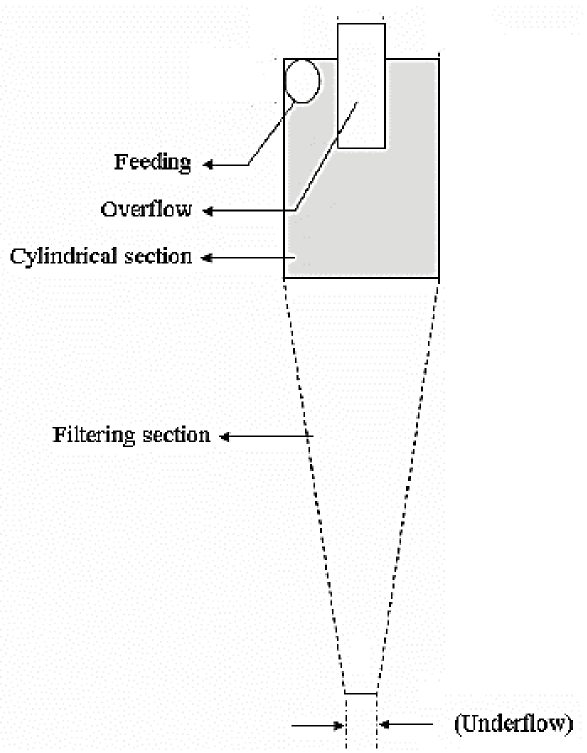

Fig. 1. Schematic drawingof a hydrocyclone.

separation steps. In fact, the recirculation of the adsorbent in a closed circuit may be increased the probability of the colorant contact with the particle surface of active carbon. On the other hand, We recover the solid particles of the adsorber, after saturation, through the underflow of hydrocyclone for the regeneration.

The overall objective of this study is to conduct a systematic experimental investigation to examine the efficiency of a combined system composed by adsorption on activated carbon and phase separation by hydrocyclone for the treatment of industrial textile wastewater. This research examines also the relationship between the effect of the size of the activated carbon particles, their charge in the medium and the feed rate in hydrocyclone, for enhanced separations. For this purpose, the Box–Behnken factorial design combined with response surface methodology (RSM) will be explored in the medialization and optimization steps in order to ensure a better improvement of particle separation efficiency.

2. Materials and methods

2.1. Chemicals: adsorbent and dye

For the study of phase separation by hydrocyclone, we used a granular activated carbon supplied from the Textile Industrial Company (SITEX). The average diameter varied between 0.5 and 3 mm and the density was about 830 kg/m^3. The technical characteristics of the activated carbon are shown in Table 1.

Table 1
Technical properties of activated carbon

Parameter	Value
Density	0.835
Pore volume (cm^3/g)	0.34
Pore diameters (Å)	14
BET surface area (m^2/g)	885

The reactive dye used for this study is NB4R (IUPAC name: tetrasodium 1,2-bis(4-fluoro-6-[5-(1-amino-2-sulfonatoanthraquinone-4-ylamino)-2,4,6-trimethyl-3-sulfonatophenylamino]-1,3,5-triazin-2-ylamino)ethane; purity 70%–80%; MW: 1,401.202 g/mol). The dyestuff is also supplied by the Textile Industrial Company (SITEX), from Merck (Germany). It was of a technical grade and used as received.

2.2. Experimental setup

Fig. 2 describes in detail the experimental setup including hydrocyclone separator, pressure gauge, centrifugation pump, mixer, and tank. We used an industrial centrifugal pump (P) to pump the liquid with granular activated carbon to the cyclone. Between the tank and the centrifugal pump, there are two gate valves (V1 and V2). And through these valves, we can carry out the adjustment of the volumetric flow rate by regulating the inlet velocity. The flow-through apex and split chamber can be controlled by the valve (V3) and the valve (V4) for emptying the pulp charge. An agitator is also employed in order to homogenize the mixture and to reduce or eliminate the adverse effects of vortices.

The tank contains the pulp charge which feeds the pump for the hydrocyclone. Its capacity is about 500 L, thus allowing the pulp to be handled without fear of spillage; for example, when adding water to reduce the solid percentage while the system is in operation. A helix is inserted into the tank to reduce the production of a vortex and ensure that a homogeneous mixture is maintained throughout the tests.

The dye solutions were prepared by dissolving the proper mass of NB4R in distilled water in the tank. The initial dye concentration of dye of NB4R solution was varied from 62.5, 95, and 127.33 mg/L which corresponds to an absorbance of 2, 3, and 4, respectively a.u. at 595 nm. These concentrations were mixed with various quantities (5, 10, and 15 g/L) of activated carbon in the tank for 2 h at ambient temperature. Then, for each trial, we turn on the system for 10 min in order to allow homogenization of the feed pulp and to reach an equilibrium regime between the operation of pump and that of hydrocyclone.

The equilibrium regime can be verified by observing the stability of the pressure and the feed rate. The targeted feed rate is easily achieved by adjusting the frequency of the pump motor. The equilibrium regime can be verified by observing the stability of the pressure and the feed rate.

Two boxes that will allow collecting the overflow and underflow, result in taking a sample of these flows individually. Then, the two streams are combined and routed to a third box where the mixture is considered as a replenishment of the feed. Fig. 2 shows the sampling points of the experimental set-up.

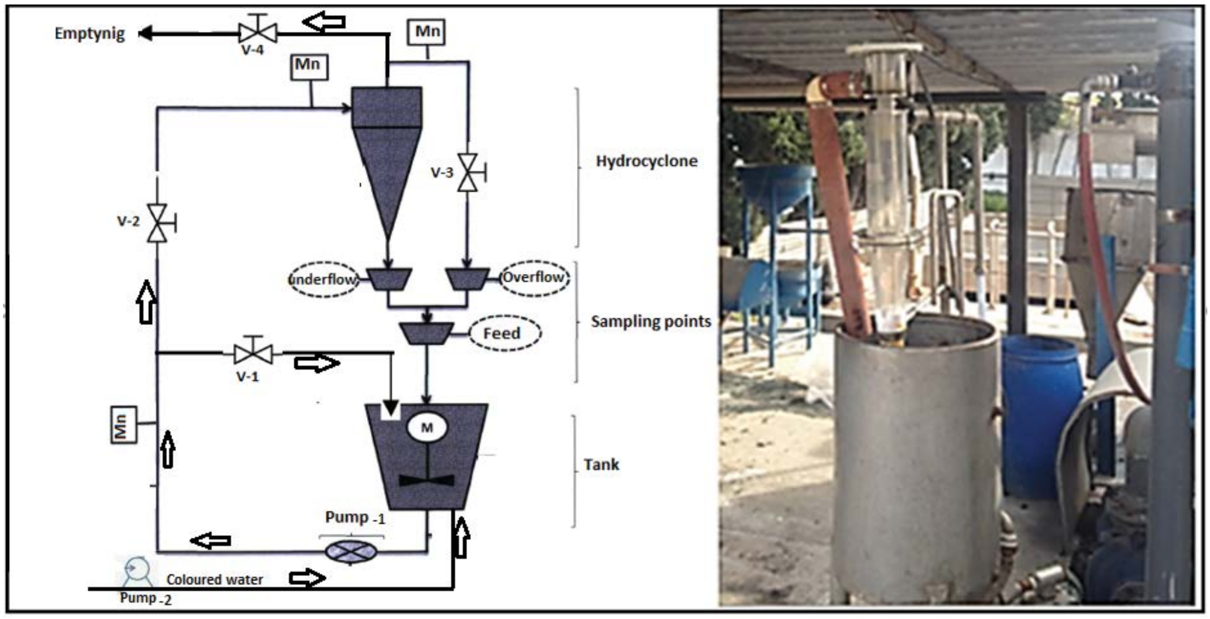

Fig. 2. Hydrocyclone experimental setup (Mn: pressure gauge; Pump_1 and 2: Centrifugation pump; Vi: valve; M: Mixer).

When the operation of the system is considered stationary, samples are taken from each box (feed, overflow, and overflow flow) with a volume of about 200 mL. The process is repeated for 3 times with an interval of 2 min. The samples are filtered using a filter paper with a porosity (7 μm). Then, the recovered particles are sent to the oven to be dried at 105°C. The dried samples are weighed to calculate the separation efficiency (%) in relation to the flow rate. For the filtrate obtained from the overflow, it is tested to calculate the efficiency of decoration by activated carbon.

The operating mechanism of this separation system can be explained as follows. First, the pump_2 is running to supply the storage tank, which contains the carbon particles, with a volume of 500 L of colored water. Then, activate pump_1 and make stop pump_2, while closing valve V-2 and opening valve V-1. At this level, the wastewater is in contact with the carbon particles in a closed circuit for a period of 1 h. It's the time needed to complete the adsorption of the color by the carbon particles. Following, inversely were V-2 open and V-1 closed for circulating the mixture in the hydrocyclone, for 10 min, until the hydrodynamic stabilization inside the hydrocyclone. Finally, to recover the treated water, the valve V-3 must be closed and the valve V-4 will be opened. The carbon particles will be collected in the storage tank by the overflow.

2.3. Geometry and dimensioning of hydrocyclone

The hydrocyclone setup used in this study was made by plexiglass (Fig. 3 and Table 2I) with the following dimensions all in mm. The inlet was the tangential and rectangular type with a width (b) of 18 mm and a height (a) of 45 mm. The diameters of overflow, underflow, and hydrocyclone were 50, 15, and 100 mm, respectively. The length of the cylindrical part was 350 mm, while the length of the cone part was 465 mm.

2.4. Experiment conditions and methodology

The investigation of all experimental parameters by the use of Box–Behnken design is very important in the case of interacted experimental variables such as the case of the proposed process in this study. So, the RSM was employed to optimize the operations variables of hydrocyclone such as feed solid ratio, flow rate to the hydrocyclone, and particle size of activated carbon.

After the preliminary study, volumetric flow rate (l/min), adsorbent concentrations (g/L), and the particle size of activated carbon (mm) were selected as the three main controllable variables (Table 3). The separation efficiency, expressed as a percentage (ES%), will be the response. We used the RSM with the "Box–Behnken" experimental design to evaluate the individual and interactive effects of variables on the cited response. Results were analyzed using the software MINITAB v 18.0. The behavior of the process was clarified by an empirical second-order polynomial model equation.

2.5. Separation efficiency calculation

Granular activated carbon with an average diameter between 0.5 and 3 mm and a density of about 830 kg/m^3 were used at the investigational testing. Suspension samples can be withdrawn at the inlet, at the underflow, and at the overflow to determine the grade efficiency separation efficiency by the use of Eq. (1) [45]:

$$\%\mathrm{ES}(x) = \frac{(W_a - W_0)}{W_a} \times 100 \tag{1}$$

where $W_a = \mathrm{Dv}_a[\mathrm{Char}]_a P_a(x)$; $W_0 = \mathrm{Dv}_0[\mathrm{Char}]_0 P_F(x)$; %ES ($X$) is the separation efficiency by weight at particle size (%); W_a is the AC mass flow rates of the feed by weight at particle

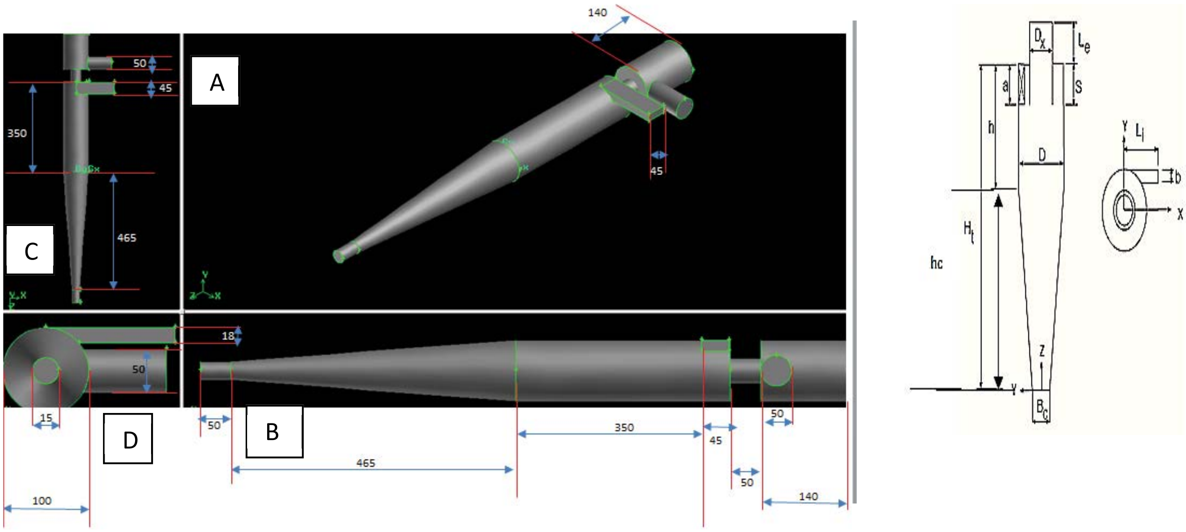

Fig. 3. Three-dimensional of a hydrocyclone for the activated carbon separation in water purifying ([A]: View according to the antero-posterior axis; [B] and [C]: View according to the longitudinal axis; [D]: transversal axis; [E]: View according to the cut plane (o,x,z)).

Table 2
Technical specifications of the cyclone

Components of the cyclone	Dimensions (mm)
Diameters of hydrocyclone (D)	100
Inlet orifice: ($a \times b$)	$a = 18$; $b = 45$
Cylindrical length: (h)	350
Length of the cone part: (hc)	465
Total length: (Ht)	815
Overflow orifice: (Dx)	50
Length of the vortex: [(S) + (Le)]	S = 75; Le = 50
Under flow orifice: (Bc)	15

size (kg/s); W_0 is the AC mass flow rates of the overflow by weight at particle size (kg/s).

In the tank, the dye solutions were prepared by dissolving the proper mass of Novacron Blue 4R (NB4R) (62.5 mg/L) in distilled water and mixed with the granular activated carbon. So, the solution was mixed with the adsorbent in a beaker at ambient temperature with a NaCl concentration of 3.5 mg/L. The pH (370 pH meter Jenway) was fixed at 11. After that, samples were removed and filtered through a 0.45 µm filter (Whatman® Glass microfiber filters, Grade GF/C, from Sigma-Aldrich). All experiments were replicated with an experimental error of 3%. All chemicals used were supplied from Sigma-Aldrich and used as received.

2.6. Decolorization measurements

In order to estimate the effectiveness of activated carbon in the decolorization of the described wastewater, absorbency before and after treatment was measured by a HACH Lange DR 3900 spectrophotometer, under a wavelength of 595 nm, which corresponds to the absorbency wavelength of the selected dye. The color removal efficiency (CR%) was calculated as follows [46]:

$$\text{CR }\% = \frac{\left(\text{Abs}\left(t_0\right) - \text{Abs}\left(t_1\right)\right)}{\text{Abs}\left(t_0\right)} \times 100 \tag{2}$$

where Abs at 595 nm (t_0): measuring the absorbance at 595 nm before treatment; Abs at 595 nm (t_{1h}): measuring the absorbance at 595 nm after 1 h of adsorption.

2.7. Statistical analyses

The SRM employed in this study was followed by mathematical analyses, using the Minitab software v 18.0, in order to establish optimum conditions for the color removal analyses included adequacy of the model (ES%), analysis of variance (ANOVA) with a confidence level of 95% ($P < 0.05$), correlation coefficient R^2 which measures the fitness of regression model, residual plots, response surface plotting, analysis of the main effects plot, and interactions plot.

3. Results and discussion

3.1. Response regression equation

Multiple regression coefficients of a second-order polynomial model have shown the separation efficiency by hydrocyclone of the particle size of activated carbon (mm). The regression model equation was as follows:

$$\begin{aligned}\text{ES}(\%) &= 101.79 + 3.33\left(\left[\text{Char}\right]\right) - 0.025\left(\text{Dv}\right) - 63.15\left(T\right) \\ &- 0.1\left(\left[\text{Char}\right]^2\right) - 0.005\left(\text{Dv}^2\right) + 8.94\left(T^2\right) - 0.001\left(\left[\text{Char}\right] \times \text{Dv}\right) \\ &- 1.69\left(\left[\text{Char}\right] \times T\right) - 0.48\left(\text{Dv} \times T\right)\end{aligned} \tag{3}$$

$R^2 = 87.5\%$

where ES (%) is the separation efficiency (%); Dv is the flow rates of the feed (L/min); *T* is the particle size of activated carbon (mm); [Char] is the concentration of activated carbon (g/L).

The regression equation of the response (ES%) and the variance analyses (ANOVA) are presented in Table 4, also, given the squared multiple correlation coefficient R^2 is equal to 87.50% with high accuracy >0.8. We can assume that the model may be predictable and can allow a good correlation between independent variables and response. Furthermore, we can deduce from the equation that the coefficients of particle size (63.147) and concentration (3.325) of absorbance value are the most important terms affecting the response value of separation efficiency (ES%).

3.2. Variance analyses

Table 4 provides a summary of the results of the ANOVA for the separation by hydrocyclone. The statistical significance of mean square variation ratio, due to regression, mean square residual error and the significance and adequacy of the model was tested using ANOVA.

An ANOVA shows that the sum of squares (SS) related to residual error is very low as compared to the total sum of squares for the model incorporating the response (1,388.8 << 11,099.2). The analysis of variance shows that the *P*-value is less than 0.05 ($\alpha = 0.05$ or 95% confidence) for regression and linear coefficients. On the other side, the *F*-value for the regression model equation was 15.54. Furthermore, the regression adjusted average squares and the linear regression adjusted average squares were 12.58 and 7.40, respectively for the response.

These parameters show that quadratic RSM models can navigate the design space well and the model accuracies are adequate to predict the performance of the separation process by hydrocyclone.

3.3. Residual plot

The normal probability diagram (Fig. 4a) shows that the points form a straight line that follows Henry's line. Therefore, it can be deduced that the residues are normally distributed. A simple check of the reliability of the model requires an observation of the plot of the residuals against the fitted values. This diagram (Fig. 4b) represents a random pattern of residues on both sides of zero (0) (Fig. 4b). According to these plots, the residuals of response (ES%) model has been randomly distributed. The histogram of the residuals in Fig. 4c shows a symmetrical distribution of residuals with respect to the order of the data. The histogram of residuals in Fig. 4c shows an almost symmetrical distribution of the residuals with respect to the order of the data. Fig. 4d shows that the residual appears to be randomly scattered about zero. These results indicate that the regression terms are correlated with each other.

3.4. Contour plot

Fig. 5a shows the relationship between the residual response ES% and the two variables (volume flow rate and carbon concentration). From the figure, it seems that at a fixed flow rate and particle size at 1.75 mm, the separation efficiency ES% decreases from 50% to 34.36% by varying the carbon concentration from 5 to 15 g/L. However, at a fixed coal concentration and flow rate varying from 81.83 to 163.67 L/min, the ES% increases from 44.47% to 53.17%.

Fig. 5b gives the relationship between the response and the two variables (concentration and particle size of the carbon). This figure clearly shows that at a fixed concentration of the coal and by varying the size of its particles from 0.5 to 3 mm, while keeping the flow rate at 122.75 L/min, a decrease of the ES% was noted from 80% to 28.61%. Conversely, by fixing the size of the carbon particles and by varying their concentration from 5 to 15 g/L, the ES% decreases from 65% to 28.58%. The concentration of charcoal showed a negative effect. We can assume that this decrease is due to the fact that the increase in the carbon dose creates shocks between the particles. Therefore, it induces their breakage and their movement towards the underflow. Consequently, the ES% decreases. According to the results in Plitt et al. and Neesseet al. [51,52] showed that the increased concentration of this operational variable reduces the performance of the hydrocyclone separation.

Regarding Fig. 5c, which describes the relationship between the response and the two variables (the size of the particles and the flow rate), it can be seen that at a carbon concentration of 10 g/L, and by varying the size of the particles from 0.5 to 3 mm, while keeping the flow rate fixed, the separation efficiency is reduced from 87.32% to 28.32%. The increase in the size of particle for the same concentration leads to a partial or total blockage of the overflow, which conducts to the sending of the feed pulp of the hydrocyclone directly to the overflow [53]. For that purpose, we can explain the decrease in the ES%.

3.5. Analysis of main effect plot

Fig. 6 shows the main effects of volume flow, carbon particle size, and carbon concentration on the separation efficiency by the hydrocyclone.

Table 3
Experimental range and levels of independent process variables

Code levels	Concentration of activated carbon (g/L) [Char]	Particle size of activated carbon (mm) (*T*)	Volumetric flow (L/min) [Dv]
−1	5	0.50	81.835
0	10	1.75	122.753
1	15	3	163.670

Table 4
Analysis of variance (ANOVA) of the model of the separation efficiency (ES%)

	Source	Regression	Linear	Square	Interaction	Residual error	Total
ES(%)	DL	9	3	3	3	20	29
	SS	9,710.51	2,555.44	1,632.90	2,687.22	1,388.76	11,099.21
	MS	1,078.943	851.816	544.297	895.744	69.438	
	F	15.54	12.27	7.84	12.90		
	P	0.000	0.000	0.001	0.000		

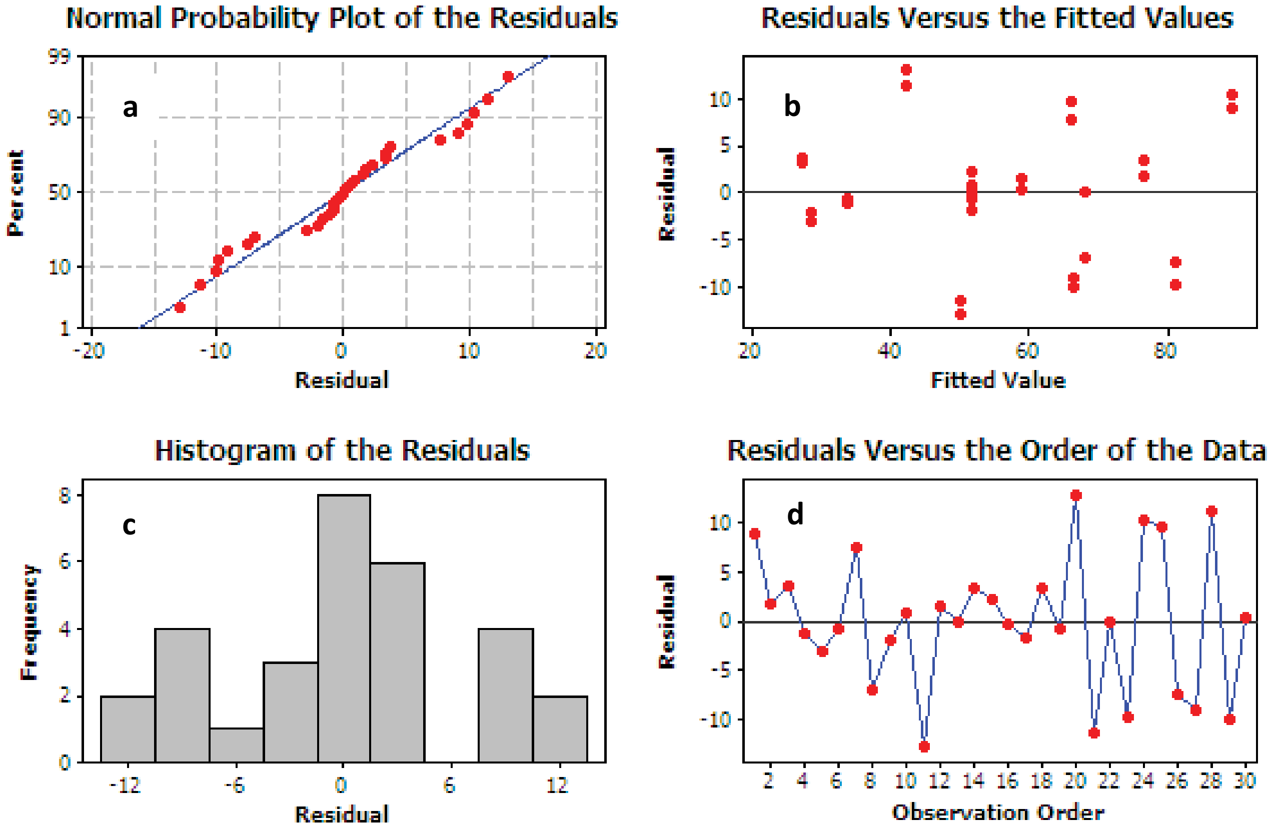

Fig. 4. Residual plots for response separation efficiency (ES%) by hydrocyclone of particle size of activated carbon.

The concentration of carbon has a major negative effect on the performance of the separation. Indeed, the efficiency separation ES% decreases considerably from 64% to 46% by increasing the concentration of charcoal from 5 to 15 g/L. The flow has a primary positive effect on the separation performance. Indeed, the ES% increases slightly from 50% to 57% by varying the flow rate from 81.83 to 163.67 L/min. Regarding the effect of particle size, we noted that if we vary the size from 0.5 to 3 mm, the ES% undergoes a strong variation from 79% to 48%. This implies that the particle size plays a major negative effect on the response.

3.6. Interaction plot

From the interaction plot, we can note an interaction when the modification in the response from high to low levels of a parameter is dependent on the level of a second parameter. Graphically, an interaction is effective when the lines are approximately not parallel [47].

The interaction effects of each factor on the separation efficiency are presented in Fig. 7. The figure shows that when the size increases from 0.5 to 3 mm, the ES% varies from 64% at a flow rate of 81.83 L/min and by 10% at a flow rate of 163.67 L/min. This could be explained by the interaction between these two parameters (size-flow rates). There is also an interaction between particle size and carbon concentration. Indeed, by varying the concentration from 5 to 15 g/L, the ES% undergoes a variation of 15% when the particle size is 0.5 mm, and of 47% when the particle size is 3 mm. Another interaction is observed between the flow and the concentration of the carbon. In fact, when increasing the concentration from 5 to 15 g/L,

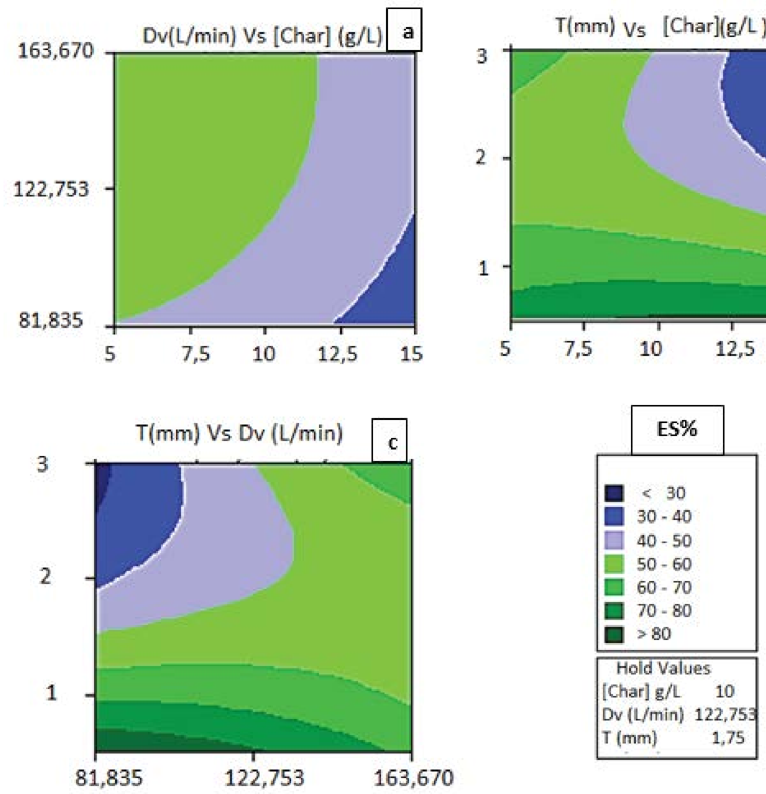

Fig. 5. Contour plots of predicted separation efficiency (%) by hydrocyclone as a function of flow rates of the feed (Dv, L/min), particle size of activated carbon (*T*, mm), and concentration of adsorbent ([Char], g/L).

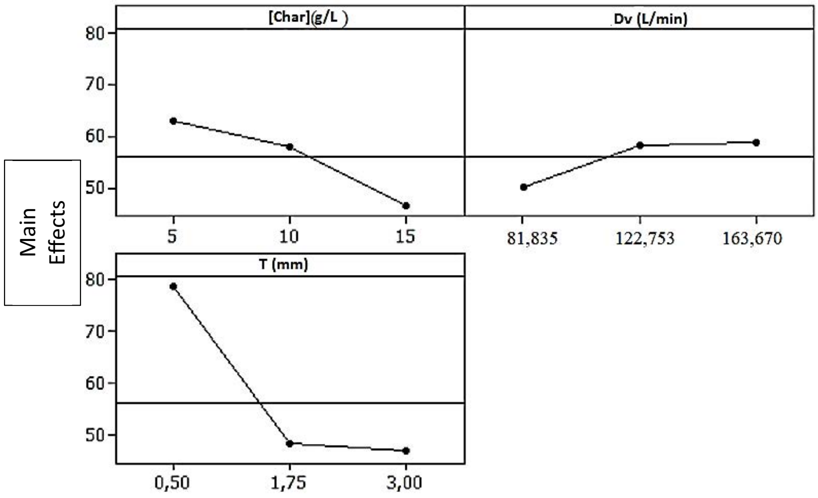

Fig. 6. Analysis of main effects plots of volume flow, carbon particle size, and carbon concentration on the separation efficiency by the hydrocyclone.

the ES% varies from 29% at a flow rate of 122.75 L/min and from 4% at a flow rate of 81.83 L/min. This difference in response variation proves the existence of this interaction.

3.7. Optimized conditions

In order to determine the optimal conditions in the proposed hybrid process that ensure the maximum color removal (CR%) and separation efficiency (SE%) by the hydrocyclone of activated carbon, the desired function methodology optimization of Derringer and Suich was used in this present study [58]. The Minitab.v.18.0. software has been used in order to find the specific point that maximizes the desirability function, for numerical optimization.

The performance of all design and response variables are shown in Fig. 8. and Table 5, where the best optimization

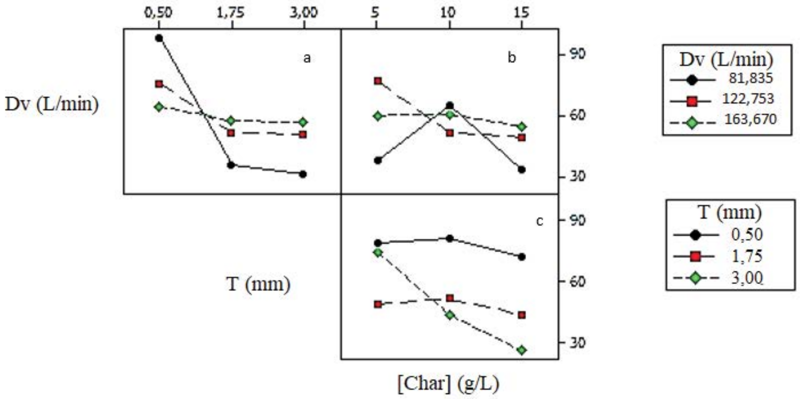

Fig. 7. Analysis of interaction plots of the separation efficiency by the hydrocyclone of activated carbon.

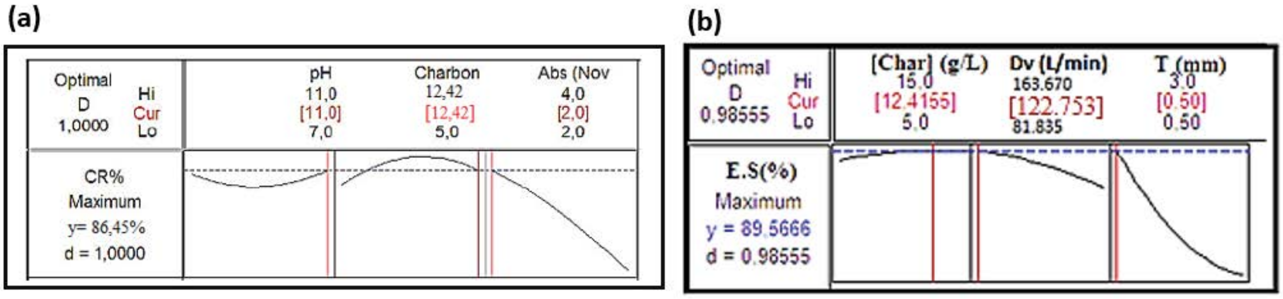

Fig. 8. Optimization of Novacron Blue 4R (NB4R) removal by adsorption (a) and separation efficiency by the hydrocyclone (b) process on activated carbon.

is approached by an overall desirability of 1 at optimum operating conditions.

For the determination of optimum process parameters for the maximum residual dye (Novacron Blue 4R (NB4R) removal) (CR%) by adsorption process on activated carbon, the performance of all design and response variables are shown in Fig. 8a and Table 5. The best optimization is approached by an overall desirability of 0.98555 at optimum operating conditions of pH at 11, carbon concentration at 12.42 g/L, and initial color absorbance at 2 a.u (corresponding to 62.5 mg/L). In these conditions, the Minitab Software gives a response value for CR% of 86.45%. When these conditions were applied experimentally, we reached a CR% = 87.15.84%. Thus, when the experimental conditions proposed by Minitab were applied to our experimentation, the values of the responses obtained are almost equal to the values obtained by calculating. Hence, the model proposed in this study is globally validated but could be more improved.

Fig. 6b suggests that the particle size of adsorbent should be kept as low as possible (0.5 mm), as ES% increases monotonically with decreasing the adsorbent size. On the contrary, the flow rate and carbon concentration should be kept moderate to optimize the response ES%. The optimal values of the first and second factors are 122.753 L/min and 12.42 g/L, respectively. Separation efficiency by the hydrocyclone at these optimal conditions is ES% = 89.57% with a desirability *d* = 0.9855. Experimentally, using the same conditions created by Minitab software, we obtained ES% = 88.74% (Table 5). Since the experimental and the predicted values are close, we could conclude that our model is globally validated.

Table 5
Comparison between experimental and predicted values using the Box–Behnken design

	Predicted response	Experimental values
Global solution	pH = 11 Do(Nov) = 2 a.u; [Nov] = 62.5 mg/L; Carbon = 12.4155 g/L	pH = 11 Do(Nov) = 2 a.u; [Nov] = 62.5 mg/L; Carbon = 12.4155 g/L
Response CR%	CR% = 86.45% Desirability = 1	CR% = 87.15%
Global solution	Débit volumique (81.83 L/min) Carbon = 12.4155 g/L Taille = 0.5 mm	Débit volumique (81.83 L/min) Carbon = 12.4155 g/L Taille = 0.5 mm
Response ES (%)	ES% = 89.55% Desirability = 0.985	$ES\%_{exp}$ = 88.74% ± 0.24

ANOVA is performed on the separation efficiency between the practical RSM and experimental results gives a *p*-value of 0.04. These results mean that we can conclude with 96% confidence that the difference is significant between the experimental and theoretical values. The confidence level is high because of the significant difference. This can be explained by the instantaneous anomalies marked by the hydrocyclone during the samples taken.

4. Discussion

All the cited results showed that the flow rate, concentration, and particle size of the adsorbent do not affect the response (separation efficiency) in the same way. The size parameter followed by the concentration of the adsorbent parameter appears to be the most influential factor in the response.

The variation in the levels of each factor shows variable profiles with respect to the performance of the treatment. The particle size of the adsorbent shows a negative effect. In fact, particles are subjected to two opposing forces namely, the centrifugal force which guides the large particles towards the overflow and the driving force directed towards the center of the water and the small particles where the air column vacuating them to the overflow [48].

It is, therefore, evident that the particles of large sizes are heading underflow. But in our case, we found that the ES% showed a drop as the particle size increased. We can explain this by the fact that the bigger size of the particles causes collisions between them and, consequently, a decrease in their size. The driving force brings the fine particles to the overflow resulting in the decrease of the ES% [49]. Indeed, Cullivan et al. [48] have proved that the performance of the separation increased proportionally with the size of adsorbent. As for the flow, we found that this parameter gives a positive effect. This can be explained by the fact that the created centrifugal force, when increasing, improves the performance of separation. This positive effect of the hydrocyclone has also been proven by Mukherjee et al. [50].

However, the concentration of activated carbon parameters showed a negative effect. Such phenomenon was related to the fact that the increase in the carbon dose creates shocks between the particles and induces their breakage and their direction to the overflow inducing the fall of the ES%. In the same way, the investigations of Plitt et al. and Neesse et al. [51,52] have made it possible to prove the impact of the increase in the concentration of this operational variable on the performance of the separation of the hydrocyclone.

Concerning the efficiency of the hydrocyclone under the influence of inlet flowrate, Bradley [59] reported that, when the flowrate increases from 1.17×10^{-5} to 3.1×10^{-5} m^3/s, the separation efficiency, for particles of about 6 μm was increased from 10% to 47%. Moreover, the suspended solids with different sizes can be more clearly separated. In any case, the cut-size value and separation efficiency were dependent on the feed flowrate.

A previous study carried out by Puprasert et al. [45] into the separation efficiency by hydrocyclone of the micro sand particle (d_{50} = 5 μm), with different raw water concentrations. Results showed that, for the same value of inlet pressure (4 bars), the removal efficiency enhanced from 38% to 57% when the water concentration decreased from 3 to 1 g/L.

It should also be noted that the variation of operating conditions of the hydrocyclone could explain the abnormal functioning of the separation process. This can cause a partial or total blockage of the overflow nozzles, leading to the sending of the hydrocyclone feed slurry directly to the overflow [53]. This could explain the decrease in the ES% in the different cases.

We also noted several interactions between the explored factors. The most important interaction was between the size and the concentration of adsorbent where it shows a synergy of action. For a given concentration, the increase in the size of the particles induces a decrease in the response under the effect of the increase in shocks between these particles. By increasing the concentration of adsorbent, the probability of having a variation in particle sizes increases, which leads to an increase in the probability of having collisions between these particles and, therefore, causes a decrease in the response.

The particle size of the adsorbent and the flow rate showed antagonistic effects. The interaction between these two parameters has been explained in many researches [54,48], by proving that the injection rate of the feed pulp acted on the separation process created in the hydrocyclone. Indeed, this separation is due to the centrifugal and driving forces applied on the particles, which act on their movements in the hydrocyclone, towards the overflow. These forces depend strongly on the characteristics of the particles such as size and density. This explains why the increase in flow favors the centrifugal force which directs the particles towards the overhead part explaining the increase in the ES%, but it induces shocks between the particles of the carbon thus limiting the performance of the separation.

Similar findings were also reported by Hwang et al. [60], who investigated in their study the separation of micro-sand particles by hydrocyclone. It was concluded that there is an interaction between the diameter of particle and the feed flowrate. The effect of the feed flowrate was noted as high when the particle size distribution between 2.5 and 5 μm. In fact, when the flowrate increases from 6.96 to 9.75 m/s, separation efficiency increases from 27% to 42% at the particulate size of 2.5 μm and from 55% to 87% at a particulate size of 5 μm. However, the effect of flowrate could be considered as negligible when the diameter of particle, beyond the limits of the interval 2.5 to 5 μm.

The injection flow rate and the carbon concentration show two antagonistic actions. In fact, for a fixed concentration, increasing the flow improves the response under the effect of centrifugal force which promotes separation. But when you increase the concentration, the number of particles also increases for the same particle size. Any increase in the flow rate may improve separation but may also increase the risk of particles impacting against the hydrocyclone wall.

The ES% optimization study shows that for a better separation efficiency, the hydrocyclone requires a relatively low flow rate, a high concentration of carbon and a smaller particle size (Fig. 8). Under these optimal conditions, the process ensures an experimental separation efficiency of 88.74%. The previous study conducted by Mukherjee et al. [49] on the separation of an adsorbent by the hydrocyclone showed that the maximum separation efficiency is 93% while Otto et al.'s [55] study showed that the efficiency of the separation of activated carbon does not exceed 79.5%. In fact, the variations in the separation efficiency do not only depend on the operational parameters [56,52] but also the optimization of the geometric parameters of the hydrocyclone [56,57]. We can then conclude that the improvement of the ES% requires a modification of the geometric variables of the used hydrocyclone.

5. Conclusion

The main objective of this study was to create a new hybrid process for the treatment of textile wastewater highly charged by residual dyestuff. The proposed process is a combined system between adsorption treatment using activated carbon and a phase separation using hydrocyclone for the recovery of the particles of the adsorbent. The originality of this work is manifested by the application of conventional methods gathered in a well-determined chronological and logical order, taking the advantage of each phase of treatment in the favor of the other step which follows it, in order to solve the technical problems and improve the final wastewater quality. Results showed that the proposed coupling between the treatment of Novacron dye (NB4R) by activated carbon and the separation of the solid–liquid phases by the hydrocyclone ensured a high level of color removal. A higher level of decolorization was reached (CR (Nov)% = 87.15%) in optimal conditions which were particle size at 0.5 mm, the pH at 11, and the carbon concentration at 12.42 g/L.

After adsorption, the hydrocyclone-integrated system ensured better recuperation of carbon particles so that they do not disturb the progress of the subsequent treatment steps. The exploration of the hydrocyclone separation system parameters and its interactions with variables of the adsorption system showed also that the particle size of the activated carbon was the most influencing factor on the processing and that the performance of the separation improved by decreasing the size of these particles for this domain study [0.5–3]. The better separation efficiency noted under optimal conditions was ES% = 88.74%. During the adsorption phase with activated charcoal, statistical analysis showed that the concentration of the adsorbent was the most influencing factor in the response with a positive effect on the decolorization process when we fixed the level of pH at 11. On the other hand, setting the particle size at 0.5 mm, the pH at 11, and the carbon concentration at 12.42 g/L, the optimal discoloration noted at the cited conditions was CR (Nov)% = 87.15%.

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Nomenclature

$[\text{Char}]_a$ — Feed concentration, g/L
$[\text{Char}]_0$ — Overflow concentration, g/L
$P_a(x)$ — Percentage by weight at particle size = /x (mm), at feed stream
$P_0(x)$ — Percentage by weight at particle size = x (mm), at overflow stream
Dv_a — feed flowrate, L/min
Dv_0 — overflow flowrate, L/min
CR% — decolorization efficiency
AC — activated carbon
NB4R — Novacron Blue 4R
(ES%) — Separation efficiency

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