

Coagulation–flocculation–decantation of dye house effluents: concentrated effluents

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Abstract

The treatment and valorization (recycling of water and mineral salts) of dyeing effluents constitutes a major economic and environmental issue. Cotton dyeing using reactive dyes is actually the most polluting in terms of discharge. In order to treat and valorize the effluent from a dyeing process using reactive dyes, the firm Clariant and the LPPE have developed and patented a process allowing the recovery of most of the water and salts present in the effluent and also of a concentrate containing the hydrolyzed reactive dyes and the highly concentrated auxiliaries. This paper describes the coagulation–flocculation–decantation process used to optimize water recovery from the concentrate. Coagulants and flocculants are used in the industry for the treatment of effluents produced by the synthesis of dyes in general (acid, disperse, reactive). Although the settling velocity is relatively low, the results show that it is possible to recycle a great volume of water using a small decanter area, since the concentrate flow rate is low. These effluents are substantially discolored, it is possible to envisage recycling them.

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1. Introduction

Water is a scarce resource since only 0.03% of the world reserves are available for human activities. Population growth and industrial booms have given rise to an increasing need for water, while the offer has remained constant. Moreover, bad water management has contributed to definitively reduced supply. When dyeing textiles, water serves two purposes. Water vapor acts as heating agent for the bath while liquid water ensures the transmission of the color onto the fiber. Cotton is the most commonly used fiber world-wide and also the material whose preparation requires the greatest volume of water. Dyeing 1 kg of cotton requires about 150 L of water, 0.6 kg of NaCl and 50 g of reactive dyes. Over 80,000 tons of reactive dyes are produced and consumed each year. This gives us an

idea of the total pollution generated by their use. After dyeing, a strongly colored dyeing bath highly loaded in organic compounds and highly concentrated in mineral salts is discharged in the municipal wastewater treatment plant. The aim of the study carried out by the firm Clariant AGBasel (Switzerland) and the Laboratoire en Procédés Propres et Environnement (LPPE—Clean Processes and Environment Laboratory, Marseille, France) is to develop an industrial process to recycle the fresh water as well as the mineral salts used during dyeing and to retain the unfixed hydrolyzed reactive dyes and the auxiliaries: a small volume of highly concentrated liquor [1]. The water and the brine can be re-used for another dyeing process. Recycling the mineral salts used during dyeing is of great interest in view of the high consumption of salts (thousands of tons of mineral salts per year) and the unacceptably high salinity of the discharge.

The aim of this study is to examine the treatment of the ultimate waste generated by this industrial process. First, we will

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detail the main processes available for the treatment of these dyeing effluents and list their advantages and disadvantages. Then, we will present briefly the industrial process developed and patented by Clariant and the LPPE, and we will study the influence of the operating conditions on the decrease of the ultimate waste generated by this industrial process using coagulation–flocculation–decantation in order to determine the minimum area of the decanter to be installed. Due to the membrane treatment, we obtain an industrial effluent with a high concentration of hydrolyzed reactive dye and a very small quantity of mineral salt: it is the first time that the potentiality of a coagulation–flocculation–decantation process is studied for the decolorization of this type of effluents.

2. Dyeing process and treatment and valorization of dye house effluents

The short and long term prospects of the dyeing industry show that, although the general production/consumption of dyes has leveled off, the production/consumption of reactive dyes has increased to the detriment of other classes of dyes. This is due to the fact that cotton is the most widely used textile and that reactive dyes have technical economic advantages [2]. Schematically, a reactive dye takes the form of a colored molecule bearing one or several solubilizing groups (Fig. 1). On this molecule has been grafted a reactive group that will exchange a covalent bond with the cellulose. The reaction with the fiber may be either a substitution or an addition. The reactive dye is applied properly, rinsed, soaped in order to remove the non-fixed hydrolyzed reactive dye, and therefore now intimately bound by covalence to the cellu-

losic fiber. The textile industry spans a wide range of activities, from preparation of the raw material in order to obtain a thread or a fabric to ennoblement treatment. Ennoblement is the collective term for pre-treatment, dyeing, printing and finishing. All these industrial activities use a great quantity of energy and of water and are highly chemically polluting. Dyed cotton is the leading dyed fiber. More than half the production of cotton is dyed using reactive dyes, on account of their technical characteristics. Unfortunately, these dyes are the least environmentally friendly of all dyes since the effluents generated are relatively colored and highly concentrated in salts. In order to dye one hundred kilograms of cotton, the dyeing protocol usually involves eight baths of 1000 L each. Bath No. 1: bleaching, which is used for removing the natural color of the cotton. Bath No. 2: rinsing, which removes the decomposition products (paraffin, etc.). Bath No. 3: neutralizing. The cotton is bathed before dyeing at room temperature. Bath No. 4: dyeing. For the dye to penetrate the textile, it still has to be accepted by the fiber. The dyeing of cotton by reactive dyes involves two distinct phases: a substantive phase which allows the dye to “set” onto the fiber thanks to the highly concentrated mineral salts present in the bath, and a fixation phase that allows the dye absorbed by the fiber to bind covalently to the fiber thanks to the alkalis (sodium carbonate, caustic soda) added to the bath. Bath No. 5: rinsing, which eliminates the dyes that did not bind to the fiber. Bath No. 6: soaping, which is used to check that the dye is well bound onto the fiber. Bath No. 7: rinsing. Bath No. 8: softening.

There are a great number of feasibility studies concerning the treatment of dyeing effluents [3,4]. There are four different types of treatment: (i) standard treatments, (ii) electrochemical processes, (iii) ozone treatment, and (iv) membrane processes.

One of the standard treatments is biological degradation [5–7], which reproduces natural that is the processes by which an aquatic medium recovers its original quality after pollution. The coagulation–flocculation–decantation treatments [8–12] and the absorption on powder activated carbon [13–16] are usually used to eliminate organic matter. The electrochemical process [17,18] requires an electrolysis cell containing iron electrodes around which the dyeing effluent circulates. The ozone [12,19–23] attacks the double bonds responsible for the coloration. For this reason, the discoloration of an effluent by the ozone alone is not always accompanied by a significant decrease of the COD. Moreover, the implementation of an ozonation process brings about extra costs. Reverse osmosis, nanofiltration, ultrafiltration and microfiltration are examples of membrane processes. In the case of reverse osmosis [24–27], the retention of mineral salts, hydrolyzed reactive dyes and the removal of the auxiliaries can be done in one single step, and this allows us to obtain a discolored pure water. The nanofiltration membranes [28–38] separate the low molecular weight organic compounds and/or the divalent and monovalent salts. In our case, it is possible to separate the mineral salts from the hydrolyzed reactive dyes

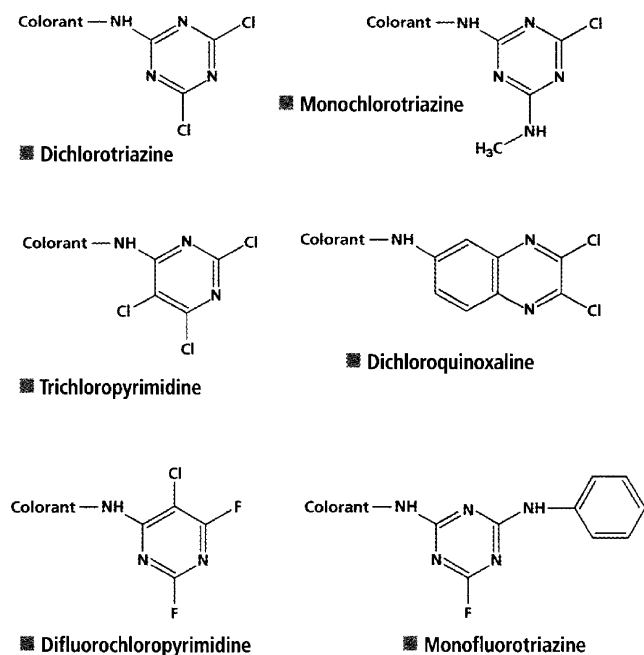


Fig. 1. Molecule structures of a reactive dye.

Table 1
Advantages and disadvantages of process

Treatment	Name	Advantages	Disadvantages
Standard	Biological degradation [5–7]	Efficiency of oxidizable matter 90%	Low biodegradability of dye The salt concentration stay constant
	Coagulation–flocculation [8–12]	Good elimination of insoluble dyes	Cost of sludge treatment Garbage dump
	Adsorption on active carbon powder [13–16]	Coupled with coagulation process: reduction of suspended matter, organic matter and low influence on color	Fast fouling of suspended matter Cost of active carbon powder
Electrochemical	[17,18]	Adaptation at different charges of pollutant and different flow rate	Formation of iron hydroxide sludge
Ozone	[12,19–23]	Good elimination of color	No diminution of COD values Extra costs
Membrane processes	Reverse osmosis [24–27]	Retention of mineral salt and hydrolyzed reactive dyes and auxiliaries	High pressure process Fouling with high concentrations
	Nanofiltration [28–38]	Separation of mineral salts, hydrolyzed reactive dyes and auxiliaries Treatment for complex solution with a high concentration of pollutant	
	Ultrafiltration [7,28,39–41]		
	Microfiltration [26,42–44]	Low pressure process	Inadequate quality for re used the permeate

and auxiliaries. Ultrafiltration [7,28,39–41] is used as a single phase treatment of the secondary wastewaters of the dye houses: the permeate has a quality allowing its re-use in minor processes (rinsing, washing) of the textile industry, but it does not have the quality necessary to be re-used in sensitive processes such as dyeing with reactive dyes. Microfiltration [26,42–44] is suitable for the treatment of dyeing baths with pigment colors. Table 1 shows the advantages and disadvantages of these various processes.

The process of treatment and valorization of dyeing effluents developed by Clariant and the LPPE aims at treating dyeing baths with a view to re-using the water and mineral salts in the dyeing process. There are four steps in this process: pre-filtration, neutralization, nanofiltration and reverse osmosis (Fig. 2) [1].

The pre-filtration aims at eliminating the small cotton fibers and other big-sized compounds that may block the membranes. The second step is the neutralization by hydrochloric acid of the alkalis present under the form of carbonate and soda to transform them into sodium chloride and carbon dioxide. It is essential to avoid the formation of bicarbonate in the water after treatment. Its presence in the recycled water would have an undesirable buffer effect during the new dyeing process, preventing the dyes from binding optimally to the fibers. The CO₂ is therefore eliminated by air bubbling. The nanofiltration separates the hydrolyzed reactive dyes and auxiliaries from the sodium chloride and water. Reverse osmosis is used to concentrate the brine if necessary and to recover the fresh water and a concentrated enough brine, which will be both sent back to the beginning of the

process in order to be re-used for the next dyeing. The performances obtained with this new process are the same as those presented in Fig. 3. A dyeing process using recycled water and brine does not differ from a dyeing process carried out with the water entering the dye house. When 85–90% of the salts as well as a great quantity of water are recycled, an ultimate waste containing hydrolyzed reactive dyes and auxiliaries with a high concentration is obtained.

The objective of the present work is to study the potential of the coagulation–flocculation–decantation process on the ultimate waste. Since the flow rate and the concentration of this ultimate waste are different (respectively, 30 times lower and 30 times higher) from those of untreated dye house effluents, the management of the sludges is different.

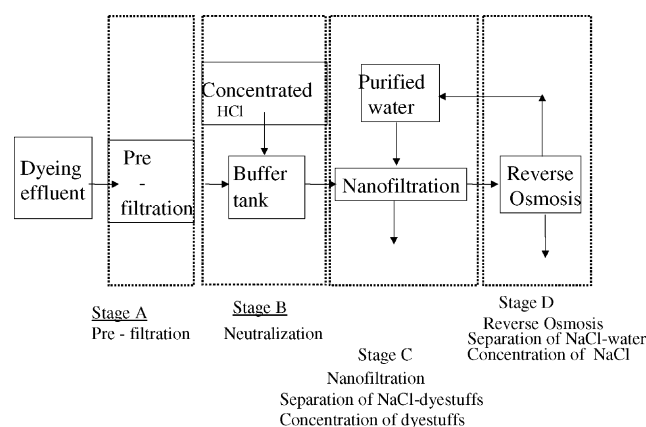


Fig. 2. Steps of patented process.

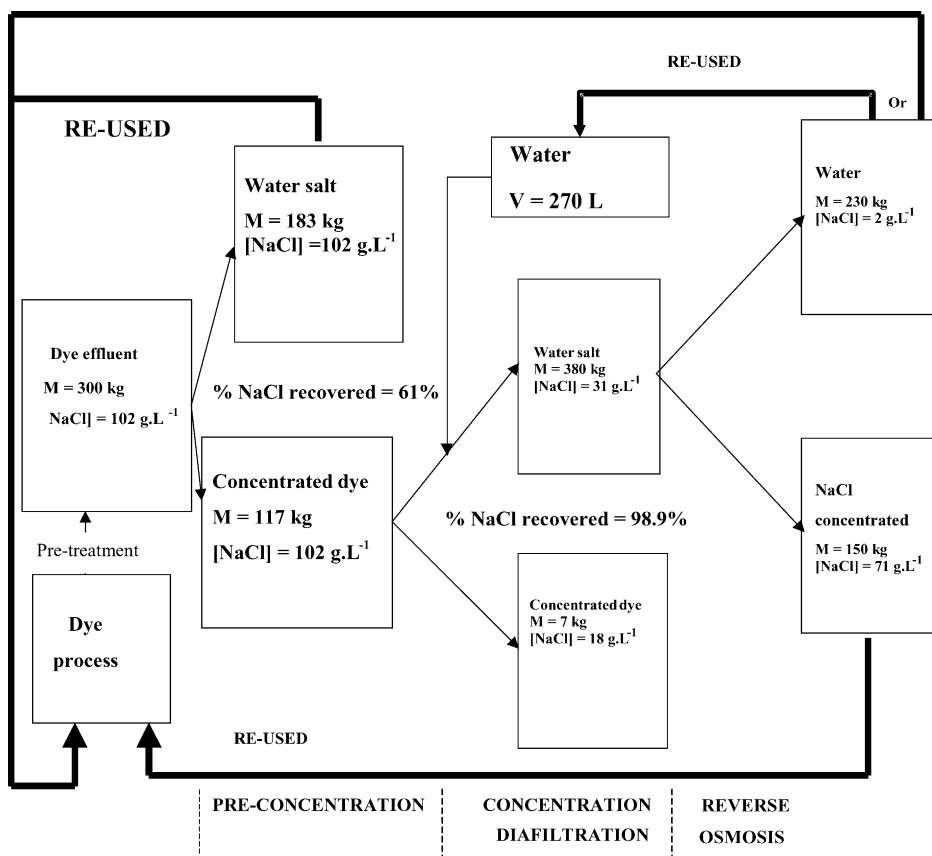


Fig. 3. Mass balance for a treatment by pre-concentration–diafiltration and concentration (dye: Drimaren HF).

3. Materials and methods

3.1. Color measurements

The color is measured by absorbance. The measurements are performed with a spectrophotometer (Thermo Spectronic, Aquamate, Rochester, USA). A graph is obtained (visible) and the absorbance of the solution is taken at the wavelength corresponding to the color of the dye considered.

3.2. Solution

Five mixtures of industrial dyeing baths from the dye house Sotrater (Troyes, France) were studied. Four of these baths contained the monochlorotriazine dyes and one bath contained reactive black 5. These tests covered almost all the reactive dyes used in the dyeing industry including all the molecular weights possible. The solutions introduced into the dyeing machines are given in Table 2. After the textile had been dyed, the effluent generated by each machine

Table 2
Characteristics and compositions of effluents

Number	Dye	Initial composition (g L ⁻¹)	NaCl (g L ⁻¹)	Na ₂ CO ₃ (g L ⁻¹)
1	Crimson ProcionHexl	0.35	45	10
	Dark blue ProcionHexl	0.006		
2	Yellow ProcionHexl	0.0007	39	13
	Dark blue ProcionHexl	0.014		
3	Yellow ProcionHexl	0.21	60	15
	Crimson ProcionHexl	0.13		
	Navy ProcionHexl	0.32		
4	Yellow ProcionHexl	0.24	78	20
	Crimson ProcionHexl	0.09		
	Navy ProcionHexl	0.27		
5	Black Saiazol	0.014	80	15

Table 3
Name and characteristic of coagulants and flocculants

Name	Manufacturer	Characteristics
Sandofix WE liq	Clariant, Switzerland	Cationic polymer, low toxicity
Eurochem Basic	Clariant, Germany	Cationic polymer, low NH ₂ groups in the structure than Sandofix WE liq so lower toxicity
Locron L	Clariant, Germany	Neutral compound with polyaluminium chloride
Flonex FI 28P4	Fleurger, France	Cationic polymer
Cartaretin 10CE (India)	Clariant, India	Cationic polymer very low toxicity
Sandotex FAR (India)	Clariant, India	Cationic polymer using without Al or Fe
FeCl ₃		Reference

was mixed with the other effluents (20 L of each effluent) to give the “Sotrater mixture”: mixture of five dyeing effluents. It then underwent the process of treatment and valorization described above. After 90–95% of the salts present in this effluent had been recovered, the hydrolyzed reactive dyes and the auxiliaries were concentrated into the ultimate waste (absorbance: 8.9; dry matter = 8.24 g L⁻¹) that was treated by coagulation–flocculation–decantation. This ultimate waste cannot actually be reduced further using membrane processes since this would cause irreversible fouling.

3.3. Experimental protocol

The experimental procedure of coagulation–flocculation–decantation was the one used in the municipal wastewater treatment plant where the effluents of the firm Sotrater are treated. Table 3 gives a list of the coagulants and flocculants used in this study. First, the efficiency of each coagulant and flocculant was assessed, according to the same operating mode for each of them. Then, the operating mode was adjusted depending on the result obtained. The pH of the ultimate waste was adjusted up to a value of 10.5/11 with soda. A known volume (Table 4) of the first coagulant was added, at a stirring speed of 200 rpm for 5 min. The solution became acid and the pH was re-adjusted to a value of 5.5/7. The flocculant was added, then the solution was stirred slowly at a speed of 20 rpm for 20 min. Once the stirring was stopped, the decantation started. The solutions were poured in 1 L graduated cylinders and the Kynch curves were drawn. Coagulation is the destabilization of the electrostatic interactions that may exist between the molecules of hydrolyzed reactive dyes or auxiliaries and the water, by addition of a chemical reagent, the coagulant. In practice, generally FeCl₃ or Locron are used. The flocculation reagents are synthetic polymers with a linear structure and a high molecular weight.

Table 4
Variation of supernatant absorbance as a function of flocculant nature

Test	A
Sandofix WE liq 0.6 mL/FeCl ₃ 7 mL	0.71
Sandotex FAR 0.15 mL/FeCl ₃ 7 mL	1.39
FeCl ₃ 7 mL	1.65
Flonex FI 28P40.3 mL/FeCl ₃ 7 mL	1.46
Eurochem Basic 0.4 mL/FeCl ₃ 7 mL	0.83
Cartaratin 10CE 0.1 mL/FeCl ₃ 7 mL	1.52

Table 5
Variation of supernatant absorbance as a function of the Sandofix WE liq quantity

Test	A
Sandofix WE liq 0.6 mL/FeCl ₃ 7 mL	0.71
Sandofix WE liq 0.8 mL/FeCl ₃ 7 mL	0.38

The flocculant must be added under slow stirring in order to avoid the breaking of the flocs during their growth. Plug settling characterizes the activated sludges and the flocculated suspensions when their concentration is higher than 1 g L⁻¹. The particles adhere to each other and settle forming an interface between the flocs and the supernatant liquid. Using to Kynch’s theory, we can calculate the minimum area of the decanter.

4. Results

In view of the results obtained with ferrous chloride, Locron L was abandoned for the rest of the tests.

4.1. Absorbance

The absorbance was measured for each supernatant obtained by addition of the coagulant FeCl₃ without any flocculant or with the various flocculants at the given concentrations. Table 4 shows that the best supernatant absorbance is obtained with Sandofix WE liq at a concentration of 0.6 mL L⁻¹ and Eurochem Basic at a concentration of 0.4 mL L⁻¹. The tests were performed with Sandofix WE liq first, since it is more economical than Eurochem Basic. The concentration in Sandofix WE liq was increased to compare the two supernatants obtained. The absorbance of the supernatant obtained with 0.6 mL of Sandofix WE liq is twice as high as that of the supernatant obtained with 0.8 mL of

Table 6
Variation of supernatant absorbance as a function of the coagulant quantity

Test	A
Sandofix WE liq 0.8 mL/FeCl ₃ 7 mL	0.38
Sandofix WE liq 0.8 mL/FeCl ₃ 5 mL	0.43
Sandofix WE liq 0.8 mL/FeCl ₃ 2 mL	0.293
Sandofix WE liq 0.8 mL/FeCl ₃ 1 mL	0.335

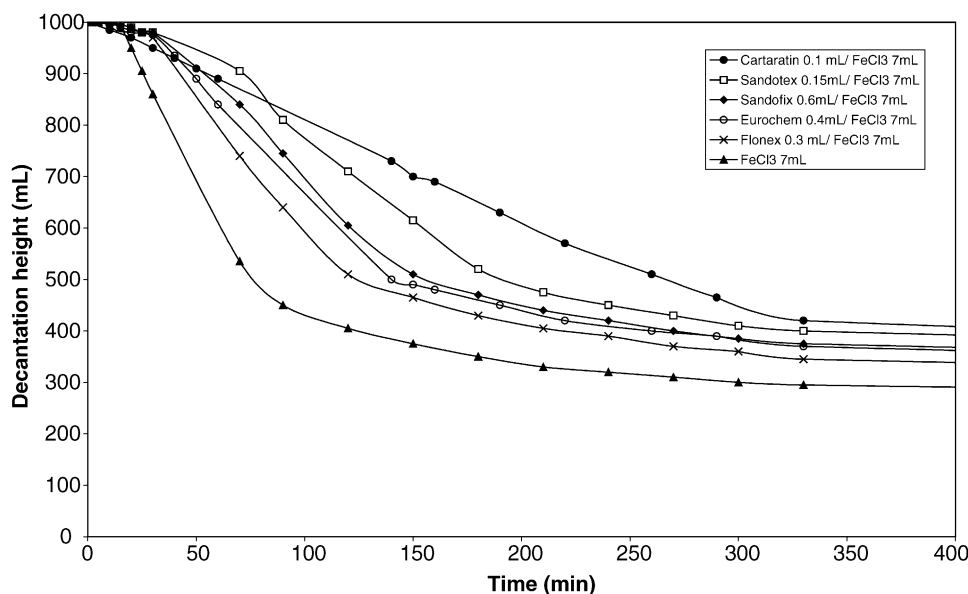


Fig. 4. Settling velocities as a function of flocculant.

Sandofix WE liq (Table 5). A dilution of a factor 30 (norms used in some European countries, such as Italy) would allow tax-free discharge. We then optimized the proportion of coagulant. Table 6 shows that the best absorbance is obtained with 2 mL of FeCl_3 .

4.2. Settling velocity

We completed the study of the absorbance of the supernatants by performing the Kynch tests. The settling velocity of the particles obtained with the optimum operating protocol allowed the determination of the decanter area. Fig. 4 shows that Flonex and FeCl_3 gave the fastest settling velocities but too high supernatant absorbencies. Sandofix WE liq and Eurochem Basic gave intermediate settling velocities and, in view of the supernatant absorbency, they were perfectly suited for this decantation. These settling velocities were low, of the order of $10^{-3} \text{ cm s}^{-1}$. Our objective was to show that the area of the decanter to be installed was not too big and also that the supernatant could be used directly as process water in the dyeing industry. Increasing the quantity of Sandofix WE liq did not modify the settling velocity of the particles (Fig. 5). Since we decided to consider the absorbance, it was thus more interesting to work with 0.8 mL L^{-1} of Sandofix WE liq flocculant. Fig. 6 shows that a more interesting settling velocity is obtained with only 1 mL of FeCl_3 . Table 7 gives a summary of the main tests carried out in plug settling. In order to determine the minimum decanter area, a quantity of coagulant of 2 mL L^{-1} was chosen. It is well known that Kynch theory is not directly valid for practical application due to compression. Basically, Kynch assumes that the settled solid at the bottom of the vessel is incompressible. To simplify and to approximate the size of

the decanter, the compression velocity is not taken into account and no compression model are used. In the case of a dye house treating 8–10 tons per day of cotton dyed with reactive dyes (any shade: light, medium, dark), the flow rate of effluents to be treated is $300,000 \text{ m}^3$ per year (250 working days). Out of these $300,000 \text{ m}^3$, $35,000 \text{ m}^3$ are dyeing baths that would be treated according to the membrane process patented by Clariant and the LPPE. After treatment of the effluents issued from the dyeing baths, a concentrate is recovered with an average flow rate of 0.285 L s^{-1} . For a critical sludge concentration of 17 g L^{-1} , the minimum flow rate was about $0.199 \text{ g s}^{-1} \text{ m}^{-2}$. The dry matter per kilogram of sludge was the initial concentration, $C_M = 8.2 \text{ g L}^{-1}$, of hydrolyzed reactive dyes, auxiliaries and other chemicals contained in the Sotratrex effluent mixture. The minimum area of the decanter will have to be 11.5 m^2 .

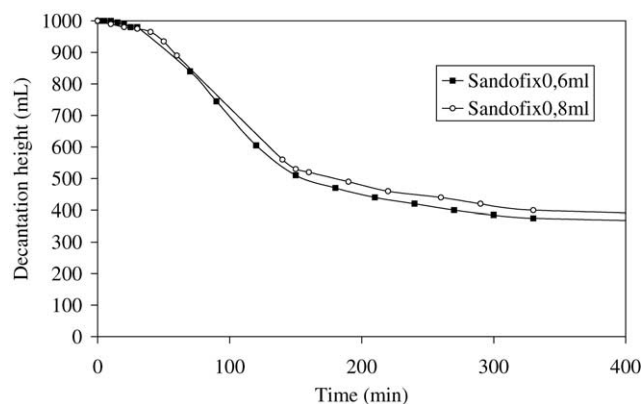


Fig. 5. Settling velocities as a function of flocculant quantity.

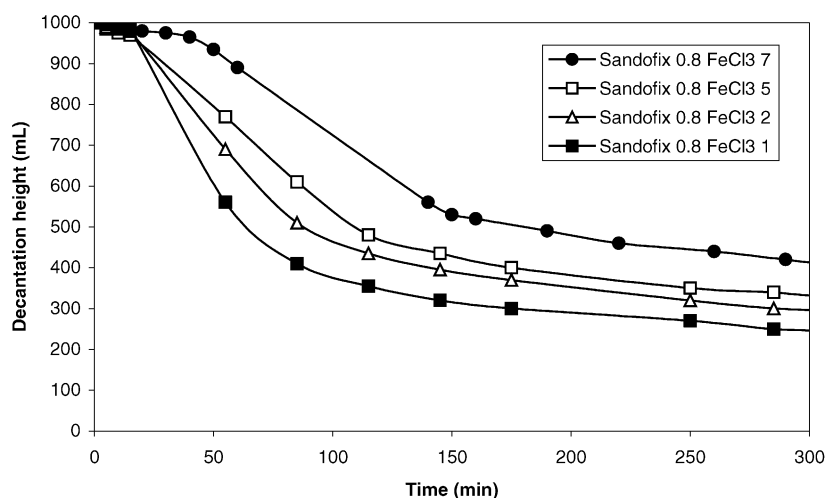


Fig. 6. Settling velocities as a function of coagulant quantity.

Table 7
Results of decantation test

Test	A_S	V_{sludges} (mL)	V_{sur} (mL)	ρ_b (kg/m ³)	Dried matter (g/kg _{sludge})	VCF ^a = A/A_S
Sandotex FAR 0.15 mL/FeCl ₃ 7mL	1.39	280	730	1013	32.7	6.40
FeCl ₃ 7 mL	1.65	230	790	1017	36.4	5.39
Flonex FI 28P40.3 mL/FeCl ₃ 7 mL	1.46	250	765	1012	32.8	6.10
Eurochem basic 0.4 mL/FeCl ₃ 7 mL	0.83	255	745	1009	33.3	10.72
Cartaratin 10CE 0.1 mL/FeCl ₃ 7 mL	1.52	260	740	1029	33.4	5.86
Sandofix WE Liq 0.6 mL/FeCl ₃ 7 mL	0.71	270	740	1015	31.7	12.54
Sandofix WE Liq 0.8 mL/FeCl ₃ 7 mL	0.38	275	725	1014	32.7	23.42
Sandofix WE Liq 0.8 mL/FeCl ₃ 5 mL	0.43	230	770	1007	31.1	20.70
Sandofix WE Liq 0.8 mL/FeCl ₃ 2 mL	0.293	190	810	1007	29.2	30.38
Sandofix WE Liq 0.8 mL/FeCl ₃ 1 mL	0.335	160	840	995	28.3	26.57
	A	C_M (g L ⁻¹)				
Final retentate after treatment of Sotratex mixture	8.9	8.24				

^a VCF: volumic concentration factor calculated with the absorbance ratio.

5. Conclusion

This study demonstrates the feasibility of the coagulation–flocculation–decantation treatment of highly concentrated effluents issued from the innovating process patented by Clariant and LPPE allowing the valorization of effluents from a dyeing process using hydrolyzed reactive dyes. We have shown that the flocculants and coagulants used in the industry must be carefully selected—the settling velocity could be too low, the supernatant absorbance could be too high. The best results were obtained with 0.8 mL L⁻¹ of Sandofix WE liq to which 2 mL L⁻¹ of FeCl₃ were added. The minimum area of the decanter to be installed was estimated at 12 m². Even though the settling velocities obtained with this coagulation–flocculation–decantation process were low, the significant decrease of the volume and the color of the supernatant leads us to believe that its re-use as process water is possible.

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