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Contribution to optimize the biological treatment of synthetic tannery effluent by the sequencing batch reactor

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Abstract

Tanning industries, widespread in the city of Fez, release massive amounts of waste (tanned and other wastewater) directly into the Sebou River without treatment. Faced with this situation, we have tried to study the effect of concentrations (500 and 1000 mg.L^{-1}) of total chromium in synthetic wastewater of tannery by Sequencing Batch Reactor SBR in order to optimize biological treatment process at the laboratory scale. Both treatments have proved to be quite effective, and the best one corresponds to a total chromium concentration of 500 mg. L^{-1}, with one cycle per day, and an aeration time of 23 hours. The corresponding results showed a reduction of all chemical parameters with a removal rates for total chromium (100%), COD_S (100%), total nitrogen (95.6%) and suspended solids (100%). While the synthetic sewage treatment at a total chromium concentration of 1000 mg.L^{-1} present a lowest performance.

Keywords: Chromium, Synthetic Tannery Effluent, Biological treatment, Optimization, Sequencing Batch Reactor SBR, Wastewater.

1. Introduction

In the developing countries heavy metal pollution has become serious due to mining, mineral, and smelting and tannery industry [1]. The leather industry is recognized as a serious environmental threat all over the world because it is associated with the generation of huge amounts of heavy metal contaminated solid wastes and disposal of these wastes is a serious problem [2]. The discharge of untreated effluents and sludge from treatment plants of tannery into nearby Fez areas is a growing problem in Morocco.

The city of Fez in a population which was estimated at 947.000 inhabitants in 2004 is currently as projections of about 1.073.000 inhabitants and reach 1.19 million in 2015 belongs to the region Fez–Boulmène which is located in north-central united [3]. Discharges of wastewater generated by this population in addition to those generated by industrial activity. Fez is of the order of 110.000 m^3 a day and reaches in 2015 (130.000 m^3) a day[1]. These discharges are characterized by a heavy industrial pollution (in the absence of pretreatment) especially since the release of vegetable oil and heavy metals (resulting from tannery, brassware processing). Therefore, the Sebou River which is the receiving end of all the wastewater from Fez is the most polluted nationally, with 28% of the total pollution. The sub-basin of Fez alone generates 40% of pollution. This creates a negative impact along the basin of Sebou and in particular on health causing waterborne diseases and water quality irrigation [4].

One study estimated the annual economic losses related to pollution of the Sebou, to over 50 million Dollars **[5]**. Because of these problems, it should be noted that the city of Fez also had a much deteriorated sewage system causing serious problems: polluted wastewater is discharged in rivers. Moreover, such pollution causes so many problems for the inhabitants of the Fes city. The report of the commission of the National Council Environment [5] points out to the existence of some industrial pollution in Sebou Basin, especially between its upstream and downstream. Table 1 explains the degree and the nature of such pollution.

These industrial discharges into the Sebou River produce large loads of metal traces. For example, tanneries are, the main sources of chromium in the Fez region, is pouring more than 255 tones a year^{-1} [6]. Analysis of measurements performed by National Office of Drinking Water [7] revealed that the chromium contents

greatly exceed acceptable standards (205 à 6400 mg.L^{-1}). Concentrations of total chromium tannery effluent can reach greater value [8;9]. In fact, Cr^{III} can be oxidized to Cr^{VI} in the presence of chemical oxydants (MnO_2,…) and microorganisms, taking into consideration that the latter (Cr^{VI}) is more toxic than the former ones. The chromium content in the effluent will be transmitted into the environment where it can undergo oxidation reactions to convert it on Cr^{VI} [10].
Many works have been interested to the poor water quality and toxicity of Sebou river [11-13] but the important chromium pollution stills arising.
The goals of such research is to optimize the treatment of synthetic tannery effluent, based on the study of the effect of 500 and 1000 mg.L^{-1} concentrations of chromium in three different effluents.

Table 1: Industrial pollution Sebou Basin, between the upstream and downstream [5].

Locality	BOD_5 (T.an^{-1})	COD (T.an^{-1})	P_T (T.an^{-1})	N_T (T.an^{-1})	Chromium (T.an^{-1})
Sefrou	174	412	2	3	1
Fès	11911	25482	74	416	101
Mechraa Bel Ksiri	713	1650	-	15	-
Sidi Allal Tazi	878	2115	-	32	-
Total	13681	29659	76	466	102

2. Materials and Methods

2.1. Biological Material

In this study, we used activated sludge from the wastewater treatment plant of Akrache in the Rabat, Morocco.

2.2. Characterization of three tanneries effluents

The chemical composition of effluents from three tanneries in Fez are illustrated in (Table 2).

Table 2: *Chemical and physical* characterization of tannery effluents.

Parameters	**Concentration in (mg.L^{-1})**		
	Effluent 1	**Effluent 2**	**Effluent 3**
Temperature (°C)	31	23	24
pH	3.58	10.94	3.68
COD_T	17511	8944	4193
COD_S	14746	5069	3225
BOD_5	3000	2000	1000
SS	5165	4870	5110
P_T	62.80	62	17.30
N_T	50	30	32
NO_3^-	37.80	10.80	21.30
NO_2^-	9.86	17.20	8.36
NH_4^+	0,90	1.10	1.97
Cl^-	7100	124425	10650
Phenolic compound	10295	6045	870
Cr_T	-	-	1000

Effluent 1: effluent containing phenol compound.
Effluent 2: effluent containing sulfates, lime.
Effluent 3: effluent containing chromium

2.3. Effluent treated

The effluent used was a synthetic wastewater simulating a real tannery effluent whose chemical composition is shown in (Table 3).

Table 3: Composition of synthetic tanneries effluents.

Chemical composition of synthetic Tanneries effluents ($g.L^{-1}$)		**Chemical composition of elements traces** ($g.L^{-1}$)	
Glucose	3.64	$FeCl_{3,H2O}$	1.5
$(NH_4)_2SO_4$	0.91	H_3BO_3	0.15
$MgSO_4,7H_2O$	0.025	KI	0.03
$FeSO_4,7H_2O$	0.02	$MnCl_2,4H_2O$	0.12
KH_2PO_4	0.088	Na_2MO_4,H_2O	0.06
K_2HPO_4	0.09	$ZnSO_4,7H_2O$	0.12
Na_2CO_3	0.066	$CaCl_2,6H_2O$	0.15
$NaHCO_3$	0.105	$CuSO_4,5H_2O$	0.03
$CaCl_2$	0.03	-	-
$KCrO_4$	*1.86	-	-

2.4. Process treatment description

The treatment process of effluent is by aeration in a sequencing batch reactor (SBR). It is based on the principle of aerobic biological treatment of effluent in sequential mode. Each day, a volume of 260 ml of synthetic wastewater (prepare every day) is introduced into the SBR containing two liters of capacity activated sludge (sixth volume of water used) with a feed time of 4 minutes, and aired 23 hours until elimination of biodegradable organic matter. After stopping the aeration-agitation, settling phase begins which lasts 54 minutes allowing the separation of sludge from the treated effluent. At the end of the cycle, a volume of 260 ml of treated effluent is withdrawn and replaced by a new raw effluent synthetic with a working volume of 540 ml and 37.5 $ml.j^{-1}$ of sludge is removed with a 30 days aged sludge, after the sludge still at rest for 2 min for the next filling, then a new cycle begins (Figure 1). The different phases of feeding, aeration, settling, decanting the treated effluent and withdrawal of excess sludge are carried out by programmers. The biomass concentration is stabilized between 3 and 6 $g.L^{-1}$ of suspended solids SS by a purge of excess sludge.

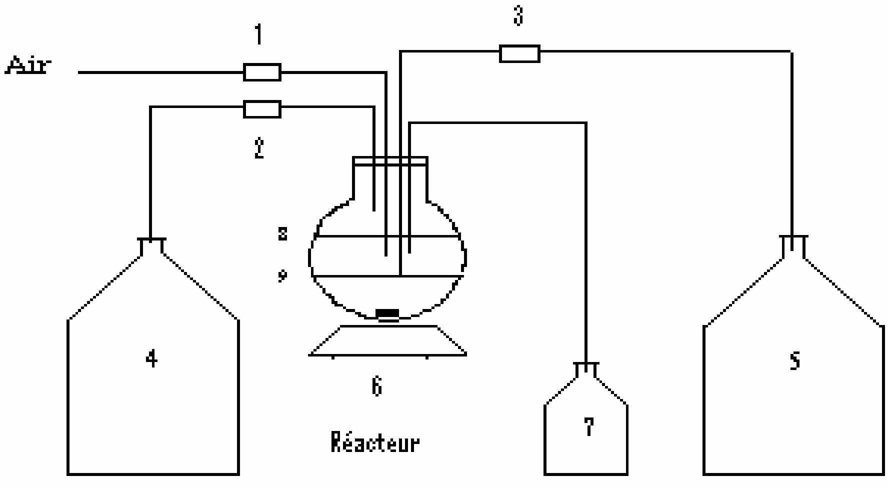

Figure 1: Schematic of the SBR process laboratory scale.

1
2 Timers providing the programming aeration (1), food (2) and withdrawal (3)
3
4 - Synthetic raw wastewater.
5 - Wastewater treated.
6 - Stirrer.
7 - Excess sludge.
8 - Level of mixed liquor used corresponding to a volume of 1500 ml (the reactor operation).
9 - Level of mixed liquor after settling and racking, which is the volume of settled sludge corresponding to a volume of 750 ml.

2.5. Chemical and physical methods

The Chemical and physical wastewaters methods were made using the methods described in [14]. They focused on the following parameters :

2.5.1. Chemical Oxygen Demand (COD)

The determination of COD was performed by the method of potassium dichromate. The method is based on oxidation to a boil (150°C for two hours) of oxidizable matter with an excess of potassium dichromate in acid medium and in presence of silver sulphate (catalyst) and sulphate of mercury (complexing). COD values were determined by spectrophotome- ter type UV / visible brand JENWAY6105 and a wavelength of 585 nm [14].

2.5.2. Biological oxygen demand (BOD)

Measuring the dissolved oxygen content is effected by incubating a volume of 150 ml of water sample to be analyzed in a BOD meter type OxitopR IS6 (AND 618-4/619-4). The sample incubation lasts five days in the dark at a temperature of 20°C. BOD_5 values are expressed in $mg.L^{-1}$ of O_2 [14].

2.5.3. Determination of Total Chromium (Cr_T)

Elemental analysis of samples of total chromium was carried out by plasma emission spectrometry inductively (ICP-AES) [14].

2.5.4. Total Nitrogen (N_T)

Nitrogen compounds present in the effluent are oxidized to nitrate by alkaline persulphate solution in an autoclave (120°C/15 min). Then, the nitrates are determined by the method of sodium salicylate. The positive reaction produces a yellow colored product named paranitro sodium salicylate which is detectable at a wavelength of 415 nm [14].

2.5.5. Nitrate (NO_3^-)

Reaction with the acid nitrate sulfosalycilique (formed by adding to the sample of sodium salicylate and sulfuric acid). The derivative obtained in the presence of ammonia gives a yellow color stable. Measuring spectrophotometrically at 415nm wavelength of this coloring [14].

2.5.6. Nitrite (NO_2^-)

Method to zambelli reagent. The sulfuric acid in hydrochloric acid medium in the presence of NH_4^+ ions and ions with phenol forms NO_2^- yellow colored complex whose intensity is proportional to the concentration of NO_2^- [14].

2.5.7. Ammonium ions (NH_4^+)

Ammonium ions are determined by the indophenol method. In alkaline medium and in the presence of catalyst such as sodium nitroprusside, they react with the phenol and sodium hypochlorite, and form the indophenol blue likely a colorimetric assay. Reading the optical density in a spectrophotometer is at a wavelength of 630 nm. The concentration of ammonium ion is derived from a range of standard solution of NH_4Cl at concentrations between 1 and 10 $mg.L^{-1}$ of NH_4^+ [14].

2.5.8. Orthophosphates (PO_4^{3-})

The determination of their concentration is based on the formation of a complex antimony molybdate-phosphate. This complex is reduced by ascorbic acid compound strongly colored blue. The optical density is read at a wavelength of 882 nm. A standard curve is made from a solution of KH_2PO_4 at concentrations of between 5 and 10 $mg.L^{-1}$ of PO_4^{3-} [14].

2.5.9. Total Phosphorus (P_T)

The determination of total phosphorus is carried out in two stages. The first step is a digestion "hot" in an acidic medium which converts all of the phosphorus present in the sample to be analyzed in orthophosphates (PO_4^{3-}) [14].

2.5.10. *Suspended Solids (SS)*
TSS are determined by centrifuging a 20 ml sample at 3000 × g for 20 minutes. The pellet was placed in a previously weighed porcelain dish and dried in an oven at 105 ° C for 24 hours. The difference between the dry weight of the sample and that of the cup determined by the ratio of suspended solids, expressed in mg. L^{-1} [14].

2.5.11. *Dry residue (DR)*
A volume of 20 ml is placed in a previously weighed porcelain dish and dried in an oven at 105°C for 24 hours. The difference between the dry weight of the sample and that of the cup determined by the rate of RS, expressed in mg. L^{-1} [14].
The calculation of reduction rate of a parameter x, expressed in percentage is based on the following formula:
Removal rate = (C_i(x) – C_f(x) / C_i(x)) . 100
C_i(x) = Initial concentration of x in the wastewater.
C_f(x) = Final concentration of x in the treated wastewater

3. Results and discussion

3.1. Discution of three effluent tanneries analyzed
As part of the study of the biological treatment of tannery effluents from the city of Fez, we conducted this study on three real effluents from tanneries to prepare synthetic tannery effluents. Table 2 summarizes the concentrations of the various parameters of the three real effluents of tanneries.
The results show that the wastewater quality varies considerably from one production method to another. Effluents containing phenolic compounds are the most polluting DCO_T with contents that can reach 17.511 mg.L^{-1}, 3000 mg.L^{-1} BOD_5, 10295 mg.L^{-1} of phenolic compounds and 5165 mg.L^{-1} of suspended solids. The analysis of effluent containing sulphide and lime show a significant pollutant load 8944 mg.L^{-1} of DCO_T, 2000 mg.L^{-1} BOD_5, 124 425 mg.L^{-1} chloride and 4870 mg.L^{-1} of suspended solids. Effluents containing chromium are also responsible in polluting 4193 mg.L^{-1} of DCO_T, 1000 mg.L^{-1} BOD_5, 1000 mg.L^{-1} of total chromium, 10,650 mg.L^{-1} of chloride and 5110 mg.L^{-1} of suspended solids. Despite the socioeconomic impact of the tanning industry through the creation of jobs and income, the company has a negative image of this activity due to the resulting pollution.
The purpose of the analysis of the three effluents from tanneries is the determination of concentrations of chemical composition of effluents from tanneries to prepare synthetic tannery effluents close to reality.

3.2. Performance obtained for the organic pollution removal
The evolution of CODs based on the total chromium concentration is shown in Figure 2.

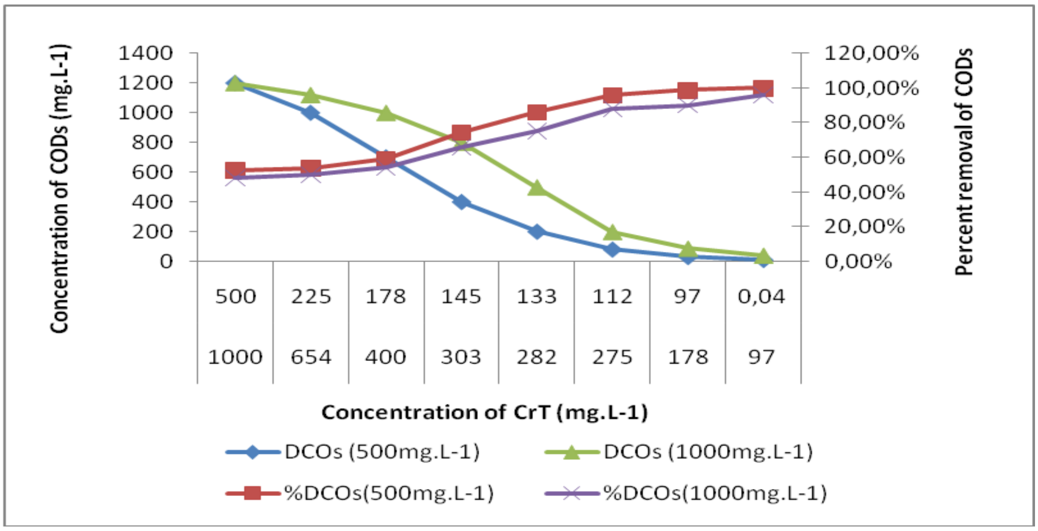

Figure 2: Variation of the concentration and elimination rate of CODs as function of total chromium concentration at 500 and 1000 mg.L^{-1} during 24 hours of treatment.

The results obtained for synthetic tannery effluent treatment at a concentration of 500 mg.L^{-1} total chromium show that the strongest performance of the SBR in terms of CODs reduction was recorded after two months of the SBR operation, since the maximum removal rate of CODs was 100% illustrated in Figure 2. Avans we arrived at a concentration that spreads to standard moin two months but prolongs to have a removal rate equal or close to 100%. The CODs concentration at the beginning of treatment in SBR was 1200 mg.L^{-1} and the output was 0.8 mg.L^{-1}. This corresponds to a maximum elimination chromium rate (100%). The treatment of synthetic tannery effluent at a concentration of 1000 mg.L^{-1} total chromium showed a slight decrease in the COD_S reduction with a maximum removal rate of 96% Figure 2, the concentration of the CODs decrease from 1200 to 40 mg.L^{-1} as shown in Figure 2. These results are closer to those reported by [15] who found tannery effluent removal rate of 90% in Turkey.
This difference in performance was explained by the adaptation of microorganisms in activated sludge to total chromium added to the synthetic effluent. From these results, we can conclude that the decreased concentration of total chromium promotes the adaptation of the activated sludge biomass and the organic pollution removal, in other words, these data show that the CODs reduction depends of total chromium concentration. Indeed, a considerable time is needed for the degradation and assimilation of organic matter by micro-organisms in aerobic conditions [16;17].

3.3. Performance obtained for nitrogen pollution removal
Early treatment of synthetic tannery effluent to a concentration of 500 mg.L^{-1} in a sequencing batch reactor, the concentration of total nitrogen is 90 mg.L^{-1}. After eight weeks of treatment, this concentration has decreased to 1 mg.L^{-1} in the reactor outlet, with a removal rate of 98.8% as shown in Figure 3. The results are similar to those of the work done in Italy whit [18], with a total nitrogen rate of 98%.

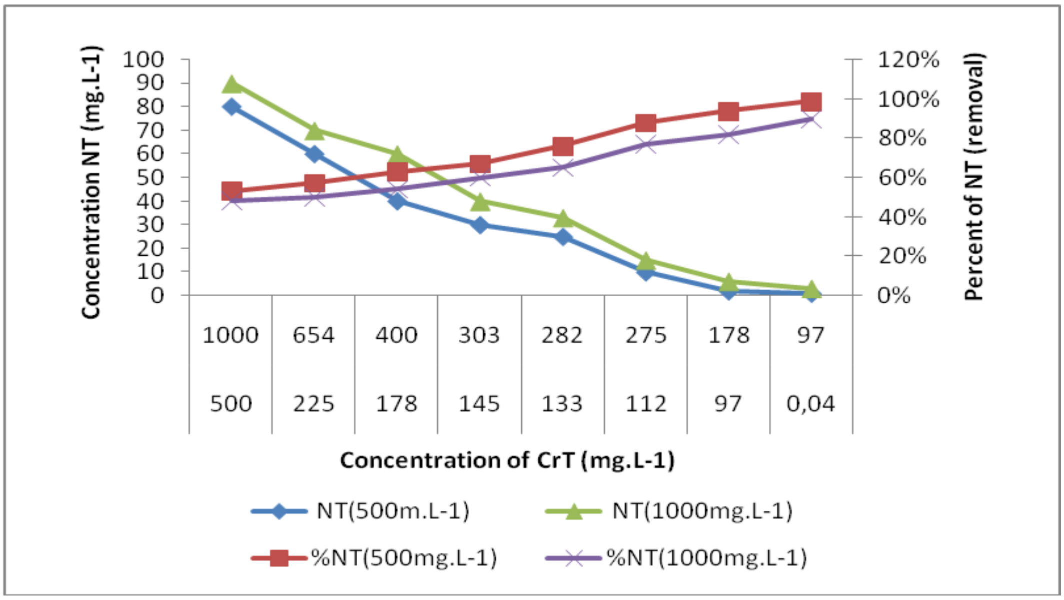

Figure 3: Variation of the concentration and removal rate of total nitrogen as function of total chromium concentration at 500 and 1000 mg. L^{-1} during 24 hours of treatment.

The decreasing concentration of total chromium in synthetic sewage led to a decrease in the average concentration of total nitrogen in treated effluent from 500 to 0.04 mg.L^{-1}, the rate of nitrogen reduction reached 99.99% in the case of the synthetic sewage treatment. At the concentration of 1000 mg.L^{-1}, the total chromium concentration decreased from 1000 to 1 mg.L^{-1}, with a removal rate of 90%. This low rate of chromium concentration (500 mg.L^{-1}) acts without toxicity for nitrifying bacteria.
Figure 2 shows that the elimination rate of CODs at 500 mg.L^{-1} of chromium is 100% and thus accompanied by a total consumption of soluble organic matter by nitrifying bacteria, which promotes the increase of bacterial biomass.

According to the literature [19], a decrease of organic matter promotes nitrification because nitrification of 1 mg $N\text{-}NH_4^+.L^{-1}$ consumes 7.14 mg $CaCO_3.L^{-1}$. We conclude that reducing the concentration of CODs is correlated with an increase of nitrifying bacteria, which, as a consequence, decreases the concentration of total nitrogen at the outlet of the SBR.

3.4. Performance obtained for phosphorus pollution removal

As it is shown in Figure 4; the best removal rate of phosphorus is obtained at 92% with the treatment of synthetic effluent at 500 $mg.L^{-1}$, but decreases at 88% when the concentration of total chromium equals 1000 $mg.L^{-1}$. During the first week of treatment, the total concentrations of chromium and nitrogen are respectively 500 and 20 $mg.L^{-1}$. After eight weeks, it decreases to 0.04 $mg.L^{-1}$ for total chromium and 0.1 $mg.L^{-1}$ for total phosphorus.

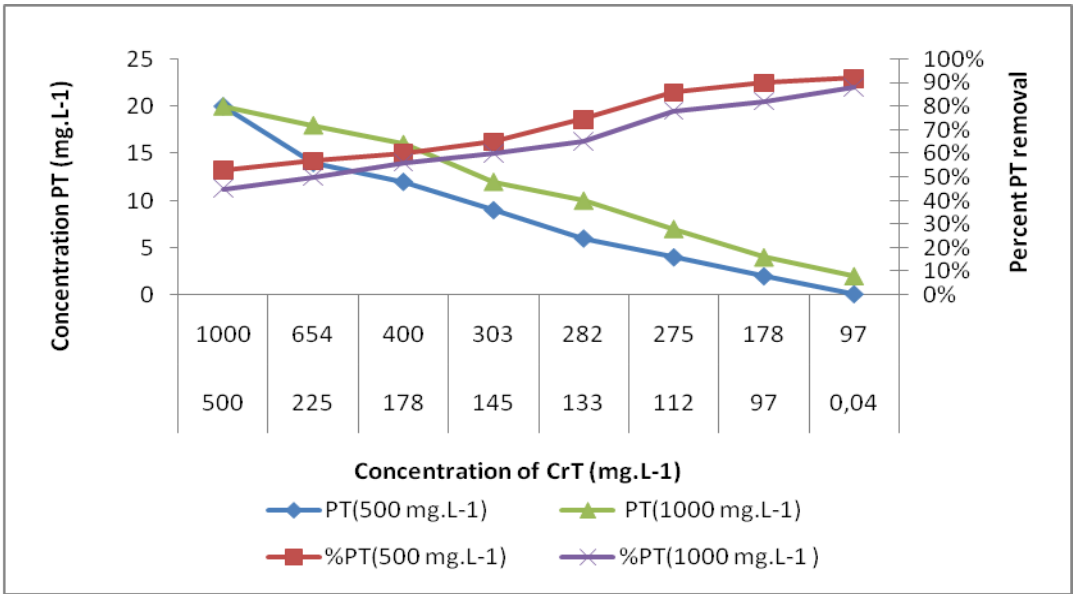

Figure 4: Variation of concentration and removal rate of phosphorus as function of total chromium concentration at 500 and 1000 mg. L^{-1} during 24 hours of treatment.

While the treatment of synthetic sewage (1000 $mg.L^{-1}$), the concentrations of total chromium and total nitrogen in the first weeks are respectively 1000 and 20 $mg.L^{-1}$. But decrease to 97 and 2 $mg.L^{-1}$ after eight weeks of treatments. Indeed, a good phosphorus removal with the aeration phase of 23 hours requires the reduction of total chromium concentration at 500 $mg.L^{-1}$at a settling period of 54 minutes. This condition has an effect on micro-organisms which release phosphorus accumulated during the aeration phase in the extracellular [20;21]. Finally, the phosphorus concentration obtained after treatment of the synthetic effluent (500 $mg.L^{-1}$ of total chromium) is below the standards discharge standards Moroccan [22]. Therefore, the rejection of the treated effluent into the environment causes no adverse effect on environment.

3.5. Elimination rates of various parameters studied after treatment of the synthetic effluent

The results of the maximum removal rate of the Cr_T, COD_S, COD_T, suspended solids SS, dry residues DR and nutrients after treatment in the SBR as function of total chromium concentrations in synthetic sewage are shown in Figure 5.

The treatment of synthetic effluent at concentration 1000 mg. L^{-1} of total chromium, with 23 hours of aeration and 54 minutes of settling per day, showed maximum removal rates of 90% for total chromium, 98.6% for the CODs and 98% for COD_T. The maximum removal rate for total chromium at a concentration of (500 mg. L^{-1}) is 100% and 99% of COD_S. These results of removal rates obtained still more interesting (or important) when they are compared to literature's data with 90% of removal rate of COD_T [23; 24].

At a concentration of 1000 $mg.L^{-1}$ of total chromium, the removal rates were 90%, 88%, 89%, 90%, 97% and 88% respectively for total nitrogen, ammonium, nitrate, nitrite, orthophosphate and total phosphorus (Figure

5). The results obtained are in agreement with those found in the effluent tannery in India [25;26]. The following findings are: reduction rates of COD_T (81.2%), total nitrogen (78.6%) and nitrate (92.8%).
Concerning our research on 500 mg.L^{-1} of total chromium, the removal rates has led to what follows: total nitrogen 96%, ammonium 94%, nitrate 93%, nitrite 94%, orthophosphate 95% and total phosphorus 92%.

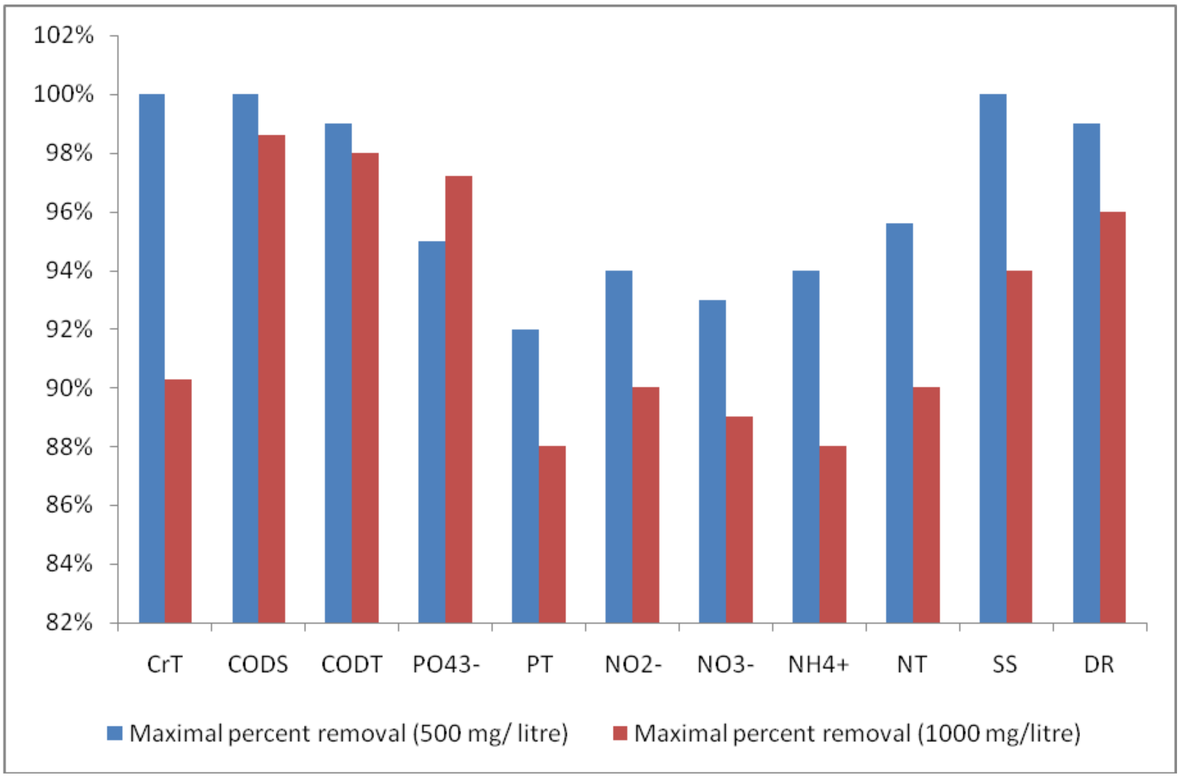

Figure 5: Maximum removal rates of Cr_T, COD_S, COD_T, SS, DR and nutrients after treatment of the synthetic effluent at concentrations of 500 mg. L^{-1} and 1000 mg.L^{-1} during 24 hours of treatment.

Hence, the reduction of these parameters increases with decreasing of total chromium concentration. This could be due to incomplete degradation of chromium at a concentration of 1000 mg.L^{-1} and the residual chromium has a toxic effect on bacterial biomass, causing a decrease in removal rate of nutrients.
The ammonium ions are converted to nitrite ions by micro-organisms of the genus *Nitrosomonas* with nitration reaction. The former ions (nitrites) are then oxidized to nitrate ions by micro-organisms of the genus *Nitrobacter* with nitrification reaction [27].
The suspended solids SS constitute an essential part of the carbon pollution. The latters elimination contributes to a better removal efficiency of COD. The results of the removal rate of suspended solids SS and dry residue DR in the SBR as a function of total chromium concentration in synthetic sewage (1000 and 500 mg.L^{-1}) as Figure 5 shows. The elimination rates of suspended solids SS and dry residue DR also increase with reduction of total chromium concentration, and present 94% and 96% at a concentration of 1000 mg.L^{-1}, respectively 100% and 99% for the concentration of 500 mg.L^{-1}. This rise could be explained by the increasing of bacterial floc formation and bacteria filamentous [28;29]. The resistance to chrome could be due to a chromosomal mutation of the sulfate transport system [30]. The CrO_4^{2-} can penetrate into the cells by the enzyme called permease sulfate (SO_4^{2-} ions). In general, transport systems are sets of proteins embedded in the membrane of the cell. These proteins come together and combine, and are selective for one or more substances in the membrane [19].
Elimination rates improve with the decrease of total chromium concentration in the effluent synthetic. This improvement could be due to better assimilation of organic matter by micro-organisms at low concentrations of total chromium. The maximum removal rates of total nitrogen, nitrite, nitrate, total phosphorus and orthophosphate increases with reduction of total chromium concentration in synthetic sewage.

Conclusion

The sequencing batch reactor method was applied to the treatment of synthetic effluent with two different concentrations of total chromium (500 and 1000 mg.L^{-1}). Following the results obtained, we have come to conclude that:

- A greater reduction of all chemicals parameters in relation to 500 mg.L^{-1},
- The high removal rate rises when the concentration of total chromium decreases (1000 to 500 mg.L^{-1}), thus presenting maximum yields of eliminating rate: 100% for total chromium and CODs. In addition to this, we have total nitrogen 96% and their derived such as ammonium ion 94%, nitrate 93%, nitrite 94%, while the total phosphorus and orthophosphorus present 92 and 95%.
- Finally, suspended solids and dry residue show a removal rate percent of 100% and 99% at the concentration of 500 mg.L^{-1} respectively 94% and 96% for 1000 mg.L^{-1}. These results improve the efficiency of the treated effluent with decreasing the concentration of total chromium at 500 mg.L^{-1}.
- The treatment with a concentration of 1000 mg.L^{-1} total chromium gave a reduction rate of 96% and 90% respectively for total chromium and CODs. Thus, the results showed a 90% removal for total nitrogen, 88% for ammonium, 90% for nitrite, 89% for nitrate, 88% for total phosphorus and 97% for orthophosphate.

Both treatments of synthetic tannery effluent (500 and 1000 mg.L^{-1} of total chromium) by SBR system have proved to be quite effective. However, the first one (500 mg.L^{-1}) is found to be more performing than the second one (1000 mg.L^{-1}). Therefore, we highly recommended it for future large scale experimentation.

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