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Using a bio-flocculent in the process of coagulation flocculation for optimizing the chromium removal from the polluted water

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Abstract
Coagulation-flocculation is a process often used in the industrial wastewater treatment. To replace the use of the chemical products in this kind of process and eliminate their downside effects on the water disposal process, a bio-flocculent extracted from the racket of *Opuntia ficus indica,* "cactus juice", has been utilized. Two types of wastewater have been used in the experiments: A synthetic solution of chromium sulfate at 100ppm or 500ppm and a real effluent coming from a tannery at 90ppm or 900ppm. The results obtained show that the flocculation is rapid in the range of pH 7-9. Neutralization and lime's coagulation followed by cactus juice flocculation greatly reduce metal charge values, and this was confirmed by the appropriate ICP's measurement (eg with 4 ml of cactus juice for 500 ml of effluent at 90 ppm, the initial charge of 176 mg/l was reduced by 99.9%. Same operations lead also to a significant drop in COD and BOD5 estimated at (94% and 95%) respectively. In the case of tannery effluent at 900 ppm, a turbidity reduction of 98.6% has been achieved. The flocculation results obtained with the two solutions tested were satisfying compared to those obtained with the chemical flocculent DKFLOC.

Keywords: Coagulation-flocculation; industrial effluent; bio-flocculent; chromium.

1. Introduction
The coagulation-flocculation is a process commonly used for the treatment of raw water and in particular wastewater treatment. It allows the removal of colloidal particles and hydrophobic metal. Several chemical coagulants such as iron or sulfate, iron chloride or aluminum sulfate are often used in the first operation **[1-2].** Also, the flocculation process requires addition of some chemical products, such as polyacrylamide or the sulfate polyferric **[1]**. In order to replace these chemicals and avoid their costly recycling treatment, biological coagulants have been proposed. Already published studies have demonstrated the effectiveness of the chitosan as a coagulant **[3-6]** or flocculent **[4].** The seed of *Moringa oleifera* has been known for decades for its coagulation properties **[7-8].** Recently other extracted biological products have been proposed **[9-11].** It was also reported that the *Opuntia ficus indica* may also act as a good coagulant or flocculent **[14].** In this work, we used the juice extracted from the racket of this cactus to estimate its flocculation ability in the total chromium elimination process. Bio-flocculents, as they are biodegradable and available all across the country, would be a good alternative for the treatment of polluted waters especially for those containing heavy metals such as the chromium discharged in the tannery effluent.

2. Material and methods
2.1. Preparation of cactus juice
The coagulation-flocculation tests were carried out following the standard practice for coagulation-flocculation testing of raw water to evaluate the chemicals dosages and conditions required to achieve optimum results.

The bio-flocculent used in our experiments is extracted from the racket of the prickly pear *(Opuntia ficus indica)*. This cactus grows in abundance across Morocco, was collected in the vicinity of the city of Fez. The racket is carefully cleaned with water and then crushed and pressed. The juice is filtered, diluted by 20% with distilled water then homogenized by stirring for 20 minutes. The product obtained (Fig.1) is a water miscible viscous liquid, green color and pH = 6.5. Its bulk density is 1027 kg/l and contains 92% water. It is tested for its ability of flocculation during 15 days without any preservative system.

Photo 1: Cactus juice

2.2. Use of cactus juice in the process of coagulation- flocculation

Two types of polluted water were treated for chromium elimination using the physicochemical process "coagulation-flocculation" with cactus juice (bio-flocculent).

- First in a purely synthetic chromium sulfate solution ($Cr_2(SO_4)_3$) prepared in the laboratory at 100 ppm and 500 ppm with a turbidity of 102 and 242 NTU respectively.

- Secondly, in a sample of an industrial effluent taken from a tannery unit in full activity at 90 ppm and 900 ppm with turbidity of 152 and 316 NTU, respectively. In this case, the most important physicochemical parameters (temperature, pH, conductivity, turbidity, suspended matter, BOD5, COD, NH_4^+, nitrates, nitrites and orthophosphates) were immediately determined.

Samples of 500ml of synthetic solution were placed in a Jar-test apparatus (model JLT 6 Leaching test - photo2), and the pH (pH meter HANNA Instruments) was adjusted by NaOH. The colloid coagulation was established by adding the limewater Ca(OH)2. with 95 % of purity in a 10 g/l solution form. Stirring was set on 120 revolution/min (rpm).

Thereafter, to trigger the flocculation mechanism, we added (2, 4, 6 or 8 ml) of cactus juice in some samples and (2 ml) of chemical flocculent (DKFLOC, K=436/VH MW LOT.GA3816; MET25G) in the other samples. The stirrer speed was set on 60 rpm for 20 minutes.

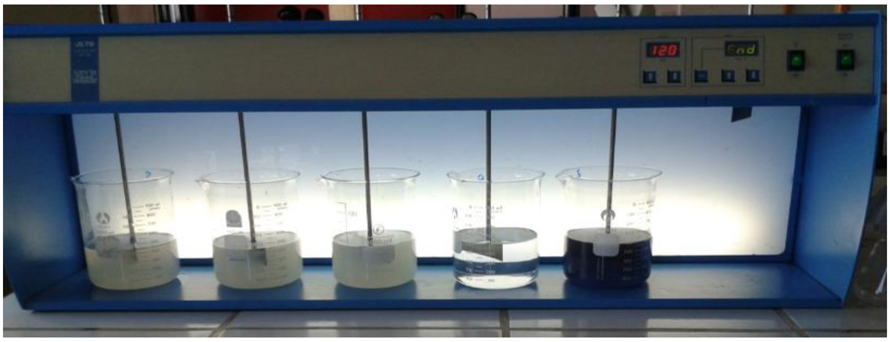

Photo 2: Jar test

The stirrers were turn off and the flocks allowed settling for 15 min, then we proceeded to measure the conductivity (type water quality instrument; YSI Scientific), turbidity (turbidity AQUAfast AQ3010, Orion), UV-visible absorbance using spectrophotometer (type JP Selecta) in the supernatant as well as the quantification of the total chromium in an emission spectrometer with inductive coupled plasma (Jobin-Yvon ULTIMA model).

The same steps were followed for the samples of the effluent coming from the tanning industry.

The removal efficiency is given by the equation below:

$$\mathbf{R\%=(1-C_f/C_i) * 100}$$

C_i: Initial concentration before treatment;
C_f: Final concentration after treatment.

3. Results and discussion

3.1. Characterization of tannery effluent

The evaluation of raw waste water pollution degree is based on determining a number of physicochemical parameters that characterize this wastewater. The table 1 summarizes the Physicochemical characteristics of the effluent before and after the tanning treatment.

Table 1: Physicochemical analysis of the tannery effluent at 90 ppm

Parametres (mg / l)	Raw tannery	Effluent + lime	Abatement rate treatment by JC	Abatement rate of treatment IF	Limit values of indirect discharges
Temperature (° C)	19.3	19.3	15.2	15.2	**30 ° C**
pH	3.5	9.6	7.7	8.4	**6.5 - 8.5**
Conductivity (µS/cm)	1400	1500	1300	1000	_
Turbidity (NTU)	37.3	158	6.31	4.04	_
[Cr]	176 *	176	0.015	0.026	**2**
COD	3850	3850	260	230	**1000**
BOD_5	508	508	25.7	23.5	**500**
Nitrate	73.95	73.95	5.11	8.83	_
Nitrite	3.71	3.71	0.98	0.83	_
Ammonium	10.64	10.64	1.73	1.48	_
Orthophosphate	2.62	2.62	2.2	1.39	**10**
MY	5300	5300	430	180	**600**

(*): Dilute 10 time

In the absence of the bio-organic flocculent treatment, the physico-chemical characterization results of tannery effluents show that the pollution level is fairly high and by nature its components are quite heterogeneous (Table 1).

Indeed, standard comparison of the recorded values before treatment with the flocculent shows that most studied parameters values exceed by far the alarming pollution limits.

The tannery effluent treatment using our bio-organic flocculent at a total chromium concentration of 176 mg/l showed good performance of coagulation-flocculation in terms of reduction of the concentration those elements: COD (94%), BOD5 (95%), turbidity (96%), suspended matter (91.9%), ortophosphates (17%), nitrates (73%), nitrites (93%) and ammonium (83%).

The bio-organic flocculent and coagulant in the coagulation-flocculation process has been also used to remove turbidity, heavy metals, organic charge, suspended solids and alkalinity samples, and it was the subject of several studies **[15, 17].**

The solution pH is an important factor in the total chromium elimination, turbidity, and the suspended matter **[18].** Our study demonstrated that the optimum results were obtained at pH neutral and pH = 9. Solution pH affects not only the surface charge of the coagulant but also the stability of the suspension. Therefore, the study of this parameter is required in order to determine the optimum pH condition in the treatment system **[15].**

In addition, it has been pointed that all parameters studied are in tendency toward same influence with different reduction efficiency values. Our results show strong concordance with those reported by Faouzi and al. **[21]** who found an efficiency of 96% for chromium removal from tannery effluents using a biological treatment.

Besides these results, our bio-flocculent provided a great COD reduction efficiencies comparing to those achieved by Ariffin and al. **[15]**, who achieved a COD removal efficiency of 72.5%.

3.2. Determination of the optimal dosage of cactus juice

The optimal dosage of cactus juice for a better flocculation was investigated in the case of the synthetic solution and the industrial effluent. To optimize the pH of the coagulation process, jar tests were conducted over

different pH values. The best result obtained at pH 9 and neutral pH where the colloidal particles destabilization occurs. Flocks are formed quickly as soon as we add the bio-flocculent in the different samples.
Coagulation-flocculation tests were carried out on raw solution of a pH of 3.5, and no changes in the solution status have been noticed.
As the lime is added, both synthetic solution and industrial effluent's pH increases and becomes neutral, except for the industrial solution at 900 ppm where the pH does not vary and remains acid at pH 3,6.
In this case the pH value was maintained at neutral, the flocculent (cactus juice) dosage ranged from 2 to 8 ml, flocks formed are small size in the synthetic solution, and almost constant in the effluent solution (between 1 and 1.5 mm).

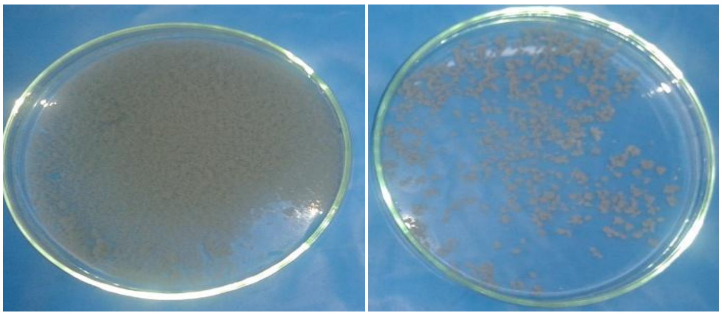

Photo 3: Dimensions of the flocks in the case of neutral and basic pH (neutral pH on left image and basic pH on right image)

Also, Coagulation-flocculation tests were performed in the same manner as mentioned before but the pH value was adjusted at pH 9 with 2 ml of cactus juice as flocculent. Flocks formed in the synthetic and real effluents are both of small sizes. Flocks become macro and their sizes vary from 3 to 5 mm (Photo 3) starting from 4ml of the bio-flocculent added to 500 ml of the effluent except for the case of the tannery effluent at 900 ppm (Tables 2-5). Mixing process during the physicochemical treatment ensures a good distribution of coagulant agents and chemical destabilization of colloids, besides it eases contact among particles and prevents damage to the formed flocks **[19].**

Table 2: Formation and removal of chromium flocks according to cactus juice dosages: Case of a synthetic solution at 100 ppm Cr.

Agents	Control CH + SS	10 ml CH + 2 ml J	10 ml CH +4 ml J	10 ml CH +6 ml J	10 ml CH +8 ml J	10 ml CH + 2 ml IF
[Cr] ppm at neutral pH	**100**	0.36	0.32	0.32	0.32	0.28
[Cr] ppm at pH = 9	**100**	0.20	0.19	0.17	0.17	0.27
Observation	**Absence of flocks**	**MI F**	**MAF**	**MAF**	**MAF**	**MAF**

* CH: Lime
* SS: Synthetic solution
* J: Juices
* IF: Industrial flocculent
* MIF: Micro-flocks
* MAF: Macro-flocks

The treatment of synthetic and industrial polluted water led to very significant chromium removal efficiency values. In the case of the synthetic solution, we found in the supernatant a chromium concentration of about 0.2 ppm. This value is comparable to that obtained using the industrial flocculent (Tables 2 and 3). The chromium concentration found in the sample was around 0.3 ppm fort the industrial rejection of 90 ppm and pH = 9. Same values obtained in the presence of industrial flocculent (Table 4). When the industrial rejection set at 900 ppm, the macro-flocks were formed with a bio-flocculent dosage of 6 ml and the chromium values detected vary between 1.22 to 1.45 ppm against 1.1 ppm in the presence of DKFLOC flocculent (Table5).

Table 3: Formation of flocks and elimination of chromium relay on cactus juice dosages: Case of synthetic solution at 500 ppm of Cr.

Agents	Control : CH + SS	10 ml CH + 2 ml J	10 ml CH +4 ml J	10 ml CH +6 ml J	10ml CH + 8ml J	10 ml CH + 2 ml IF
[Cr] ppm at neutral pH	**500**	0.6	0.59	0.52	0.49	0.24
[Cr] ppm at pH = 9	**500**	0.29	0.27	0.30	0.30	0.22
Observation	**Absence of flocks**	**MIF**	**MAF**	**MAF**	**MAF**	**MAF**

Table 4: Formation of flocks and chromium removal based on cactus juice dosages: case of industrial effluent at 90 ppm Cr

Agents	Control : CH + IE	10 ml CH +2 ml J	10 ml CH +4 ml J	10 ml CH +6 ml J	10 ml CH + 8ml J	10 ml CH +2 ml IF
[Cr] ppm at neutral pH	**90**	0.61	0.57	0.60	0.61	0.55
[Cr] ppm at pH = 9	**90**	0.39	0.29	0.31	0.32	0.30
Observation	**Absence of flocks**	**MIF**	**MAF**	**MAF**	**MAF**	**MAF**

* IF: Industrial effluent

Table 5: Formation of flocks and chromium removal based on cactus juice dosages: case of industrial effluent at 900 ppm Cr

Agents	control : CH + IE	10 ml CH + 2 ml J	10 ml CH +4 ml J	10 ml CH +6 ml J	10 ml CH + 8ml J	10 ml CH +2 ml IF
[Cr] ppm at pH = 9	**900**	1.25	1.22	1.31	1.45	1.10
Observation	**Absence of flocks**	**MIF**	**MAF**	**MAF**	**MAF**	**MAF**

Treatment of the synthetic and the industrial solutions with the cactus juice dosages of 4-8 ml lead to very interesting results in term of clean up water clouding and developing clear large flocks of 2 to 4 mm in diameter. Same large flocs have been achieved when the industrial flocculent was used. Miller & al. **[20]** also found unclear large flocks that trigger easily the sedimentation process.

3.3. Reduction of turbidity

The decrease in turbidity was evaluated with same studied samples. Figure 1 shows that in neutral pH solution at 100 ppm and for a dosage of 8 ml of cactus juice, turbidity removal yield was about 97.6% and reaches 98.64% for a pH = 9 with 4 ml of cactus juice. The turbidity decreases by 98.25% with DK FLOC. The turbidity efficiencies for the synthetic solution at 500 ppm, with 6 ml of cactus juice at pH neutral and pH=9, and for DK FLOC were about 99%, 99.46% and 99.2%, respectively.

In the case of the industrial samples at 90 ppm, with 4 ml of cactus juice, turbidity removal efficiency reached 96.1% at pH neutral and to 97.6% at pH 9 against 97.45% for the referential flocculent. In addition, regardless the flocculent used, this efficiency exceeded 98% when the charge is as high as (500 ppm) (Figure 1).

In certain treatment, *Opuntia ficus indica* was evaluated as coagulant in the elimination of effluent turbidity **[12, 13, 20-24]**, as well as in the reduction of the bacteria **[14].**

Therefore, this bio-coagulant was considered as primary coagulant or as adjuvant coagulant **[3,15,25].**

In the present study, we evaluated *Opuntia ficus indica* as coagulant or flocculent and excellent turbidity reduction results have been achieved for both synthetic and industrial tannery solutions. Thus, the efficiency of the turbidity removal varies between (95% and 99.2%) for both cases, against (97% and 99%) when the DKFLOC was used. These results are in strong concordance with those obtained by Miller and al. [**20**], Shilpa and al. [**14**] and Kannadasan and al. [**25**].

3.4. Reduction in conductivity

As it was illustrated in the Figure 2, the conductivity in the synthetic solutions decreases significantly in both neutral and basic pH during coagulation-flocculation process using cactus juice as bio-flocculent. The conductivity decrease is lesser compared to the case of tanneries effluents.

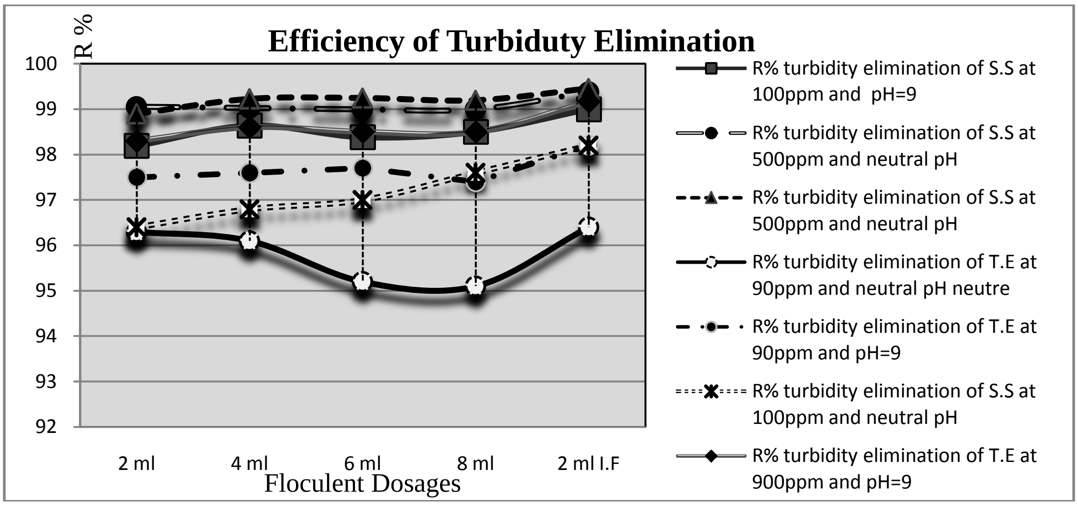

Figure 1: Reduction of turbidity in different polluted water using the coagulation and flocculation process with the cactus juice as bio-flocculent or DK as chemical flocculent.

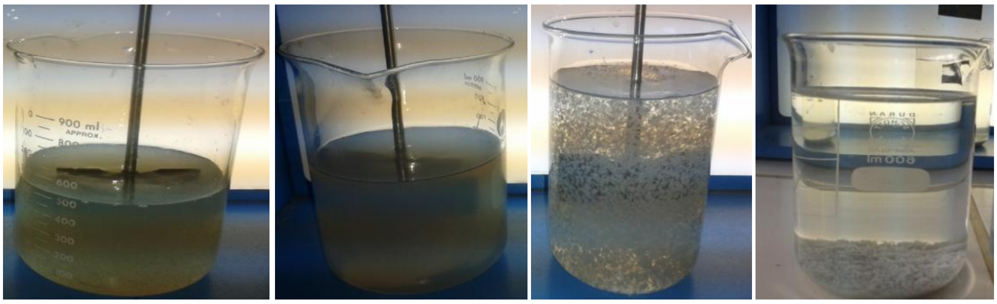

Photo 4 : Steps of coagulation-flocculation and sedimentation of the tannery effluent

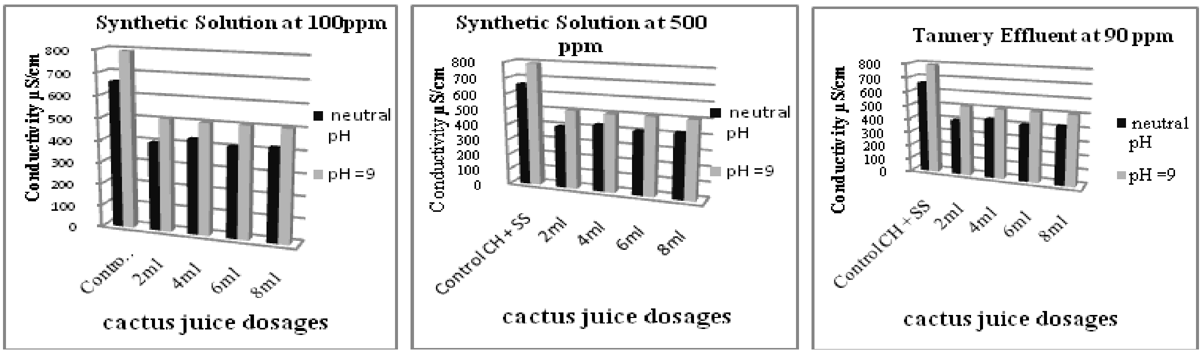

Figure 2: Variation of the conductivity in different polluted water treated by coagulation-flocculation process using the cactus juice as bio-flocculent.

In the absence of bio-flocculent, the conductivity obtained in a solution at 100 ppm is about 500 µS/cm, and it does not exceed 350 µS/cm with 8 ml of cactus juice at pH=9. In the case of a solution at 500 ppm, the conductivity stabilizes around 400 µS/cm at pH=3 and found of about 500 µS/cm at pH=9. These values stay often lower than that of the control samples that are set on 650 and 750µS/cm, respectively, in same pH conditions. When the industrial liquid is charged at 90 ppm, the conductivity values become less pronounced in the case of the effluent at 900 ppm, because at such high ppm, the coagulation-flocculation cannot be obtained without adjusting the pH values

3.5. Determination of chromium by ICP

Using ICP technique the total chromium removal values from different tested samples were found greatly important. Tables 6 to 8 show the average values of the chromium removal in the samples tested. The chromium concentration decreases strongly in both synthetic and effluent supernatant. Much better, there is a tendency toward a zero chromium values when we used the bio-flocculent in the treatment of either synthetic or effluent solutions at 90 ppm and at a pH neutral or pH=9. Same measurements was carried out on same effluent at 900 ppm, a residual concentration average of 0.319 mg/l obtained with the addition of 6 ml of 4 or cactus juice, and 0.566 mg/l was recorded with adding only 2 ml of the bio-flocculent .

Table 6: Changes in the chromium concentration, determined by ICP, in a synthetic solution at 100 ppm treated by coagulation flocculation process with the cactus juice as bio-flocculent

Agents	control : CH + SS	10 ml CH +2 ml J	10 ml CH + 4 ml	10 ml CH +6 ml J	10 ml CH + 8 ml J	10 ml CH +2 ml IF
[Cr] in mg/l at neutral pH	**112**	0.41 0	0. 320	0. 150	0. 144	0.093
[Cr] in mg/l at pH 9	**112**	0.018	0.009	0.015	0.015	<0.01

Table7: Changes in the chromium concentration, determined by ICP, in a synthetic solution at 500 ppm treated by coagulation flocculation process with the cactus juice as bio-flocculent

Agent	control: CH + SS	10 ml CH +2 ml J	10 ml CH +4 ml J	10ml CH +6 ml J	10ml CH + 8ml J	10ml CH + 2 ml IF
[Cr] in mg / l at neutral pH	**560**	0.280	0.562	0.271	0.170	0.155
[Cr] in mg / l at pH 9	**560**	0.013	0.056	0.021	0.018	0.076

Table8: Changes in the chromium concentration, determined by ICP, in a tannery effluent at 90 ppm treated by coagulation flocculation process with the cactus juice as bio-flocculent

Agents	control : CH + IE	10 ml CH +2 ml J	10 ml CH +4 ml J	10 ml CH+ 6 ml J	10 ml CH + 8ml.J	10 ml CH + 2 ml IF
[Cr] in mg / l at neutral pH	**176**	0.135	0.157	0.169	0.125	0.153
[Cr] in mg / l at pH 9	**176**	0.025	0.021	0.0035	0.037	0.026

Table 9: Changes in the chromium concentration, determined by ICP, in a tannery effluent at 900 ppm treated by coagulation flocculation process with the cactus juice as bio-flocculent

agents	control : CH + IE	10 ml CH +2 ml J	10 ml CH +4 ml J	10 ml CH +6 ml J	10 ml CH + 8 ml J	10 ml CH + 2 ml IF
[Cr] in mg / l at pH 9	**1189**	0.566	0.329	0.319	0.540	0.067

Efficiencies of the total chromium elimination in the case of synthetic solution reach 99.68% with the cactus juice dosage of 4 ml at pH neutral and slightly increase to 99.81% at pH=9, against 99.72% when the cactus juice is replaced by the industrial flocculent (DK) in same dosage conditions.
With the synthetic solution 500 ppm, efficiency of the total chromium elimination was also strongly high and exceeded 99.9% with a cactus juice dosage of 8 ml and with no pH adjustment and reached 99.96% with the addition of sodium hydroxide.
Now with the industrial solution 90 ppm, efficiency of the total chromium removal was around 99.37% with a cactus juice dosage of 4 ml with no pH adjustment and reached 99.68% with the addition of sodium hydroxide and same results as above obtained when we used the chemical flocculent (DK).
All tests carried out on both synthetic and industrial tannery solutions illustrate that the cactus juice exhibits great flocculation ability in the presence of lime alone or mixed with the soda. After the neutralization and coagulation with the lime, the bio-flocculent used here lead to a great chromium elimination value of 0.2 ppm which is comparable to 0.3 ppm obtained when the industrial flocculent has been used. Our results are also in concordance with what Abid and al. [24] found as chromium elimination value of (0.4 ppm). On the other hand, the efficiency of the chromium removal reported in those works varies from 98.50 to 99.50% versus an efficiency ranged between 99.3 and 99.9% when we used the cactus juice as bio-flocculent. While the ICP technique has been developed for dosing the heavy metals, to our knowledge, it has been never used in the

chromium elimination dosage. Therefore, in order to confirm the previous results, we have assessed the chromium elimination dosage using this technique and we have achieved an efficiency of 99.9%.

Conclusion

1. In this work, a bio-flocculent extracted from the racket of *Opuntia ficus indica* has been used and it showed a great efficiency in the removal of the total chromium discharged in the tannery effluent.
2. The flocculation was found rapid in the range of pH 7-9
3. The bio-flocculent provided great COD reduction efficiencies.
4. Excellent turbidity reduction results have been achieved for both synthetic and industrial tannery solutions.
5. The appropriate ICP's measurement confirmed that the metal charge values have been greatly reduced by adding NaOH or lime's coagulation to cactus juice flocculation.
6. The bio-flocculent used in this study is biodegradable and available all across the country, would be a good alternative for the treatment of polluted waters especially for those containing heavy metals such as the chromium discharged in the tannery effluent.

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