

Coagulation–adsorption–ultrafiltration for wastewater treatment and reuse

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Received 3 August 2000; accepted 16 August 2000

Abstract

We considered the treatment of domestic wastewater by coagulation–adsorption coupling with ultrafiltration as an alternative for wastewater reclamation and reuse. The flocculation–adsorption process showed good performance in removing the chemical oxygen demand 86% at a FeCl_3 concentration of 40 mg/l, and powdered activated carbon concentration of 20 mg/l. In the same way the turbidity passed from 18 NTU to 3.5 NTU at pH = 5.5. Laboratory tests showed that the turbidity is passed from 18 NTU to 0.5 NTU by the process flocculation–adsorption–ultrafiltration and removal efficiency of chemical oxygen demand 82.7% (where the value passed from 77 to 13mg/l).

Keywords: Coagulation; Adsorption; Ultrafiltration; Wastewater; Reuse; Organic matter

1. Introduction

Industrialisation and urbanisation have accelerated pollution in the water environment, making water a limited resource. Recycled wastewater can reduce stress on the environment as well. Wastewater can be an alternative water source which can reduce the demands for fresh water. Membrane separation (ultrafiltration or microfiltration) increasingly used in the field of

water and wastewater treatment is adequate for producing disinfected clear water suited for different kinds of applications. However, fouling of microfiltration or ultrafiltration membranes is the main limitation. Coagulation and adsorption permit removal of organic colloids which play an important role in fouling phenomena. Al-Malack et al. [1] have shown the effect of using alum, polyaluminium silicate sulfate (PASS), and lime as coagulants on the performance of cross-flow microfiltration of domestic wastewater. The

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coagulants were added to the circulation tank of the experimental set up at the beginning of each run. Doses of 20 to 120 mg/l of alum were investigated at a pH of 7. The results showed a 50% improvement in flux values with regard to direct filtration without a coagulant.

Seo et al. [2] are interested in the coupling of biological powdered activated carbon (BPAC)–microfiltration for wastewater reclamation and reuse. The average organic removal efficiency was 83% for an initial concentration of the effluent in total organic carbon (TOC) equal to 9.8 mg/l. Maleriat et al. [3] considered natural agents of combined flocculation to the cross-flow filtration. The reduction of the turbidity and matters in abeyance are important by coupling. The coupling of flocculation and microfiltration for a synthetic solution (bentonite) has been studied by Mieuton-Peuchot and Ben Aim [4]. A cellulose membrane of a pore size of 0.2 μm (WAC) was used as the flocculant. Their results show that the coupling clearly improves the flowrate of filtration and gives an excellent quality of filtrate. S. Vigneswaran et al. [5] showed that the cross flow microfiltration with in line flocculation reduces the clogging of membranes thus leading to high quality product water at an economic filtration rate. The filtration rate can be increased by more than 200% by adopting in line flocculation.

Table 1

Middle characteristics of the secondary effluent during the period May–June–July 99

Temperature, °C	18
pH	7.4
Turbidity, NTU	18
SS, mg/l	35
BOD ₅ , mg/l	30
COD, mg/l	77
Conductivity, $\mu\text{s/cm}$	1350
Zeta potential, mV	4.118

The goal of this work is to study the elimination of organic matter still present in the secondary effluents. We have thus studied different processes for removal of organic matter, such as coagulation, adsorption on powdered activated carbon and ultrafiltration. We will study the evolution of the residual turbidity, the chemical oxygen demand, and the Zeta potential in different concentrations of the FeCl_3 coagulant and the powder activated carbon for different pH values.

2. Experimental conditions

Tests have been made on a secondary effluent of the treatment station of Staoueli (Algeria).

2.1. Characteristics of the raw water

The characteristics of the effluent are given in Table 1.

2.2. Equipment

Ultrafiltration is achieved in a cell using an Amicon laboratory in a static mode. The active surface of the membrane is 26.4 cm^2 . We used an organic membrane with a 50,000 Dalton molecular weight cut off. The shearing to the surface of the membrane is assured by a magnetic agitator of a speed of 250 tr/min rotation, which permits a better homogenisation of the solution at the time of the addition of FeCl_3 and powdered activated carbon, and the reduction of the gradient of concentration near the membrane and fouling effects. The total volume of the cell is 180 ml. A circuit of nitrogen permits maintenance of the transmembrane pressure at $\Delta P = 1$ bar.

The characteristics of the membrane are reported in Table 2.

Table 2
Characteristics of the membrane

Membrane	Process	Classification	Cut off	Material
Tech Sep	UF	IRIS 3042	50000 Da	Acrylonitrile

2.3. Analysis methods

Classical methods for determination of retained parameters have been used [6] (AFNOR standard). The turbidity has been measured with the help of the HACH turbidimeter, model 2000 and calibrated. The pH has been measured with the help of a pH meter calibrated, CRISON DEIT 501 type; the Zeta potential is deduced from the measure of the electrophoretic mobility determined with the help of mass transfer analyser Micromeritics 1202 type. The chemical oxygen demand has been measured with AFNOR method (T90-101). The coagulant used is $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$ with a molecular mass of 270.3 g/mole. The powdered activated carbon PAC (anticromos) obtained from Ceca Italiana, the BET surface area is approximately 700 m^2/g . The iodine number is about 760 mg/g and the humidity is 15.6%. The powdered activated carbon has been rinsed with the distilled water, boiled during 3h in distilled water, then dried at 120°C during 4h and stocked inside a desiccator.

2.4. Experimental procedures

Step 1: Variable doses of the coagulant FeCl_3 are added (10 to 50 mg/l) to different values of pH between 4 and 8. This last is adjusted with 0.1N HCl or 0.1N NaOH, before addition of the coagulant. The coagulation–flocculation is achieved in a Jar Test while observing perikinetic phase (fast agitation: 250 tr/min) for 1 min, orthokinetic phase (slow agitation: 60 tr/min during 15 min), and a phase of settling during 45 min, the optimal conditions of coagulation

were determined: two optimal doses of coagulant to pH corresponding to the best elimination of the turbidity as well as the organic matter chemical oxygen demand (COD).

Step 2: We added powdered activated carbon to the coagulant and proceeded to a coagulation flocculation, to the previously determined optimal conditions.

Step 3: We introduced directly the variable doses of coagulant FeCl_3 (10–50 mg/l) inside the AMICON cell with ultrafiltration under agitation in a static mode.

Step 4: We added the variable doses of powdered activated carbon (PAC) with the optimal dose of coagulant obtained previously and proceeded to ultrafiltration.

3. Results

3.1. Determination of the coagulation optimal conditions

3.1.1. Turbidity and COD reductions

Fig. 1 shows the variation of turbidity and the COD as a function of coagulant concentrations (FeCl_3) at different pH values.

Fig. 1a showed that low turbidity is obtained at pH = 6.5. It can be seen that the best removal efficiency 66.1% occurs in two sets of conditions pH = 5.5 with FeCl_3 concentration $C = 40$ mg/l where the value of the COD passes from 77 mg/l to 26 mg/l and pH = 6 for a concentration $C = 30$ mg/l. A residual COD of 20.8 mg/l with a removal efficiency of 73% as shown in Fig. 1b. According to these results, we have pursued the Jar Test with powdered activated carbon at the optimal coagulation conditions previously determined.

3.1.2. Variation of the Zeta potential

Fig. 2 shows variations of the Zeta potential, according to the concentration of coagulant FeCl_3 .

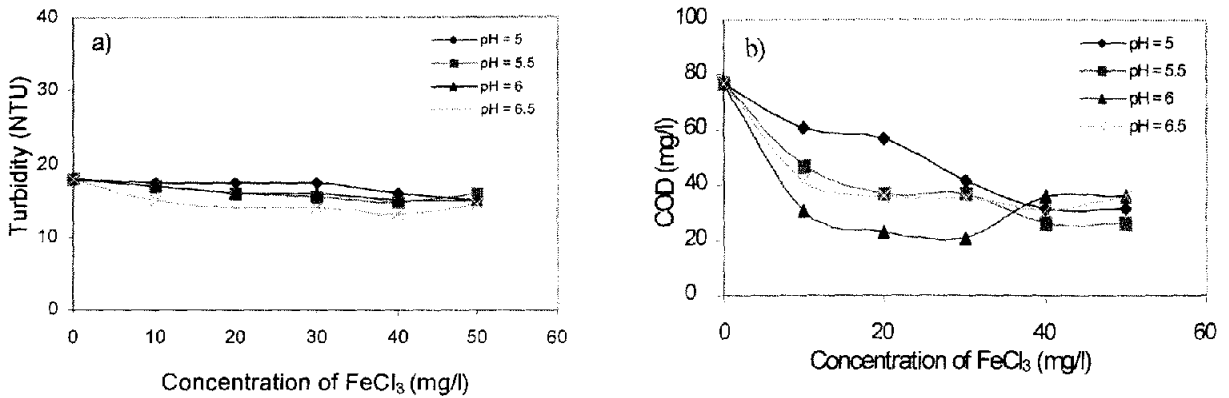


Fig. 1. Test of coagulation-settling in the Jar Test: removal efficiency of (a) turbidity and (b) COD.

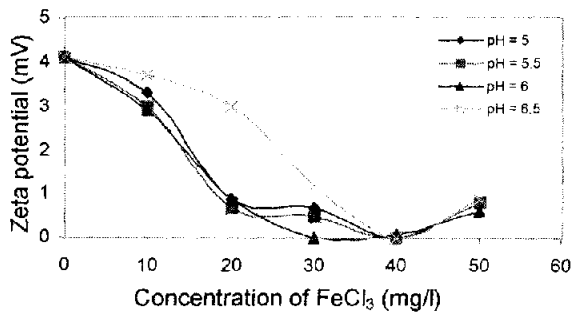


Fig. 2. Zeta potential as a function of coagulant FeCl₃ doses at different pH values.

One observes the inversion of the Zeta potential near to the optimal concentrations of the coagulant FeCl₃ (pH = 5.5 FeCl₃ concentration C = 40 mg/l; pH = 6 for a concentration C = 30 mg/l).

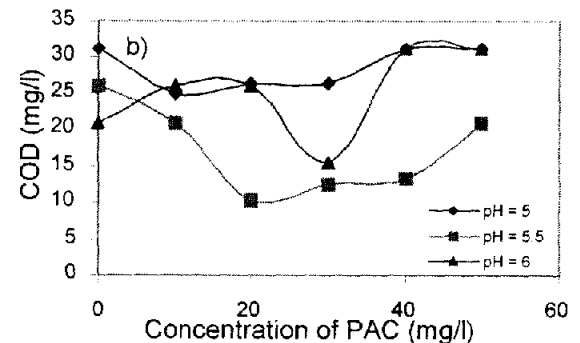
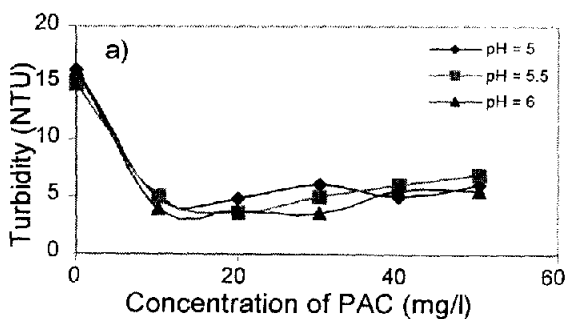


Fig. 3. Test of coagulation-settling-adsorption in Jar Test: removal efficiency of (a) turbidity and (b) COD.

Once the optimal dose coagulant had been obtained, we tried to determine optimal conditions of the adsorption with the powdered activated carbon.

3.2. Determination of optimal conditions for coagulation/adsorption

On the basis of previous results, we determine the optimal conditions for adsorption with the powdered activated carbon.

3.2.1. Turbidity and COD reductions

The turbidity passes from 14.75 NTU to 3.5 NTU (Fig. 3a) with the optimal concentrations in FeCl₃ equal to 40 mg/l and in powder activated carbon equal to 20 mg/l. Results show in Fig. 3b a

residual chemical oxygen demand of 10.4 mg/l, with the removal efficiency of 86.5% at pH = 5.5 and by way of consequence 1. At pH = 6, we got a COD of 15.6 mg/l with a 79.74% elimination of a powdered activated carbon concentration equal to 30 mg/l.

From these results we conclude that the adsorbant the powdered activated carbon permits a reduction of the organic matter.

3.2.2. Variation of the Zeta potential

The variation of the Zeta potential (Fig. 4) is 4 mV. At the start, we note a fall in Zeta potential. This last confers to these particles their stability, then annuls near the optimal concentration $C = 30$ mg/l of powder activated carbon at pH = 5.5 and $C = 40$ mg/l of powder activated carbon at pH = 6.

3.3. Determination of optimal conditions for coagulation/ultrafiltration

For the works following, we retain two values of pH (5.5, 6) for the variable doses of FeCl_3 (10–50 mg/l).

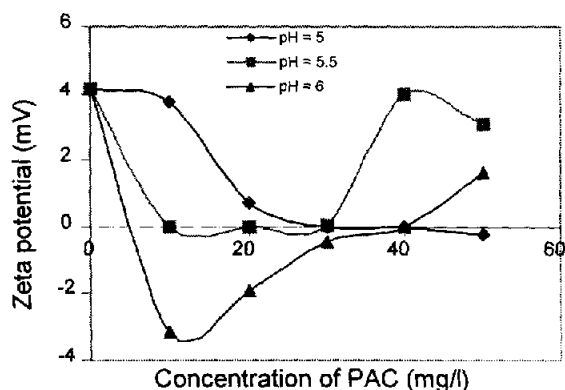


Fig. 4. Zeta potential as a function of powdered activated carbon concentration at different pH values for FeCl_3 concentration = 30 mg/l.

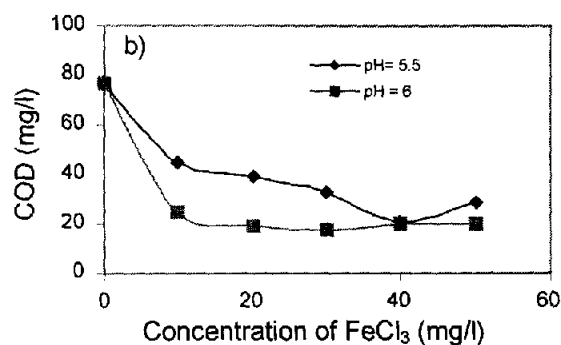
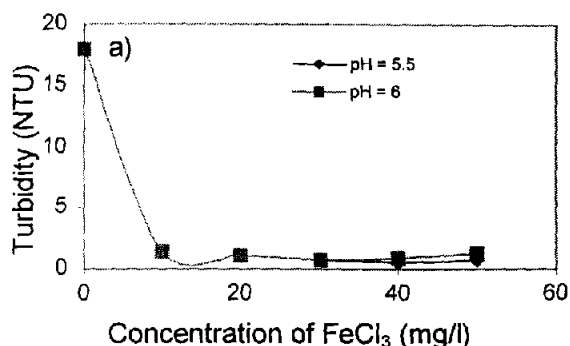


Fig. 5. Test of coagulation–ultrafiltration: removal efficiency of (a) turbidity and (b) COD (b) at pH = 5.5 and pH = 6.

3.3.1. Turbidity and COD reductions

Results show almost the same values of turbidity 0.5–0.8 NTU for the two pH (Fig. 5a). At pH = 5.5, an optimal dose in FeCl_3 equal to 40 mg/l corresponding to a COD of 21 mg/l and for a pH = 6, the optimal concentration is 30 mg/l in FeCl_3 with a COD of 17.50 mg/l (Fig. 5b).

3.3.2. Variation of Zeta potential

We note a slight variation of the Zeta potential otherwise as shown in Fig. 6.

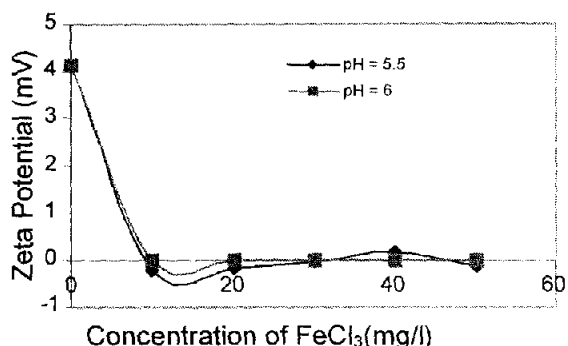


Fig. 6. Zeta potential as a function of coagulant FeCl₃ concentration (tests of coagulation-ultrafiltration).

3.4. Determination of the coagulation adsorption/ultrafiltration optimal conditions

3.4.1. Turbidity and COD reduction

Once the optimal conditions in FeCl₃ have reached C = 40 mg/l at pH = 5.5 and C = 30 mg/l at pH = 6 during the coagulation-ultrafiltration, we introduced the variable doses of powdered activated carbon in the ultrafiltration cell.

By this process, we note that the best elimination of the chemical oxygen demand (13.33 mg/l) is obtained at pH = 5.5 (Fig. 7b). We got identical values concerning the turbidity, that is 0.5 NTU for pH=5.5 and 6 (Fig. 7a).

3.4.2. Variation of Zeta potential

The optimal doses for powdered activated carbon obtained for minimal turbidity and minimal COD are confirmed by the variation of the Zeta potential according to the concentration in powdered activated carbon represented in Fig. 8.

3.5. Evolution of COD and turbidity for different processes

Fig. 9 compares the water quality obtained for the different processes tested.

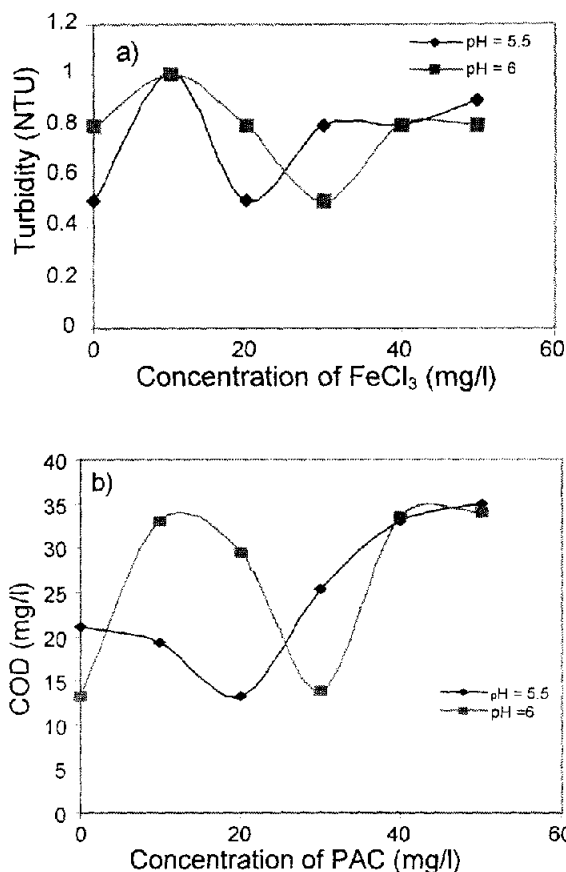


Fig. 7. Test of coagulation-adsorption-ultrafiltration: removal efficiency of (a) turbidity and (b) COD at pH = 5.5 and pH = 6.

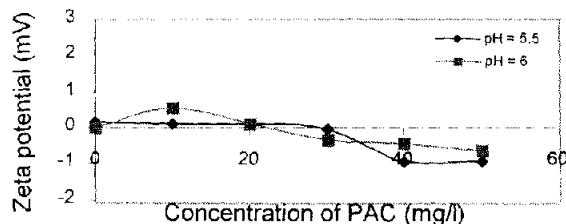


Fig. 8. Variation of Zeta potential as a function of PAC concentration.

The best result was obtained by coupling FeCl₃-PAC (coagulation-adsorption).

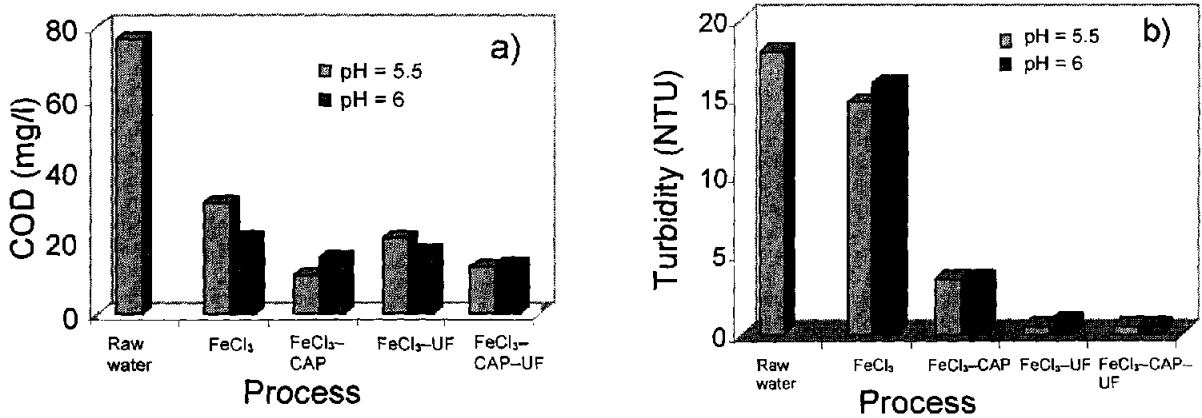


Fig. 9. Evolution of (a) COD and (b) turbidity as a function of the treatment process.

3.6. Influence of the coagulant concentration either of the powdered activated carbon on the efficiency of COD at pH = 5.5

The removal efficiency of COD by coagulation is strongly dependent on the coagulant dose (Fig. 10), it passes from 21 to 59% for 10 to 50 mg/l doses. On the other hand, the efficiency of the coupling coagulation–adsorption varies slightly with the dose of powdered activated

carbon.

The efficiency of the coupling coagulation–ultrafiltration increases between 10 and 40 mg/l of coagulant, then decreases slightly.

The process coagulation–adsorption–ultrafiltration is more efficient (75%) for 10 mg/l powdered activated carbon dose. This efficiency decreases when the concentration of powdered activated carbon increases.

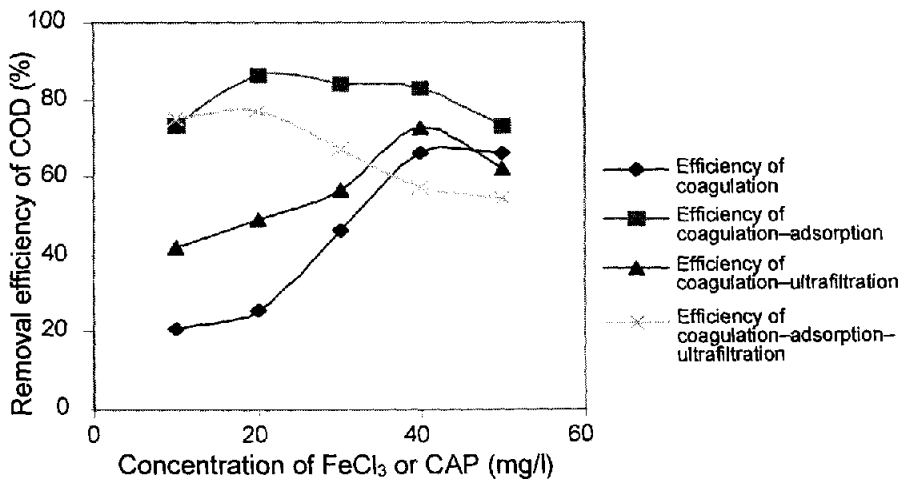


Fig. 10. Removal efficiency of COD evolution as a function of coagulant dose FeCl₃ or PAC at pH = 5.5.

Table 3
Comparison of results between the two methods of treatment

Method	Jar Test				Ultrafiltration			
	5.5	5.5	6	6	5.5	5.5	6	6
Optimal pH	5.5	5.5	6	6	5.5	5.5	6	6
Reagent	FeCl ₃	FeCl ₃ /PAC	FeCl ₃	FeCl ₃ /PAC	FeCl ₃	FeCl ₃ /PAC	FeCl ₃	FeCl ₃ /PAC
Optimal dose, mg/l	40	40/20	30	30/30	40	40/20	30	30/30
Turbidity, NTU	14.75	3.50	16.00	3.50	0.50	0.50	0.80	0.50
COD, mg/l	26	10.40	20.80	15.60	21.00	13.30	17.50	13.80
Zeta potential, mV	2.265	0	-1.247	-0.471	0.166	0.110	0	-0.332

3.7. Evolution of parameters according to the process of treatment

Table 3 summarises the results obtained. The removal efficiency of the chemical oxygen demand for the process of coagulation–adsorption is 86% and by way of consequence, the turbidity is reduced from 18 to 3.5 NTU for the coagulant and adsorbant optimal doses 40mg/l of FeCl₃ and 20 mg/l of PAC. Otherwise, the process of coagulation–ultrafiltration is not as efficient as the process of coagulation–adsorption with a percent elimination of the organic matter about 75% (low value 17.5 mg/l). On the other hand, the turbidity is reduced from 3.5 to 0.5 NTU.

4. Conclusions

In this study, we have determined the optimal conditions of tertiary treatment by coagulation–settling–adsorption–ultrafiltration of a secondary effluent. Our goal has been to specify the efficiency of the alternative treatment plant for a reclamation wastewater and reuse. We observe that the best result of the residual COD is obtained for the coupling of coagulation–adsorption, either a value of 10.4 mg/l for a pH = 5.5 and for

concentrations while coagulating FeCl₃ equal to 40 mg/l and in powdered activated carbon equal to 20 mg/l and a compliant turbidity to norms of reuse.

The process of coagulation–ultrafiltration is not very efficient, probably because the ultrafiltration membrane does not retain the low organic molecules mass less than 50,000 Dalton.

References

- [1] M.H. Al-Malack and G.K. Anderson, *J. Membr. Sci.*, 121 (1996) 59.
- [2] G.T. Seo, Y. Suzuki and S. Ohgaki, *Desalination*, 106 (1996) 39.
- [3] M. Peuchot and R. Ben Aim, *Couplage de la floculation et de la microfiltration tangentielle, Récents progrès en Génie des Procédés*, Lavoisier, Toulouse, Vol. 8a, 1989, pp. 313–319.
- [4] J.P. Maleriat, D. Trebouet, P. Jaouen and F. Quemeneur, *Proc., 7th World Filtration Congress, Hungary*, 1–2 (1990) 456.
- [5] S. Vigneswaran and S. Boonthanon, *Cross flow microfiltration with in line flocculation. Water technology*, 1992, pp. 29–31.
- [6] J. Rodier, *Traitement de l'eau*, Lavoisier, Paris, 1992.