DESALINATION

Desalination 152 (2002) 367–373

www.elsevier.com/locate/desal

Treatment of primary effluent by coagulation–adsorption–ultrafiltration for reuse

D. Abdessemed*, G. Nezzal

Faculté de Génie Mécanique-Génie des Procédés–Environnement, Département de Génie de l'Evironnement, Université des Sciences et de la Technologie Houari Boumediène, B.P.32 El Alia 16111, Bab Ezzouar, Alger, Algerie Tel. +213 (21) 247169; Fax +213 (21) 758158/525863; email: adjamal@yahoo.com

Received 30 March 2002; accepted 15 April 2002

Abstract

This study aims to find news a die of purification for a wastewater reuse. Primary effluent contains organic mineral, dissolved and suspended matter (colloids). Microfiltration or ultrafiltration is adequate for producing disinfected clear water suited for different applications. However, direct filtration on membrane is limited by the fouling phenomena which leads during filtration to constant pressure, to a strong and continuous decrease of the permeate flux. On the other hand, coagulation and adsorption make it possible to removal the colloidal fraction, which plays a significant role in membrane fouling. We considered the treatment of primary effluent by coagulation–adsorption–ultrafiltration for reuse with ultrafiltration membranes. Tests have been performed on the primary effluent of the wastewater treatment plant of Staoueli, Algeria with the average characteristics in the chemical oxygen demand (COD) of 165 mg of O_2/l, and turbidity 90 NTU. The ultrafiltration tests have been made on mineral membranes tubular CARBOSEP M5 (10 kDa), M2 (15 kDa), in dynamic mode with a transmembrane pressure ΔP = 1 bar, cross flow velocity U = 3 m/s. The reagents used $FeCl_3$ (as a coagulant agent) and powder activated carbon (PAC) as an adsorbent agent. In the first step, the optimal conditions were determined for coagulation and adsorption corresponding to the best elimination of turbidity as well as organic matter. In the second step, the efficiency of different processes (coagulation, adsorption, ultrafiltration) was compared when used alone or combined. The coagulation test showed a COD value equal to 23 mg/l at pH = 5.5 for $FeCl_3$, concentration 120 mg/l and a final turbidity equal to 12 NTU. For the process coagulation–adsorption–ultrafiltration, we obtained the best COD final value of 7 mg/l. The best value of the permeate was obtained for the coupling coagulation–ultrafiltration at coagulant concentration equal to 80 mg/l and residual COD equal to 13 mg/l for M2 membrane. Coagulation significantly improves the ultrafiltration performances. The coupling makes it possible to reduce the regeneration membranes.

Keywords: Primary effluent; Coagulation; Adsorption; Ultrafiltration; Organic matter; Reuse

*Corresponding author.

Presented at the EuroMed 2002 conference on Desalination Strategies in South Mediterranean Countries: Cooperation between Mediterranean Countries of Europe and the Southern Rim of the Mediterranean. Sponsored by the European Desalination Society and Alexandria University Desalination Studies and Technology Center, Sharm El Sheikh, Egypt, May 4–6, 2002.

1. Introduction

In view of deficiency of the water resources in Algeria, the wastewater treatment and reuse permits, on the one hand, to protect the environment and, on the other hand, to bring fertilizers to the cultures while reducing or eliminating employment of chemical fertilizers. The new techniques known as separation by membranes are applied for water treatment.

Membrane processes (microfiltration and ultrafiltration) are effective, but the fouling of the membranes is the main limitation. The processes coagulation and adsorption permits to modify the colloidal fraction, which plays a significant role in fouling.

Ademoroti [1] proceeded to the coagulation followed by sand filtration using aluminium sulphate and ferric chloride on a raw wastewater. Percentages of reduction are 83.3 for turbidity, 75 for BOD and 78% for the COD for a concentration of $FeCl_3$ equal to 300 mg/l and 400 mg/l of $Al_2(SO_4)_3$ with an optimum pH = 4.8.

Al Malack and Anderson [2] determined the coagulation optimal conditions of raw wastewater with a concentration of 200 mg/l $FeCl_3$ at pH = 9, the removal efficiency of turbidity and the chemical oxygen demand are respectively 99.6 and 99.3%.

Aguiar et al. [3] studied the organic matter removal from colored water by coagulation using ferric chloride. The results showed that the optimum pH by coagulation is between 4 and 5 for an elimination of the organic matter. Whatever the origin of the raw water, the optimal dose of coagulant was 2.1±0.2 mg Fe per mg of TOC. The same results were obtained with fulvic acid solutions. The coagulation process allowed a 60–80% elimination of the total organic carbon.

Pouet [4] studied the microfiltration of urban wastewater. The principal disadvantage of this process is the membrane fouling, often irreversible. She examined the role of various organic fractions (settable, colloids, soluble) in the membrane fouling. The chemical destabilization can reduce the hydraulic resistance of the membrane by a factor from 10 to 30 according to the wastewater nature. An optimal treatment was determined with 700 mg/l of $Al_2(SO_4)_3$ by eliminating the majority of colloids and the settable fractions, while minimizing the hydraulic resistance of the membrane.

Vigneswaran and Boonthanon [5] studied the coupling of crossflow microfiltration with inline flocculation for a bentonite aqueous suspension with an aim to reduce the membrane fouling. The filtration rate can be increased by more than 200% by adopting in line flocculation in the presence of aluminium polychloride.

A comparative study of coupling of flocculation and microfiltration for a suspension solution (bentonite) has been carried out by Mieuton-Peuchot and Ben Aim [6]. The membrane used was cellulose with a pore size of 0.2 µm. Aluminium polychloride (WAC) was used as the flocculant. Their results show that the coupling improves the flowrate of filtration and gives an excellent quality of filtrate. The optimal dose of 150 mg/l in WAC makes it possible to obtain the residual turbidity of about 0.1 NTU.

Ngo et al. [7] showed that more than 85% of fulvic acids was removed by using the microfiltration–adsorption hybrid system for the powdered activated carbon with concentration equal to 260 mg/l with Millipore membrane of 0.22 µm. The average steady permeate flux increased from 707.5 to 816.3 l/hm^2 with the increase in the concentration of PAC from 100 to 260 mg/l.

Lin et al. [8] showed that the use of the powder activated carbon, like pretreatment with ultrafiltration or combination, allowed the fouling membrane reduction. The powdered activated carbon is ineffective for solutes of molecular weights less than 300 and more than 17,000 Da.

In reference to these works, the present study relates to the treatment of a primary effluent by coagulation adsorption ultrafiltration for its reuse, with mineral ultrafiltration membranes (15,000 Da, 10,000 Da).

The following parameters are considered: COD, turbidity and permeate flux J_v for the four processes.

2. Experimental conditions

2.1. Raw water characteristics

The experimental study was carried on a primary effluent at the wastewater treatment plant of Staoueli whose average characteristics are given in Table 1.

Table 1
Average characteristics of the primary effluent

Parameters	Primary effluent
Temperature, ºC	22
pH	8.1
Turbidity, NTU	90
COD, mg of O_2/L	165

2.2. Experimental procedures

Step 1: Variable doses of the coagulant ($FeCl_3$) are added to the effluent (20–150 mg/l) at different values of pH between 5 and 6.5. This last is adjusted as a preliminary with 0.1 N HCl. The coagulation–flocculation is achieved in a jar test while observing perikinetic phase (fast agitation 250 tr/min) for 2 min, followed by 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 corresponding to the best elimination of the organic matter COD under normal atmospheric pressure.

Step 2: We added powdered activated carbon and proceeded at the jar test.

Step 3: Variable doses of coagulant $FeCl_3$ (10–50 mg/l) were introduced directly inside the feed tank of the experimental set up.

Step 4: We added the optimal dose of coagulant obtained previously with the dose of powdered activated carbon (PAC) and proceeded to ultra-filtration.

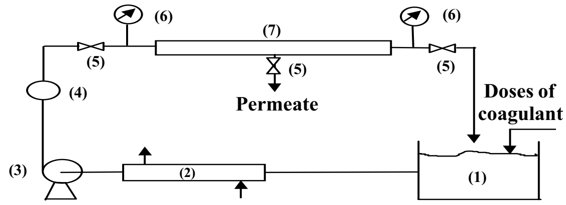

Fig. 1. The experimental setup. 1, feed tank; 2, cooling coil; 3, centrifugal pump; 4, flowmeter; 5, valves; 6, pressure gauge; 7, tubular UF module.

2.3. Experimental set up (Fig. 1)

A tubular inorganic membranes CARBOSEP M5 and M2 with respectively 10 kDa 15 kDa MWCO have been used (porous carbon support and membrane layer of ZrO_2). The transmembrane pressure $\Delta P = 1$ bar, the crossflow velocity $U = 3$ m/s, temperature 298 K.

2.4. Characteristics of powdered activated carbon

The powdered activated carbon (antichromos) was obtained from Ceca Italiana. Its characteristics are given in Table 2. The powdered activated carbon has been rinsed with the distilled water, boiled during 3 h in distilled water, then dried at 393 K during 4 h and stocked inside desiccated.

Table 2
Characteristics of powdered activated carbon

Characteristics	Description
BET surface area, m^2/g	600–800
Iodine number, mg/g	760
Humidity, %	15.6
Density, g/cm^3	0.41
Granulometry (refusal on a sieve of opening of 20 μm), %	85

2.5. Regeneration

The chemical protocol of cleaning adopted for the two membranes is as follows: rinsing with distilled water during 10 min, cleaning with

NaOH at pH = 12 and 70°C during 30 min, rinsing with water during 30 min, addition of sulphuric acid (pH = 2) during 15 min, rinsing with water during 20 min. The hydraulic permeability of the membrane remained appreciably constant during the carried out tests (between 94 and 100% of the initial value) for M5 and M2. The values of the initial hydraulic permeability are 45 l/hm^2 bar for M5 and of 72 l/hm^2 bar for M2.

2.6. Methods of analysis

Classical methods for determination of retained parameters have been used (AFNOR standard). The turbidity has been measured with the help of the Hach turbidimeter, model 2100 A, and calibrated. The pH has been measured with the help of a pH meter calibrated CRISON DEIT 501 type. The chemical oxygen demand has been measured with AFNOR method (T90-101).

3. Results and discussion

3.1. Determination of the coagulation optimal conditions

3.1.1. Turbidity and COD reduction

Fig. 2 shows the variation of turbidity and COD as a function of coagulant concentrations ($FeCl_3$) at different pH values. It can be seen that the low value of turbidity is 12 NTU, and the minimal value COD = 23 mg/l is obtained at the pH 5.5 for the concentration of $FeCl_3$ corresponding to 120 mg/l.

Municipal effluents contain colloidal suspensions generally carrying some negative charge. When the coagulant agent is dissolved in the water, the ion forms monomeric and polymeric ferric species, the formation of which is highly pH dependant: $Fe(OH)^{2+}$, $Fe(OH)_2^{+}$, $Fe_2(OH)_4^{2+}$, $Fe_3(OH)_4^{5+}$, neutral $Fe(OH)_3$ and negatively charged $Fe(OH)_4^{-}$.

All the curves have a similar pattern at the start. We note a fall in turbidity corresponding to a destabilization of the particles (first phase), then to stabilization, followed by a neutralization of the charge leading to precipitation (second phase). We notice that the turbidity increases as a result of stabilization of the particles (third phase) accounted fir by the fact that too high coagulant dose leads to a reduction of the efficiency of coagulation–flocculation process.

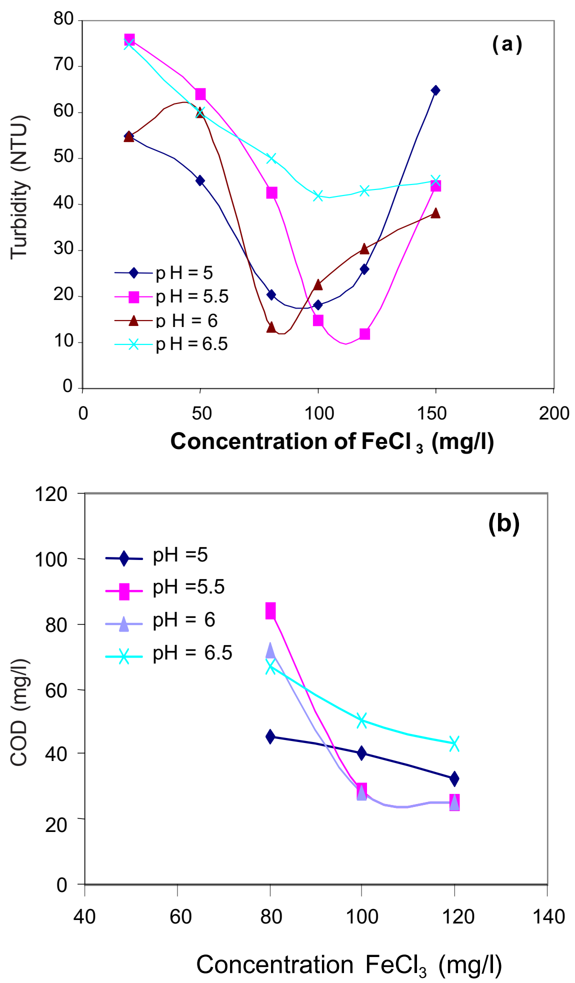

Fig. 2. Test of coagulation settling in the jar test: variation of (a) turbidity and (b) COD concentration with coagulant $FeCl_3$.

3.2. Determination of the adsorption optimal conditions

3.2.1. Turbidity and COD reduction

The powdered activated carbon addition

contributes to the elimination of the organic matter with the optimal concentration will then be determined as $C = 40$ mg/l as shown in Fig. 3b, where the turbidity and COD values are respectively 20 NTU and 57 mg/l.

3.3. The permeate flux — its evolution with time

The follow-up of the evolution of permeate flux according to the time for a crossflow velocity $U = 3$ m/s for different coagulant concentrations (from 0 mg/l to 120 mg/l) and only one concentration of powdered activated carbon (40 mg/l) is represented in Fig. 4.

Results reveal that for M2 membrane, $FeCl_3$ addition as the coagulant was a beneficial contribution for the limit permeate flux value which passed from 49 (ultrafiltration alone) to 71 l/h.m^2 (optimal concentration of $FeCl_3$) either an increase of 46.6%. With the presence of powdered activated carbon, we obtained a value of 59,8 l/h.m^2 or an increase by 22.2%.

We note that the presence of the coagulant is more beneficial than the adsorbent for the permeate flux improvement. This increase in the permeate flux is due to the flocs formation, whose dimensions do not permit the fouling of the pores. The best value of the permeate flux was obtained for the coagulant concentration equal to 80 mg/l for membranes M2 and M5.

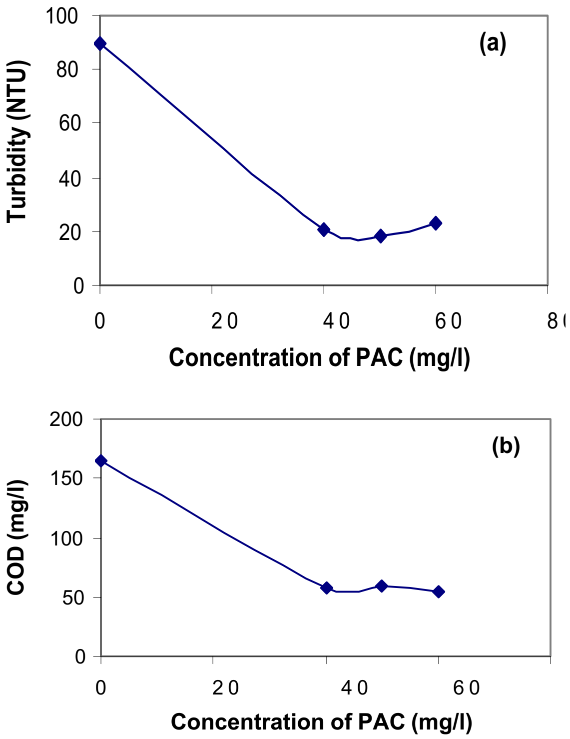

Fig. 3. Test of adsorption settling in the jar test: evolution of (a) turbidity and (b) COD concentration adsorbent .

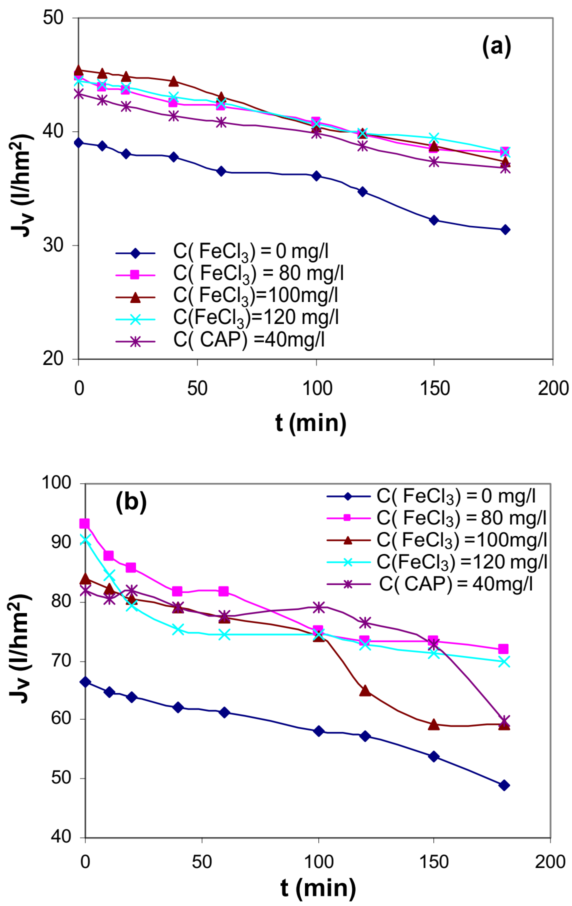

Fig. 4. Variation of the permeate flux with ultrafiltration time for different concentrations of coagulant and adsorbent for (a) M5 and (b) M2.

3.4. Variation of turbidity with time

Fig. 5 reveals a sharp reduction in the turbidity for membranes M2 and M5 — 99% of turbidity is eliminated in the first 60 min. This established fact is explained by the great capacity of the membranes to retain the suspended matter responsible for turbidity in water.

3.5. Variation of chemical oxygen demand for different processes

Fig. 6 compares the water quality obtained for the different processes tested. The best result was obtained by the process of coagulation–adsorption–ultrafiltration, with COD final value of 7 mg/l for M2 membrane and 20 mg/l for M5 membrane.

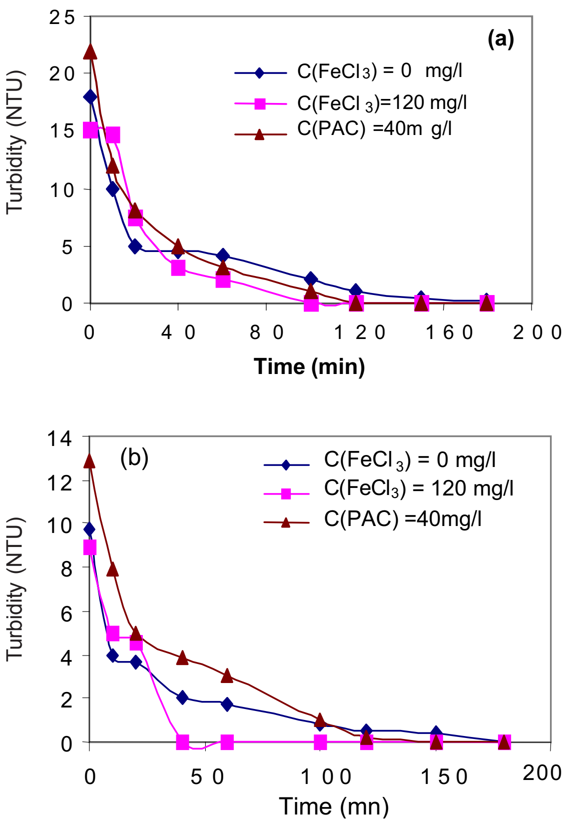

Fig. 5. Variation of turbidity according to the time of ultrafiltration for (a) M5 and (b) M2 for various concentrations of the reagent.

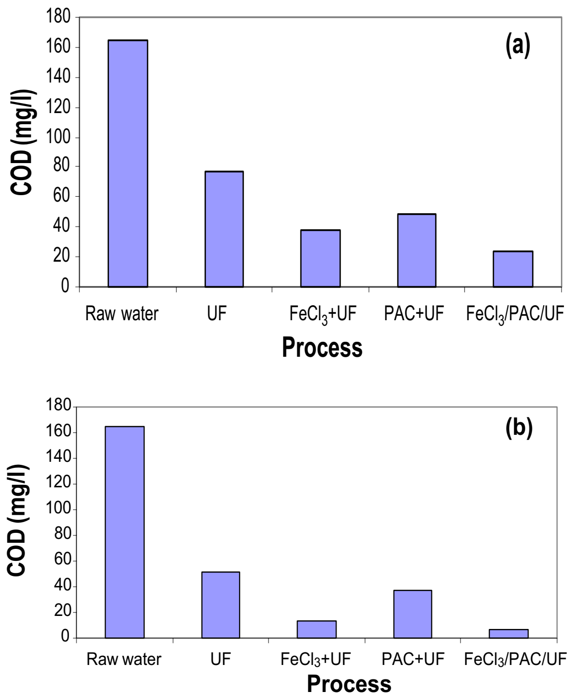

Fig. 6. Evolution of COD as a function of the treatment process (a) for M5 and (b) for M2.

4. Conclusions

The study made it possible to determine the optimal primary conditions by coagulation for effluent with pH = 5.5 and one concentration in coagulant $C = 120$ mg/l. Coagulation improves the permeate flux in a very significant way. The coupling makes it possible to modify the configuration of the deposit on the surface of the membrane by the formation of larger particles of greater dimensions (flocs). It permits too the reduction of membrane regeneration.

We observed a significant reduction of COD, which decreases on average from 165 to 40 mg/l under jar test; 20 mg/l under the test of coupling coagulation or adsorption ultrafiltration; 7 mg/l under the test of combining coagulation–adsorption–ultrafiltration.

We observed partial or total elimination of turbidity, according to cases, which decreases from 90 to 13.5 NTU under jar test, with 0 NTU under the test of coupling coagulation or adsorption ultrafiltration like under the test of a combination. Such a quality of water makes it possible to consider its reuse as industrial water.

References

[1] C.M.A. Ademoroti, The effect of pH on the coagulation and purification of wastewater, J. Effl. Wat. Treatment, Nov. (1980) 541–549.

[2] M.H. Al Malack and G.K. Anderson, Use of crossflow microfiltration in wastewater treatment, J. Wat. Res., 31(12) (1997) 3064–3072.

[3] A. Aguiar, E. Lefebvre, M. Rahni and B. Leguebe, Relationship between raw water TOC and the optimum coagulant dose (iron III chloride), Environ. Technol., 17 (1996) 381–389.

[4] M.F. Pouet, Traitements physico-chimiques associés à une microfiltration d'eau usée urbaine, Thèse de doctorat, USTL Montpellier, 1994.

[5] S. Vigneswaran and S. Boonthanon, Cross flow microfiltration with in line flocculation, Wat. Techn., Feb. (1992) 29–31.

[6] M. Mieuton-Peuchot and R. Ben Aim, Improvement of crossflow microfiltration performances with flocculation, in: Proc. 5th World Filtration Congress, Nice, June 1990, pp. 488–493.

[7] H.H. Ngo, S. Vigneswaran, S.H. Kim, A. Bidkar and H. Moon, Microfiltration–adsorption hybrid system in organic removal from water, Wat. Sci. Tech., 14(10–11) (2000) 51–57.

[8] C.F. Lin, Y. Huang and O.J. Hao, Ultrafiltration processes from removing humic substances: effect of molecular weight fractions and PAC treatment, Wat. Res., 33(5) (1999) 1252–1264.