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Performance of three ultrafiltration ceramic membranes in reducing polluting load of landfill leachate

Mohamed Zait[a], Soukayna Benalla[a], Basma Bachiri[a], Mustapha Tahaikt[a], Driss Dhiba[b], Azzedine Elmidaoui[a], Mohamed Taky[a,b,*]

[a]*Laboratory of Advanced Materials and Process Engineering, Ibn Tofail University, Faculty of Sciences, P.O. Box: 1246, Kenitra, Morocco, emails: mohamed.taky@uit.ac.ma (M. Taky), mohamed.zait@uit.ac.ma (M. Zait), soukayna.benalla@uit.ac.ma (S. Benalla), basma.bachirir@uit.ac.ma (B. Bachiri), mustapha.tahaikt@uit.ac.ma (M. Tahaikt), elmidaoui@uit.ac.ma (A. Elmidaui)*

[b]*International Water Research Institute, Mohammed VI Polytechnic University, Lot 660, Hay Moulay Rachid Ben Guerir, 43150 – Morocco, email: Driss.DHIBA@um6p.ma*

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ABSTRACT

The Oum Azza Landfill, which is located in the suburb of Rabat, receives daily 2,500–2,800 t of solid waste and produces 660 m^3/d of landfill leachate. The leachate treatment plant of Oum Azza includes a biological pretreatment (aeration tank and anoxic tank followed by a bag filter), and a membrane processes by reverse osmosis (RO). A low RO recovery rate and frequent stops caused by membranes fouling have been observed, this behavior is mainly due to the poor quality of the effluent after pretreatment. To address this problem and improve the physico-chemical quality of the effluent upstream of the RO processes, we consider in this paper the feasibility of ultrafiltration (UF) separation as a pretreatment. The purpose of this study is to evaluate and compare the performance of three ceramic UF membranes with different pore sizes (0.02, 0.05 and 0.1 μm). The transmembrane pressure (TMP) and circulation velocity effect on the quantity and the quality of permeate and on the removal efficiency of chemical oxygen demand (COD) and suspended solid (SS) are performed. The pretreatment of the leachate by UF shows that the SS and COD retention increase with TMP for all the tested membranes and reach, for a circulation velocity of 0.5 m/s, the maximum values of 85%, 77% and 72% respectively for COD and 70.4%, 62% and 55% for SS. On the other hand, the study shows that circulation velocity has no influence on the physico-chemical quality of the permeate in the range of the applied TMP. However, for an applied TMP, the flow rate increases with the circulation velocity. The results obtained show a clear improvement in effluent quality compared to the conventional pretreatment used in the leachate treatment plant of Oum Azza but pollutants indicators contents are still high and the effluent cannot be directed to the RO unit. Three methods are proposed to perfect the quality of the effluent at the entrance of the RO unit.

Keywords: Landfill leachate; Pollutant load; ultrafiltration; Ceramic membrane; Membrane fouling

1. Introduction

Fast urbanization, population growth and a strong change in production and consumption patterns generate significant volumes of waste, both in liquid and solid. With a population of 34 million and an urbanization rate of over 60%, Morocco has not escaped the inexorable growth in the quantity of waste produced which has reached 5.5 million tons of urban household waste and which will reach 9.3 million tons in 2030. At the national

* Corresponding author.

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level, the total production of waste reaches more than 7 million tons/y according to the National Household Waste Program (PNDM) [1], with a predominance of organic matter (70% causing 18% of the country's total greenhouse gas emissions). The industry generates more than 1.5 million tons annually, of which 256,000 t are hazardous waste. Finally, medical waste represents 6,000 t/y [2]. This waste has increased in volume, variety and toxicity, endangering public health, and the environment.

In 2008, morocco launched the National Household Waste Program whose objective is to meet the challenges of sustainable management of household and similar waste and to be part of the modernization process of this sector. It aims to ensure the collection and cleaning of household waste to reach a collection rate of 90% in 2022 and 100% in 2030 [1].

Morocco has moved from a policy of burial and closure and rehabilitation of wild landfills to the development of sorting, recycling and recovery sectors [1].

A landfill is a source of several sources of environmental nuisance such as: emission of odor, noise, dust, production of biogas and especially landfill leachate (LFL).

LFL is defined as the water that percolates through the waste, during this percolation, the liquid is loaded bacteriologically and chemically by mineral and organic substances, this is what is commonly called "landfill juice". The composition of buried waste, their degree of decomposition, their humidity rate and the age of the discharge are the main parameters influencing the composition of LFL [3–5]. The massive production of this "landfill juice" generates risks of pollution of soils, rivers and groundwater. It is therefore necessary to collect and treat it before discharge into the natural environment [6–8].

To overcome this problem, several processes drawn from wastewater treatment technology have been applied to treat LFL: aerobic and anaerobic biological degradation, chemical oxidation, chemical precipitation, coagulation-flocculation, activated carbon adsorption and membrane-based processes [9,10].

Coagulation–flocculation is one of the physical-chemical water treatments that has a long history and that is widely used for LFL treatment. Adsorption processes are also used for the treatment and it works in a way that leachate attaches to the surface of the absorption material and is removed from the liquid. Dissolved air flotation is another treatment technique but that is not much used in LFL treatment. Chemical oxidation consists of adding a strong oxidizing agent such as hydrogen peroxide or ozone that breaks the organic contaminants into carbon dioxide and water.

There are some biological treatment methods that aim at minimizing organic matter and nitrogen in the LFL. The biological treatment is simple and efficient, among these methods are: sequenced batch reactor, moving bed biofilm reactor, constructed wetlands, aerated lagoons.

Nowadays, membrane-based technologies have become popular for water treatment purposes [11,12]. They are increasingly used for the treatment of groundwater, surface water or wastewater. Thus, many researches currently deal with the integration of these membrane processes in the LFL treatment, some of these membrane processes are microfiltration (MF), ultrafiltration (UF), nanofiltration (NF) and reverse osmosis (RO). In 2017 Pertile et al. [13] investigated the use of MF in the treatment of LFL. The treatment allowed an abatement rate of 43% for chemical oxygen demand (COD) and 63% for biochemical demand of oxygen (BOD_5). A few years earlier, Pi et al. [14] performed a study on LFL treatment by UF using the 1 kDa membrane which reducing 86% of COD. Şeyda Özyaka et al. [15], by coupling two membranes (MP005 and ZW-UF), achieved a COD abatement rate of 33.54%. In the same way, Trebouet et al. [16] opted for different NF membranes for the treatment of LFL. With the MPT-31 NF-membrane, they reduced 80% and 98% of BOD_5 and COD respectively. Recently, Sabah et al. [17] treated LFL with RO, and obtained high abatement rates for COD (98%) and BOD_5 (97%). In 2001, Ahn et al. [18] treated LFL by coupling membrane bioreactor and RO, they reduced 97% COD and BOD_5, 99% of suspended solids (SS) and 99% of ammoniacal nitrogen ($N–NH_4^+$) and nitrate nitrogen ($N–NO_3^-$).

In a similar study carried out by Bohdziewicz et al. [19] and Hasar et al. [20], they found that COD was reduced by 99%. In other study based on a combination of activated sludge and RO, Li et al. [21] reported that the removal rate of COD, ammoniacal nitrogen and SS reached 99%, and the removal rate of nitrate nitrogen and nitrogen nitrite was less than 97% and 82.7% respectively.

Baumgarten and Sayfried [22] combined two techniques: biological contactor with RO, this combination led to a great result in the elimination of organic parameters, such as BOD_5 (>99% elimination), COD (99%) and total organic nitrogen (97%). The same efficiency was observed for inorganic substances: ammoniacal nitrogen, total inorganic nitrogen, lead (Pb) and chloride ion (Cl^-).

Pirbazari et al. [23] investigated the combination of UF and biologically activated carbon for LFL treatment, they obtained rejections in the range of 95% to 98% for total organic carbon and 97% for organic pollutants. However, these membrane processes are particularly limited by fouling due to the high pollutant load of the leachate.

The Oum Azza LFL treatment is performed in two stages: the first one is a biological pretreatment which consists of an aeration basin and an anoxic basin followed by a pocket filter, and the second stage is a membrane-based processes RO. The monitoring of the performances of the plant showed that the recovery rate of the RO is low, which leads to frequent stops for cleaning the fouled membranes due to the poor effluent quality after pretreatment. To address this problem, Ibn Tofail University and the Pizzorno Group have collaborated to study the feasibility and the efficiency of UF as pretreatment upstream RO processes.

The objective of this work is to study and compare the performances of three UF ceramic membranes of different pore sizes, as pretreatment of Oum Azza LFL. The choice of ceramic membranes over polymeric membranes is justified by several advantages: distinct pore size distribution, higher porosity, better separation, higher flux, chemical stability, higher hydrophilicity, long lifetime and reduced fouling [24]. The influence of the transmembrane pressure (TMP), the circulation velocity and the membranes cut-off on the physico-chemical quality of the permeate and on the abetment of the most pollution indicators have been studied.

2. Materials and methods

The LFL is periodically taken from the controlled dump of Oum Azza (110 ha) which is located in the commune of Oum Azza at 20 km from Rabat. This controlled Technical Landfill Center (CTC) was created in December 2007. The center receives significant tonnages ranging from 2,500 to 2,800 t/d of household waste from the three transfer centers: Rabat, Temara and Salé. The daily quantity of landfill leachate (LFL) produced in the landfill is 660 m^3/d.

Fig. 1 illustrates the stages of operation of the Oum Azza Landfill Plant.

Samples collected are transported to the laboratory to be analyzed and treated within hours following the collection.

Experiments are performed on a UF laboratory pilot supplied by the French company TIA (Technologies Industrielles Appliquées). It consists of a feeding tray with a capacity of 50 L and two pumps: one for circulation and the other for filtration (Fig. 2). The tangential speed of recirculation is in the range of 0.5–6 m/s. The TMP varies from 0 to 10 bar.

Table 1 gives the main characteristics of the membranes used. After UF tests, the membranes are cleaned with alkaline and acidic cleaning solutions according to the manufacturer's recommendations.

Permeate samples are collected and leachate parameters are determined analytically following standard methods:

COD (NF T90-101), BOD_5 (NF T90-103), and ammonium ion (NH_4^+) (NF T90-105) measurements are made according to AFNOR standards (1994). Determination of SS (NF T90-105-2) is carried out according to the AFNOR standards (1997).

Ion chromatography is used to determine the concentrations of chloride ion (Cl^-), nitrate ion (NO_3^-) and phosphate ion (PO_4^{3-}). Bicarbonate ions content is measured by the alkalimetry method. Potassium ion (K^+), sodium ion (Na^+), calcium ion (Ca^{2+}) and magnesium ion (Mg^{2+}) cations have been analyzed by atomic absorption spectroscopy.

Other parameter is the Retention R (%) which is defined according to Eq. (1):

$$R(\%) = \frac{C_0 - C_P}{C_0} \times 100 \tag{1}$$

where C_P and C_0 are permeate and initial concentrations respectively.

3. Results and discussion

3.1. Characterization of the raw LFL

The main LFL analysis are presented in Table 2. The sampling campaign was achieved during January 2019. As shown in Table 2, all the pollution indicator contents exceed the discharge limit values [25].

Table 1
Characteristics of the membranes

Characteristics	UF 1	UF 2	UF 3
Pore size (μm)	0.02	0.05	0.1
Nature	Ceramic	Ceramic	Ceramic
Surface (m^2)	0.35	0.35	0.24
Maximum pressure (bar)	10	10	10
Maximum temperature (°C)	100	100	100
pH range	3–11	3–11	3–11

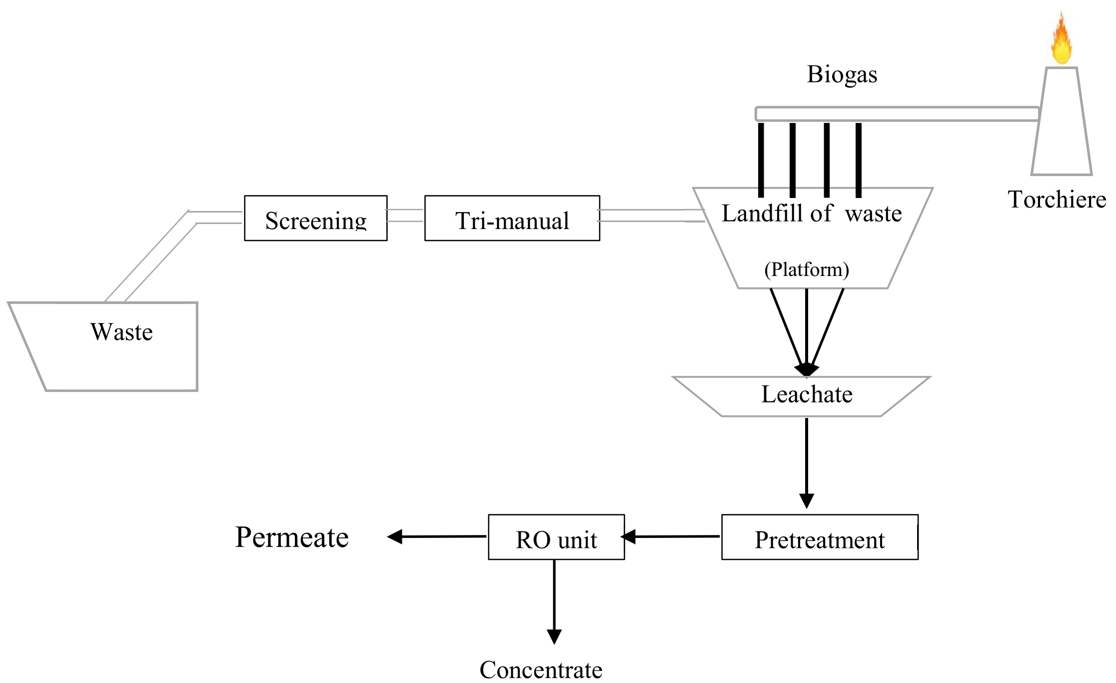

Fig. 1. Operating stages of the Oum Azza Landfill Plant.

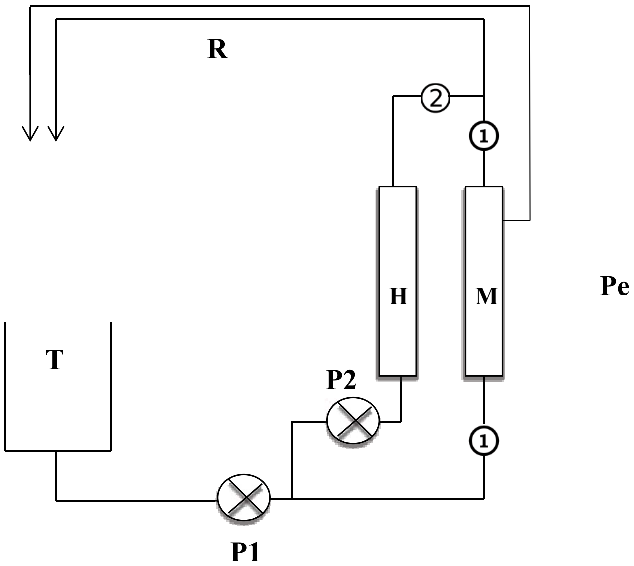

Fig. 2. Diagram of the ultrafiltration pilot plant. *T*: Tank; P_1: Feed pump; P_2: Filtration pump; *M*: Ultrafiltration module; P_e: Permeate recirculation; *R*: Retentate recirculation; *H*: Heat exchanger; 1: Pressure sensor; 2: Temperature sensor.

Table 2
Physico-chemical and organic characterization of leachate and Moroccan discharge limit values

Settings	Value	Domestic discharge limit values [25]
pH	8.5	5.5–9.5
Temperature (°C)	18.6	30
Electric conductivity (mS/cm)	30	2,700
COD (mg d'O_2/L)	8,000	500
BOD_5 (mg d'O_2/L)	4,500	100
SS (mg/L)	4,120	100
NH_4^+ (mg/L)	150	–
HCO_3^- (mg/L)	12,810	–
Cl^- (mg/L)	3,650	–
NO_3^- (mg/L)	4.1	–
PO_4^{3-} (mg/L)	4.9	–
Ca^{2+} (mg/L)	7.6	–
Mg^{2+} (mg/L)	230	–
K^+ (mg/L)	6,000	–
Na^+ (mg/L)	4,800	–

These analyses show a strong organic pollution which results in high load of BOD_5, COD, SS and a high electric conductivity. The average temperature of 18.6°C is fairly characteristic of the month during which the samples were taken and this recorded value is much lower than the value recommended for the (general limit value of discharge). Oum Azza LFL potential hydrogen (pH) is 8.5, value which is in the range of Moroccan release limits. This slight basic character has been emphasized in several works on Moroccan landfills [26–29]. One of the most features of this LFL is its high electric conductivity which is around 30 mS/cm. This value exceeds the standards of treated wastewater. Similar values were mentioned by different researchers working on LFL of Zalaghi et al. [27] and Khalil et al. [28] in Morocco.

Due to this high electric conductivity decision-makers and stakeholders have opted for the treatment of Oum Azza LFL by RO. Indeed, it is the only technology capable of providing a very significant reduction in total dissolved solids (TDS); this is why RO is currently the main seawater desalination process worldwide. However, the efficiency of RO in the treatment of LFL is largely dependent on the performance of the pretreatment. If the pretreatment is not very refined, the RO membranes will be quickly fouled, and flows will drop, which leads to the implementation of osmosis membrane washing sequence with high frequency.

For COD, the value obtained, 8,000 mg O_2/L, exceeds largely the limit value of discharges (500 mg O_2/L). This high value indicates a very high organic load and shows that these liquid effluents are in the reducing conditions (oxygen drop). Similar values of COD are reported in different studies on Moroccan LFL. [27,29,30]. For BOD_5, the content measured is 4500 mg d'O_2/L. This value is very high compared to the limit value of discharges (100 mg d'O_2/L).

And finally, analysis of SS reveals a content of 4,000 mg/L. This concentration is slightly higher than the limit value fixed by Moroccan Liquid Discharges Standards (MLDS, 2018).

3.2. Pretreatment of LFL by UF

3.2.1. Membrane permeability

Fig. 3 gives the variation of pure water flux as a function of TMP for the three membranes. The circulation velocity and temperature are respectively fixed at 0.5 m/s and 25°C.

As shown in Fig. 3, the permeate flow varies linearly with the TMP in accordance with Darcy's law for the three membranes. On the other hand, this flux increases linearly with the pore size of the membranes within the range of pressure studied. The obtained permeabilities of the three membranes are presented in Table 3.

3.2.2. Influence of pore size

Fig. 4 illustrates the evolution of flux and electric conductivity of the permeate and retention of COD and SS for as a function of pore size of the membranes. The circulation velocity V = 0.5 m/s is considered as the minimum value that can be issued by the laboratory pilot. The TMP is 5 bar, which is the medium pressure, and a temperature is maintained at 26°C. Results show that the permeate flux increases with the membrane pore size. The highest flux is obtained with the membrane that has the greatest pore size (100 L/h.m^2, 0.1 μm), the lowest flux is obtained with the membrane that has the lowest pore size (46.79 L/h.m^2, 0.02 μm) and for the third membrane, with intermediate pore size (0.05 μm), the obtained flux is 27 L/h.m^2. These fluxes are lower than those obtained with pure water (Fig. 1) as result of concentration polarization by the retained compounds and the fouling of membranes. In general, membranes, which have small pore sizes fouling up quickly due to the particles having molecular weights between 500 and 1,000 Da, which are widely present in LFL [31]. On the other hand, pore size of UF membrane has no influence on the electric conductivity of the ultrafiltrate, only a TDS retention of 6% is exhibited

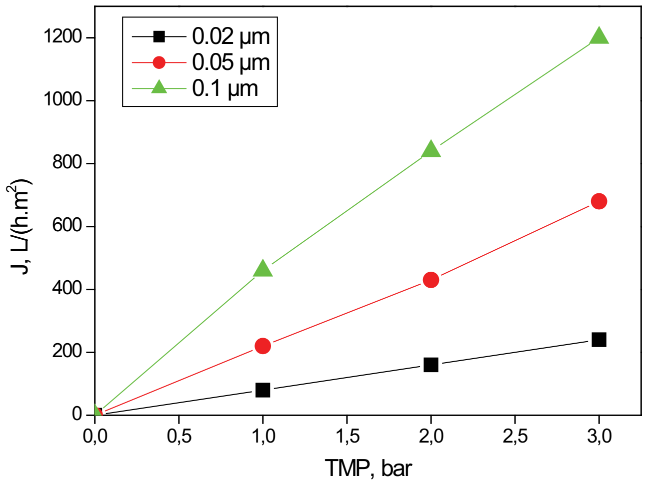

Fig. 3. The evolution of the water permeates flux as a function of TMP at 25°C.

Table 3
Permeabilities and R-squared of the three ultrafiltration membranes

	Permeability (L/h m²)	R-squared
UF 0.02 µm	80	1
UF 0.05 µm	225	0.99
UF 0.1 µm	398	0.99

by the membrane with the lowest pore size (0.02 µm). Indeed, TDS is distributed equally between permeate and retentate of UF, which means that the electric conductivity will be practically the same in permeate and retentate.

Retention of COD and SS decreases with increasing pore size of membranes. The highest retentions of COD (85%) and SS (70.4%) are obtained with membrane having the lowest pore size (0.02 µm), intermediate retention values of COD (74.14%) and SS (58.96%) are exhibited by the membrane with intermediate pore size (0.05 µm) and the lowest retentions of COD (69%) and SS (49.53%) are obtained with the membrane of the highest pore size (0.1 µm). Particle size influences the removal efficiency as ceramic membranes with smaller molecular weight cut-off (MWCO) show better retentions.

3.2.3. Influence of TMP and velocity of circulation

3.2.3.1. Influence of TMP

For the three membranes and for a velocity of circulation V = 0.5 m/s, Fig. 5 gives the variations of permeate flux, retention of COD and SS, electric conductivity and pH as a function of TMP. The effect on flux is studied in order to find the optimum TMP and control the membrane fouling.

Following to the results obtained in Fig. 5a, the permeate flux of the three membranes increases with TMP and tends towards a plateau which indicates the limiting flux. This limiting flux value becomes independent of the pressure when the clogging layer is formed.

For the two membranes of UF 0.05 and 0.1 µm, the limiting flux is observed at an applied TMP of 5 bar. On the other hand, with the 0.02 µm UF membrane, the limiting flux which is the lower one (29 L/h m²) is observed at a TMP of 2 bars. For the other membranes 0.05 and 0.1 µm, the limiting fluxes reach respectively 46.79 and 100 L/h m².

As the pressure increases beyond a certain threshold value, the flux becomes constant and independent of pressure.

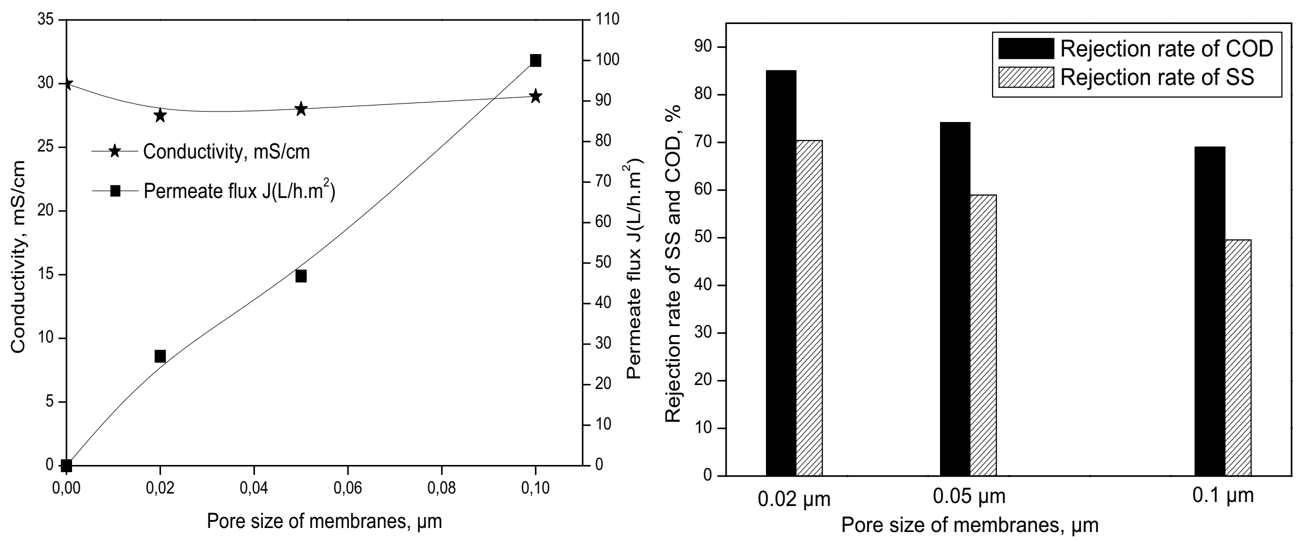

Fig. 4. Variation of flux, conductivity and retention of SS and COD as a function of pore size.

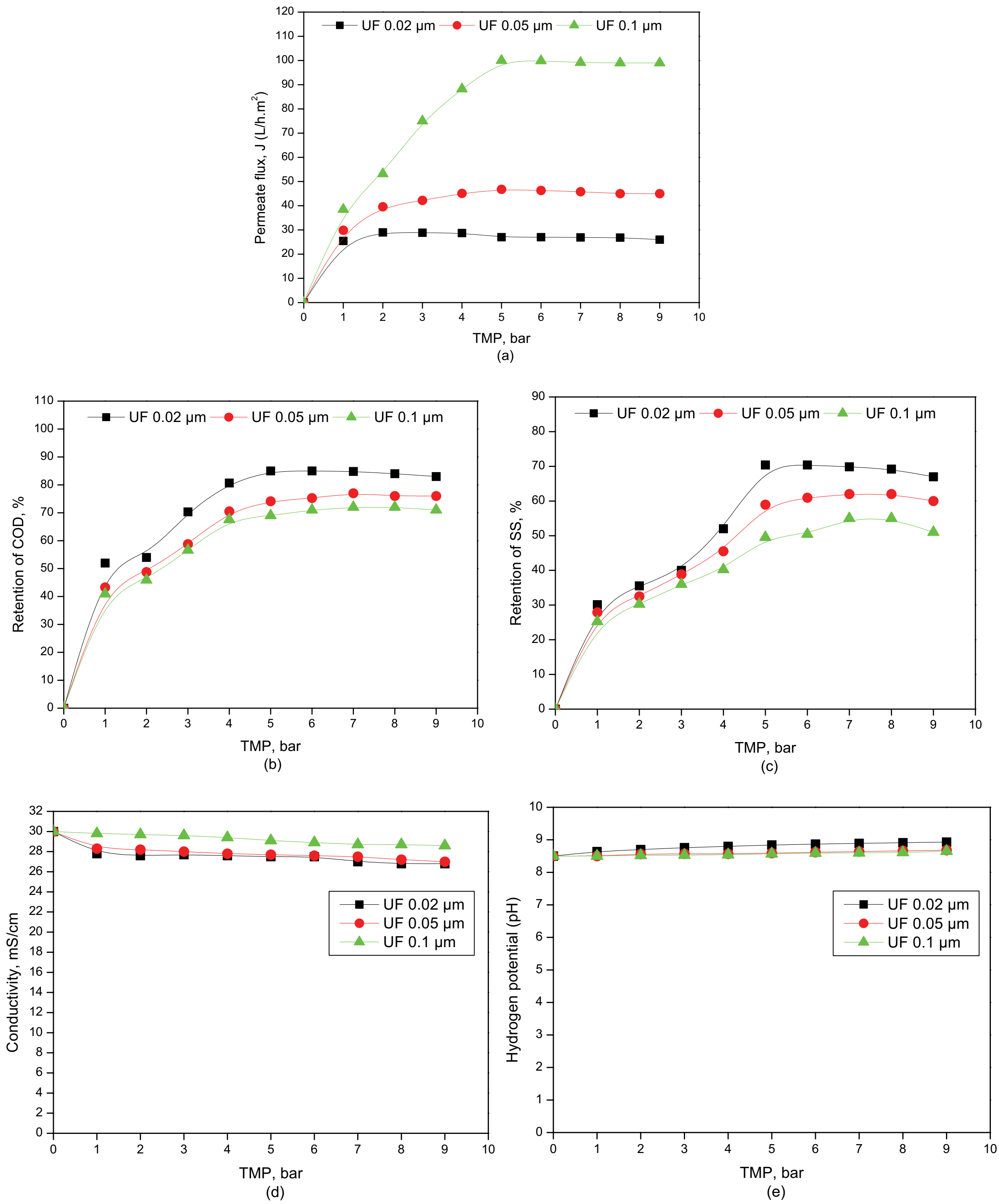

Fig. 5. Variations of permeate flux (a), COD (b) and SS (c) retention, electric conductivity (d) and pH (e) as a function of TMP (V = 0.5 m/s).

The optimum TMP is preferably 80% of the limiting pressure: P_{opt} = 1.2 bar for the UF membrane (0.02 μm), P_{opt} = 1.6 bar for (0.05 μm) and P_{opt} = 2.5 bar for (0.1 μm).

Concerning the influence of TMP on retention of COD and SS (Fig. 5b and c), it appears that initially retention increases with TMP and then tends towards a plateau that means the limit retention.

As the TMP increases, more pollutants accumulate on the membrane surface, forming a gel layer and clogging the pores, which increase filtration resistance due to higher compression of pollutants. Depending on the polarization model of the gel, the existence of limiting flux is related to concentration polarization that occurs when the feed solution containing suspended and soluble solids (colloids) passes through the membrane. The formation of a viscous, gelatinous layer is responsible for additional resistance to the permeate flux in addition to that of the membrane [32] which explain the increase in retention. The highest retentions of COD and SS are obtained with the membrane having the smallest pore size, since with these membranes the concentration polarization conditions are quickly reached. This result is in accordance with the convective transport mechanism that governs transfer through UF membranes.

As shown in Fig. 5d, TMP has no impact on electric conductivity of the permeate since the UF membranes used are porous and allow the mineral salts to pass through the membranes. Electric conductivity is almost the same and have the same in the feed, permeate and retentate.

Fig. 5e shows that the pH of permeate remains almost stable with the increase of TMP [33,34].

3.2.3.2. Influence of circulation velocity on the permeate flux

In general, the increase in tangential velocity decreases the thickness of the concentration polarization layers on the surface of membrane. It also makes it possible to reduce the deposition of large particles in suspension which are more easily trained. Finally, it generates shear forces in the vicinity of the membrane, leading to an increase in ultrafiltrate flow. This is why the study of the influence of circulation velocity is crucial for the three membranes, in order to determine the best operating conditions.

As shown previously, membrane with the higher pore size exhibits the higher flux. For the three studied membranes. The evolution of the permeation flux as a function of TMP for different circulation velocity is illustrated in Fig. 6. At equal TMP the increase of circulation velocity improves the permeation flux across membranes.

At equal circulation velocity, the increase of TMP enhances the permeation flux until it reaches a limiting flux, a plateau. This limiting flux disappears as soon as the circulation velocity reaches 6 m/s.

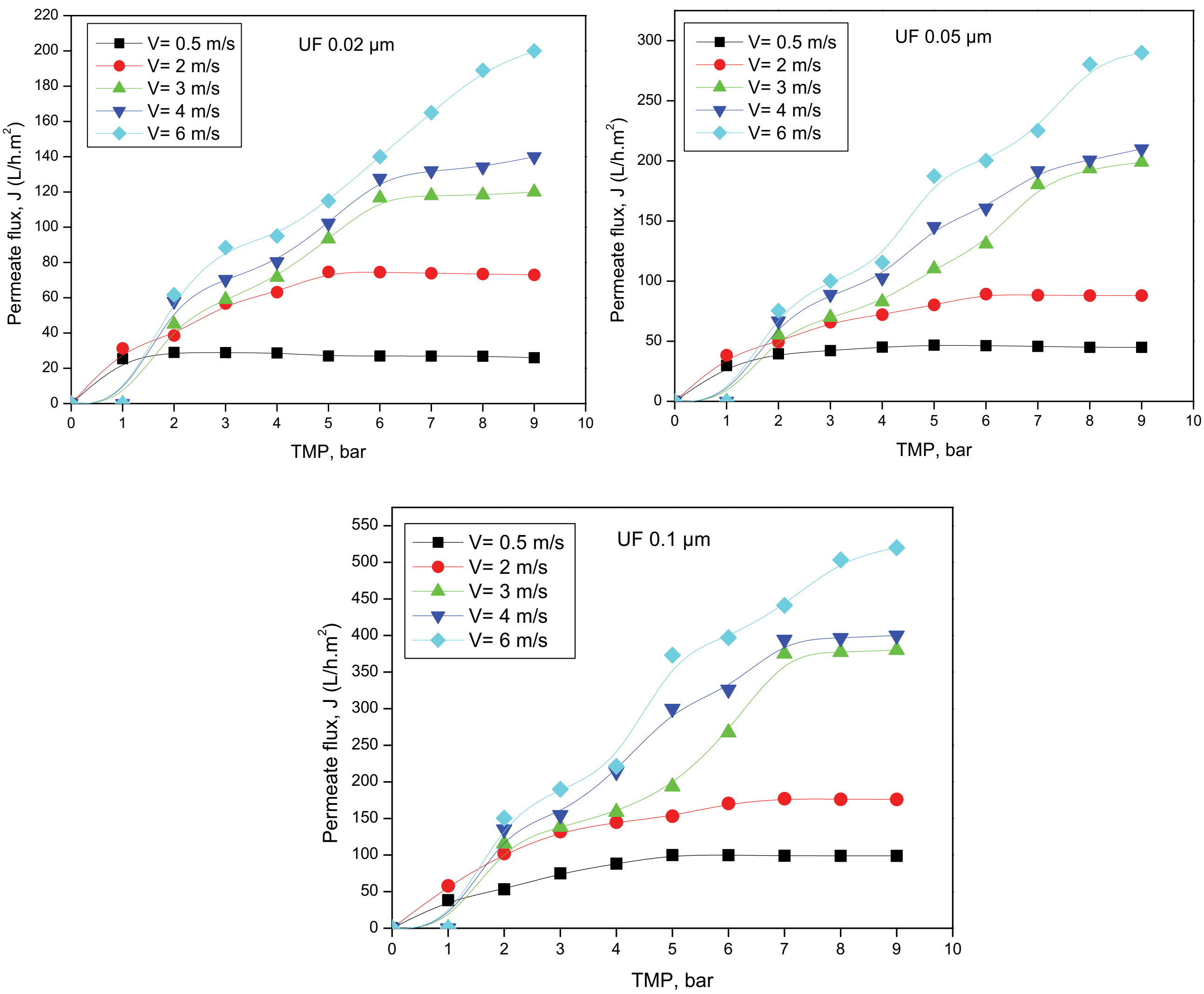

Fig. 6. Evolution of the permeation flux as a function of TMP at different circulation velocities for the three membranes studied (26°C).

An increase in circulating velocity leads to an increase in permeate limiting flux across UF membranes. Depending on the film model, an increase in recirculation velocity improves the hydrodynamic conditions by reducing the concentration polarization layer thus avoiding the formation of the fouling layer and enhancing the mass transfer coefficient resulting in an increase of the permeation flux [35]. At the higher circulation velocity (6 m/s), the permeate limiting flux is not reached meaning that in these conditions, ongoing hydrodynamic conditions reduce concentration

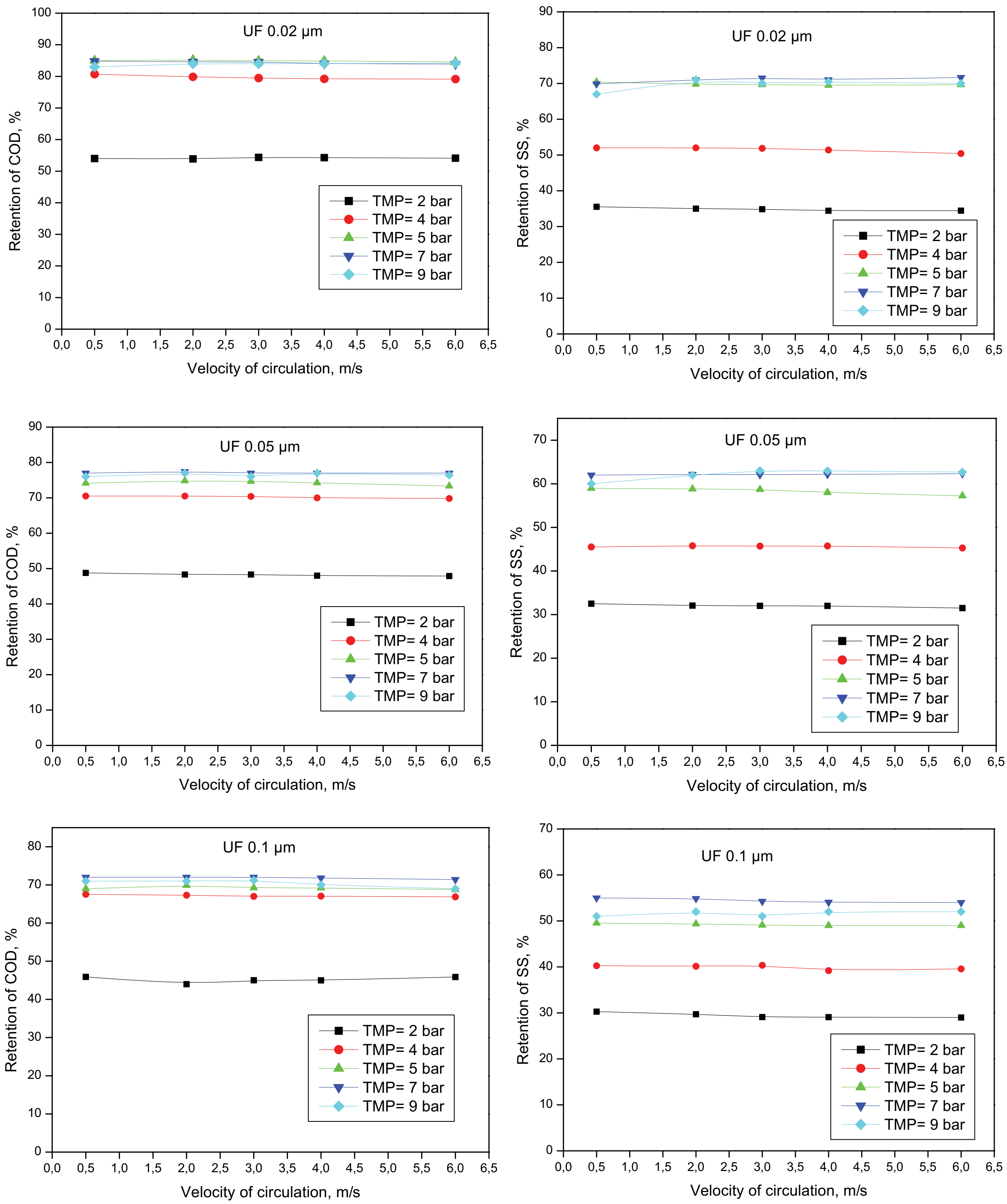

Fig. 7. Evolution of the COD and SS retentions as a function of circulation velocity for different TMP.

polarization and allow continuous leachate renewal at the solution-membrane interface for the three studied membranes. This of course has an energy cost since circulation requires energy. For this reason, a compromise must be found between the circulation velocity and the permeation flow across the membranes.

3.2.3.3. Influence of circulation velocity on permeate quality

Fig. 7 gives the evolution of the COD and SS retention as a function of circulation velocity for different TMP for the three membranes. Results show that for each applied TMP, retention of COD and SS exhibited by membranes remain stable independently of the change in circulation velocity. Circulation velocity has no influence on the retention of COD and SS and consequently on permeate quality. Thus, for the UF membrane with porosity of 0.02 μm and for a TMP of 5 bar, the change in circulation velocity from 0.5 to 6 m/s does not affect retention values of COD and SS which remain invariant, 85% and 70.4% respectively. The same phenomenon is observed with the other two membranes.

According to these results, as confirmed previously, circulation velocity greatly affects the permeation flux, disrupting the establishment of concentration polarization. But it does not affect COD and SS retentions for the three membranes which are related to the inherent structural properties of UF membrane (pore size) and the size of the particles retained.

Regarding the obtained quality of permeate, average retention of 70% of the COD and 60% of the SS have been achieved.

So, the choice of the working circulation velocity must be a subject of depth reflection in the process of optimizing LFL pretreatment by UF. A low velocity seems to be penalizing in terms of filtration quantity performance and a strong propensity to concentration polarization and fouling. Inversely, a high circulation velocity (between 4 and 6 m/s) will provide better permeation flow rate with stable retention as mentioned previously. But this flow improvement will be thanks to additive energetic consumption. The choice of operating TMP and circulation velocity must be the subject of a compromise only capable of sustaining working conditions and guaranteeing stability in terms of filtration performance.

The objective of this work is to study and compare performances of three UF ceramic membranes in the pretreatment of the fresh Oum Azza LFL, raw effluent, which has not undergone any pretreatment, in order to improve the quality of the effluent at entrance of RO plant and reduce the membrane fouling and washing frequencies. The pretreatment consists to reduce the pollutant load of LFL, in order to improve the physico-chemical quality of the effluent upstream of RO. The results obtained with a single stage show a clear improvement in the quality of the effluent compared to the existing pre-treatment provided by the operators of Oum Azza. This improvement will which will reduce the RO membrane fouling and washing frequencies. However, the abatements of the major pollutants are far from guaranteeing protection of membranes according to RO manufacturer recommendations. Three options are proposed to further improve the effluent quality at the outlet of UF step: (i) UF treatment with two or three stages, (ii) introduce a conventional pretreatment step before UF with one stage, (iii) replace UF by membrane bioreactor by treating the effluent at the outlet of the biological reactor. The investigation of this options is ongoing and the results will be published soon.

4. Conclusion

Treating the LFL by UF reduces a large part of organic pollution, however the TDS of these leachates still very high (close to 35 mS/cm). So, to meet the Moroccan discharge standards in terms of TDS, RO seems to be the unique process capable to responding to this constraint. This technology is known to be cost expensive and RO membranes are sensitive to fouling.

The aim of this work was to study the feasibility of UF membranes in the reduction of pollution indicators of Oum Azza LFL with the ultimate goal of replacing the current pretreatment in work at Oum Azza by UF separation.

This study shows that, increasing the applied TMP for the three membranes causes a rise in permeate flux until a limiting flux and improve the retention of COD and SS, but it has no influence on TDS.

Moreover, the velocity of circulation has no influence on the retention rate of COD and SS. On the other hand, the permeate fluxes and the limiting fluxes have been improved with the increase in the velocity of circulation.

A clear improvement in effluent quality compared to the conventional pretreatment used in the leachate treatment plant of Oum Azza but pollutants indicators contents still high and the effluent cannot be directed to the RO unit.

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