Studii şi Cercetări Ştiinţifice
Chimie şi Inginerie Chimică, Biotehnologii, Industrie Alimentară

2019, **20** (1), pp. 075 – 087

Scientific Study & Research
Chemistry & Chemical Engineering, Biotechnology, Food Industry

ISSN 1582-540X

ORIGINAL RESEARCH PAPER

EXPLOITATION OF ORGANIC ULTRAFILTRATION MEMBRANES IN THE REMOVAL OF PEPTONE FROM WASTEWATER

Laurențiu Tataru[1], Valentin Nedeff[1,2*], Emilian Moșneguțu[1], Mirela Panainte-Lehăduș[1], Dana Alexandra Chițimuș[1]

[1]"Vasile Alecsandri" University of Bacau, Faculty of Engineering, Department of Environmental Engineering and Mechanical Engineering, 600115, Bacau, Romania
[2]Academy of Agricultural and Forestry Sciences "Gh. Ionescu, Sisesti", Bucharest, Romania

*Corresponding author: v.nedeff@ub.ro

Received: November, 23, 2018
Accepted: March, 07, 2019

Abstract: In the present paper an ultrafiltration experiment was carried out for testing the polyacrylonitrile (PAN) membrane in the process of peptone separation from wastewater. The evaluation of the applied ultrafiltration process consisted in the monitoring and the recording of the parameters as well as of some indicators that will reveal the PAN membrane efficiency (the percentage of peptone retention on the membrane surface, the volume and quality of the resulting permeate in the pre-established time period).
The obtained results showed that the peptone retention efficiency was noticeably, around 20 % and the obtained permeate volume did not decrease greatly, only by 2.5 L over 300 minutes experiment.

Keywords: *hollow fiber membrane, industrial wastewater, peptone, polyacrylonitrile, ultrafiltration*

INTRODUCTION

The increased amount of wastewaters represents an important problem for environment, a quick proper management being required in order to avoid, as much as possible, the destructive environmental impact of different compounds present in industrial wastewater.
The treatment technology of wastewater has evolved along with the demographic expansion and, in accordance with legislation, it is mandatory. In this context, the removing of toxic compounds in water should be as effective as possible [1, 2].
Ultrafiltration membranes are continually expanding in terms of exploitation, as they offer superior qualities of the final product (respectively permeate from wastewater filtration), at acceptable costs, without harming the environment [3 – 7].
However, the ultrafiltration process cannot be the perfect solution for removing all bacteria, viruses or organic matter from wastewater given the fact that their dimensions are inferior to the membrane pores dimensions; this makes it easy to pass through the walls of the membrane, finally leading to the decrease of permeate quality [8 – 12].
Different studies [13, 14] have shown that the use of magnetic ion exchange resins represent a more effective method for the retention of wastewater fine particles compared to clotting and flocculation.
The efficacy of wastewater ultrafiltration for the elimination of some pharmaceutical compounds has also been demonstrated [15 – 19].
Peptone is a powder with relatively small particle sizes, very soluble in water, commonly used in the pharmaceutical industry (different culture media for bacteria, the production of various drugs, antibiotics), but also in the food industry [20 – 23].
The purpose of this study is to monitor the flow of permeate in the continuous ultrafiltration process over 300 minutes. The present paper focuses on the relationship between the parameters that most influence the ultrafiltration process, namely the pressure at the membrane inlet, the pressure from the concentrate and the permeate, the temperature and the flow of the liquid subjected to the ultrafiltration. The efficiency of peptone retention by the PAN organic membrane was determined through a relationship that expresses the peptone concentration retained on the membrane during the set time.

MATERIALS AND METHODS

Pilot ultrafiltration plant MP 90

The plant used to conduct the ultrafiltration experiment is shown in Figure 1 and the sketch for plant setup is shown in Figure 2.
The PAN organic membrane module belongs to the MP 90 ultrafiltration pilot plant of the "Vasile Alecsandri" University of Bacau research laboratory, the module being manufactured in Japan, by Asahi Kasei Chemical Corporation for Pall Corporation (Figure 1) [24]. The module has a number of organic fibers, hollow inside, arranged symmetrically, fibers having a pore size of 0.3 μm. The outside diameter of the fibers is 1.4 μm and the inside diameter is 0.8 μm, and can easily hold particles that exceed the size of the membrane pores [24]. The molecular weight of the membrane is 13 kDa according to the installation guide [24].

Figure 1. *Ultrafiltration plant MP 90*
(1 - PAN membrane module; 2 - Control panel, recording system parameters; 3 - Liquid suction pump from the supply tank; 4 - Homogenization feed tank; 5 - Concentrate tank; 6 - Permeate collection tank; 7 - Large particle retaining filters; 8, 9 - Flow meters for feed water and permeate; 10 - Valve; 11 - Pressure sensors)

Materials used in the experiment

Peptone was purchased from the chemical company "Sigma-Aldrich Chemie Company", as well as the acids needed to clean the membrane at the end of the experiment.

Preparation of the peptone solution

The peptone, having particle sizes substantially around 5 µm, was mixed with tap water with an electrical conductivity of about 300 $\mu S \cdot cm^{-1}$.
Peptone preparation consisted of dissolving 90 g of peptone in one liter of water at 300 rpm mixing speed, for 35 minutes, then the solution was diluted with tap water up to 30 L, in order to reach a peptone concentration of 3 $g \cdot L^{-1}$.
After dissolving the peptone, a yellowish solution similar to the citrus juices was obtained. The concentrated solution was introduced into the feed tank, diluted with water up to 30 L and recirculated for several minutes for homogenization, without the liquid penetrating into the membrane.
Throughout the experiment, distilled water was used to clean the membrane module, the pipes circuit, and the tanks. At the end of the experiment, membrane module was

cleaned by recirculation of tap water in countercurrent for remove all particle from pores.

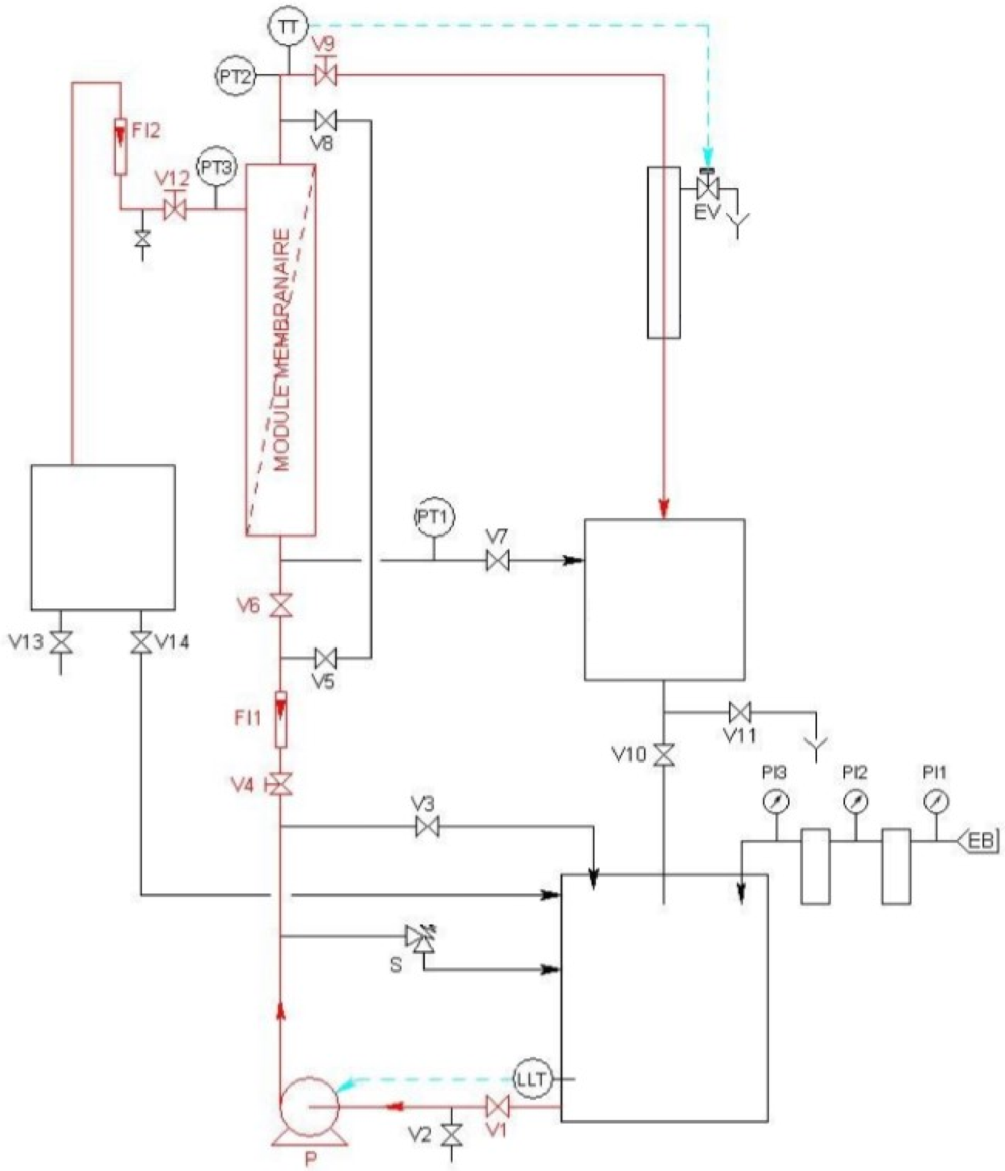

Figure 2. *Experimental sketch of MP 90 setup*
(V – valves; S – Overpressure valve; P - pump; EV – Thermocouple;
EB – alimentation with tap water; PI – pressure indicators; FI – flow indicators;
PT – Pressure sensor; TT – Temperature sensor; LLT – Level sensors)

The monitoring of ultrafiltration process

During 5 hours of experiment the following parameters have been monitored, respectively: pressure, temperature, flow and chemical indicators of the samples.
It was considered that the time chosen, respectively 300 minutes, was enough to follow the time evolution of the permeate volume obtained; the data collection was made every

30 minutes to better observe the fluctuations of the essential parameters leading to good conduct of the ultrafiltration process.

During this time it was possible to decide to what extent the ultrafiltration membrane was able to retain the peptone, in other words it was considered that the chosen time was enough to draw conclusions about membrane efficiency.

Since the beginning of the experiment, the emphasis has been placed on certain parameters [24] such as permeate flow and transmembrane pressure because they depend most on the efficiency of the membrane used in the study. An equally important indicator was the time elapsed between data collection. Random sampling, or delaying or hurrying the collection of each sample, would have implied the recording of wrong data that could have led to misinterpretation of process evolution.

At the beginning and at the end of the experiment, a sample of the solution was taken from the supply tank and then, over time, samples of the permeate obtained as a result of the good functioning of the membrane were collected, as regards the peptone retention on the active surface.

The volume of data obtained has shown that this type of membrane can function in good conditions and in the containment of peptone, and this type of substance that is present in the wastewater resulting from different activities can be retained if its particles have larger dimensions than those of the pores and the membrane in the study was not damaged.

The density of liquid flowing through the plant in the unit of time was determined using equation 1 [24]:

$$J = \frac{Q}{S} \tag{1}$$

where: J - density of liquid entering the membrane module along the experiment, expressed in $L \cdot h^{-1} \cdot m^{-2}$; S - membrane filtration surface, expressed in m^2; Q - flow of the liquid in the installation, expressed in $L \cdot h^{-1}$.

The transmembrane pressure, according to the MP 90 ultrafiltration pilot manual, was calculated using equation 2 [24]:

$$PTM = \frac{P_1 + P_2}{2} - P_3 \tag{2}$$

where: PTM - transmembrane pressure, expressed in bar; P_1 - fluid pressure at the entry into the membrane module, expressed in bar; P_2 - pressure of the concentrate, expressed in bar; P_3 - pressure of the permeate or pressure of the liquid at the outlet of the membrane, expressed in bar.

The retention rate was calculated using equation 3 [24]:

$$T_{ri} = \left(1 - \frac{C_{Pi}}{C_{Ri}}\right) \times 100 \tag{3}$$

here: T_{ri} - represents the calculated retention rate and expressed in %; C_{Pi} - concentration of the substance in the concentrate samples, expressed in %; C_{Ri} - concentration of the substance in the permeate samples, expressed as %.

As the retention rate is closer to the maximum value of 100 %, the more effective the membrane is in retaining the substances proposed for the study.

Some of the main parameters of the experiment in the present study, namely temperature, pressure, volume concentration factor, flow density retention rate, were recorded automatically, others have been calculated and analyzed later.
A WTW kit (produced by MultiLab SRL) was used for measurement of *p*H (WTW 3210 *p*H-meter), conductivity (COND 3210 Conductometer), dissolved oxygen (OXY 3210), and turbidity (TURB 430 IR), in the collected samples.

RESULTS AND DISCUSSION

Evolution of permeate volume over time relative to membrane inlet flow

Figure 3 shows the variation of the permeate flow obtained over 300 minutes. As seen in Figure 3, the permeate flow is decreasing and this is obvious because over time, the membrane tends to dirt, the blocking of the pores being assigned to the accumulation of peptone particles deposited on the pore walls or on the surface of the membrane, if the peptone particle size exceeds the peptone size.

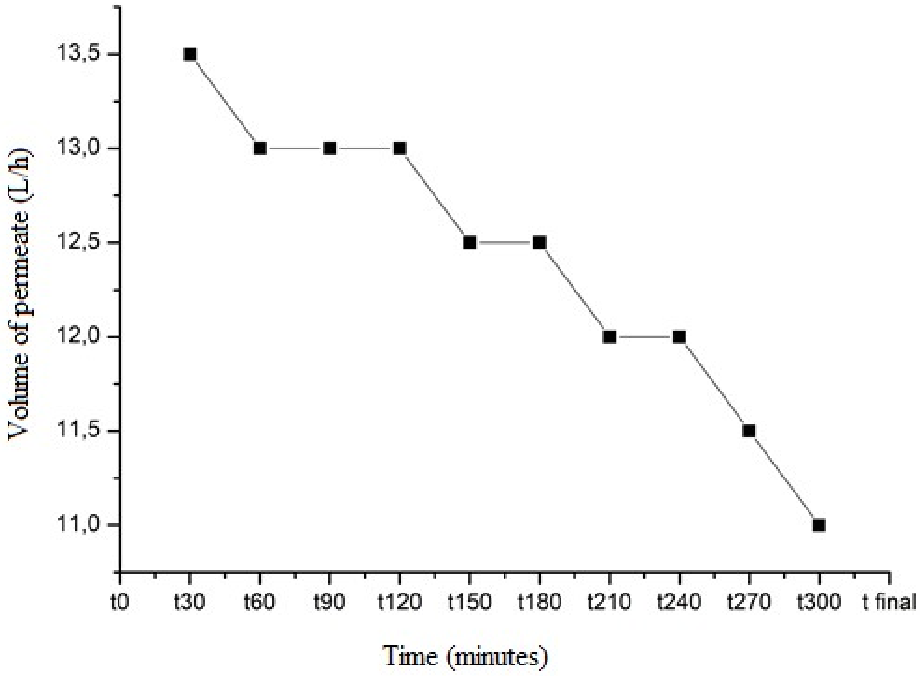

Figure 3. *The variation in time of the permeate volume at 1500 L·h^{-1} inlet flow*

The volume of permeate obtained decreased over time by about 2.5 liters. This is due to the fouling of the membrane over time. The permeate dropped by approximately 0.25 L every 30 minutes, summing up to the end of the experiment a 2.5 liters decline over the 5 hours of continuous filtration of the ultrafiltration membrane.
For example, from the 60th minute to the 120th minute, the permeate flow appears to be the same, with insignificant fluctuations. This can be explained by the fact that during this time the membrane pores are not blocked, and the membrane is not damaged as a result of wear, and this implies maintaining the permeate quality.

Variation of permeate conductivity in collected samples

With the decrease in permeate volume, the conductivity of the collected samples decreased (Figure 4). At the beginning, the decrease in conductivity is due to the fact that the permeate decreases the peptone concentration, respectively, the membrane retains the particles on its surface. But after 210th minute, the conductivity of the collected samples slightly increases, indicating that at the membrane's surface particles tend to agglomerate, fact that can block some pores of membrane and broken other pores.

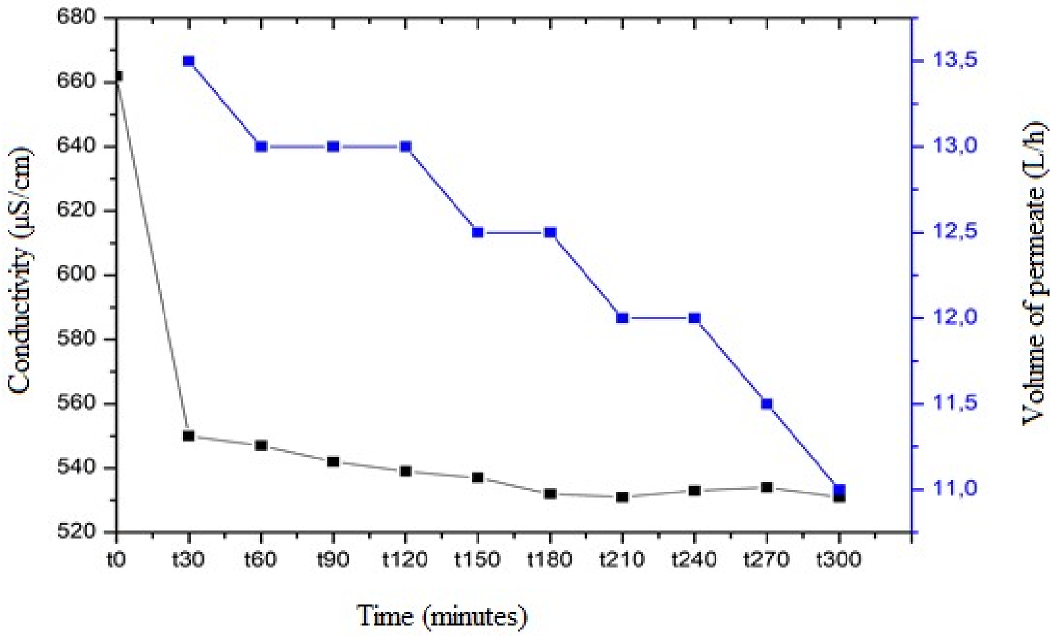

Figure 4. *Variation of conductivity in permeate samples*

In other words, it is normal that with the passing of time, increase the concentration of the substance in the sample collected and the electrical conductivity to begin to increase. This will always make the performance of the membrane decline.

The role of the temperature in the ultrafiltration process of the peptone solution

The temperature has been increasing throughout the ultrafiltration process and this is due to the fact that when the membrane holds the peptone, there is some pressure in the system which causes the fluid to continuously heat up (Figure 5).
As shown in Figure 5, the temperature variation was inversely proportional to the permeate, increasing continuously, regardless of the flow of liquid entering the plant during the ultrafiltration process.
The flow rate of the peptone-based liquid was equal to 1500 $L \cdot h^{-1}$ (Figure 5). This flow did not show any decline because the peptone is not a substance with large particle size that can quickly block the membrane pores and thus decrease the ultrafiltration performance of the membrane. Otherwise, the drop in input flow would have been felt as early as the first minutes of the process.

The temperature increased significantly, so that the experiment began at room temperature and laboratory conditions, respectively 21 °C, and its fluctuations finally reached the value of 27.1 °C, as shown in Figure 5. This value is quite high having in view of the fact that the experiment took place for only 300 minutes.
Probably, if the experiment would have been done for 10 hours, the maximum temperature reached could easily exceed the double value recorded in this experiment, assuming that the temperature linearly increases with time. Temperature recording was done automatically, being read only from the control panel shown in Figure 1 and recorded as stated in the research procedure of this experiment.

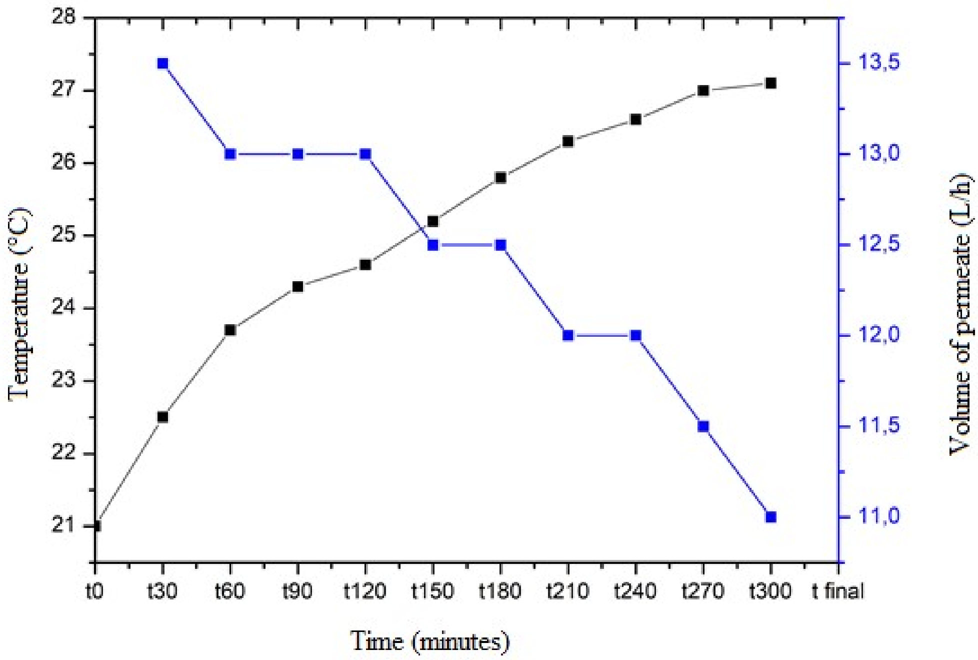

Figure 5. *Temperature increase over time at 1500 L·h^{-1} inlet flow*

Influence of pressures in the system

As can be seen in Figure 6, the pressure has had very little influence over the whole process, varying insignificantly.
The pressure fluctuations were only slightly visible on the concentrate side, ranging from 0.7 to 0.8 bar (Figure 6). At the inlet, the pressure was kept constant because fine particles of peptone did not put the liquid through the installation in difficulty and this did not create any pressure in the process because the blocking of the pores did not take place in an accentuated manner.
This time, the temperature variation was not directly proportional to the pressure, and this involves conducting the ultrafiltration process under easy conditions without disturbing the other important system parameters.

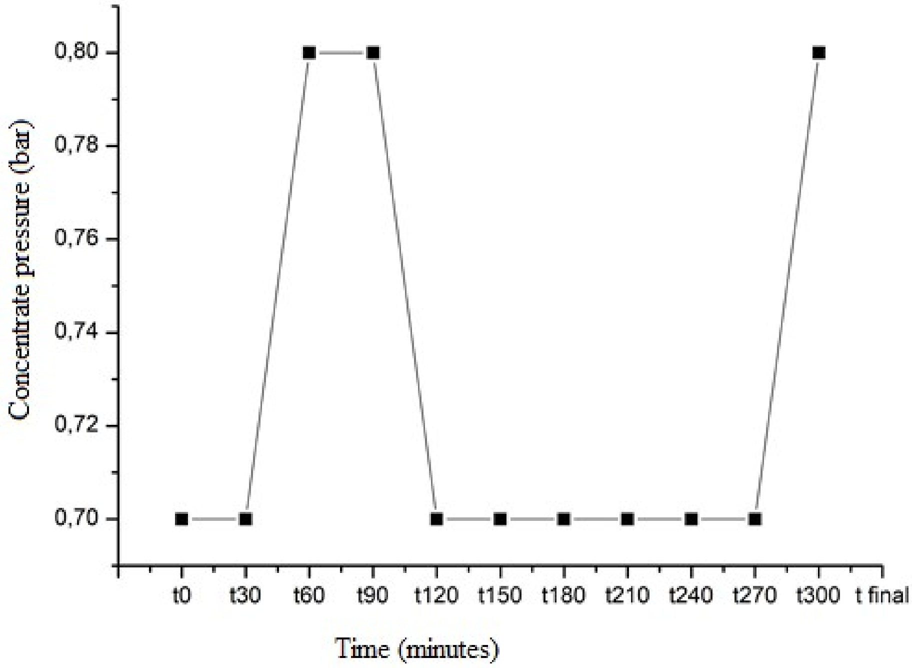

***Figure 6**. Pressure of concentrate related to 1 bar inlet pressure*

Transmembrane pressure varied to a small extent, from 0.85 bar to 0.9 bar, due to fluctuations that occurred in minutes 60 - 90 - 300.

Variation of chemical indicators in permeate samples

As can be seen in Figure 7, *p*H has grown steadily from 7.7 to 8.4.

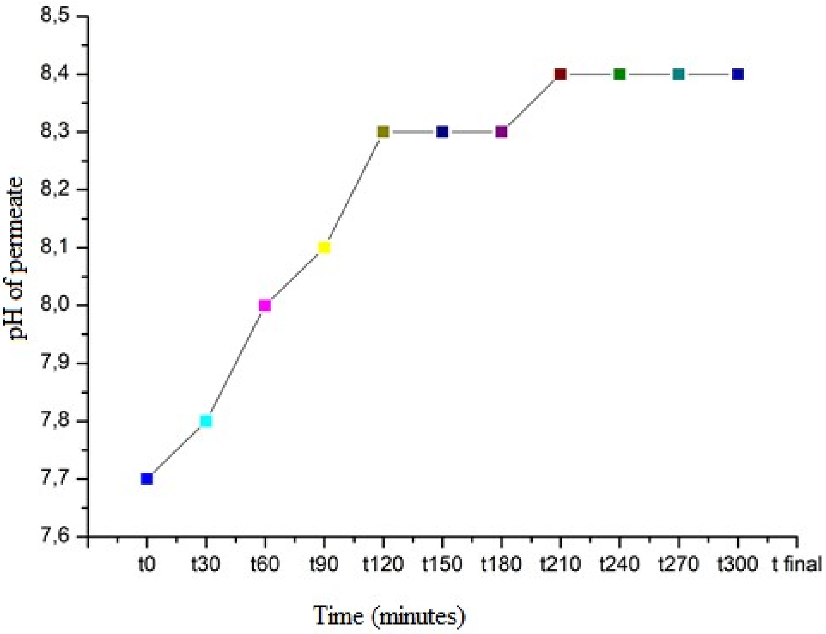

***Figure 7**. Variation of pH in each sample of permeate*

This means that the decrease in peptone concentration in the permeate samples renders water slightly basic as a result of its retention by the membrane.
Figure 8 shows the fluctuations of the main chemical indicators that were determined for each sample collected throughout the experiment.
Contrary to *p*H variation, dissolved oxygen varies dramatically until near the end of the experiment, where its value increases slightly. This increase can be explained by the fact that with the membrane force caused by fouling, some pressure is accumulated on its surface, causing the particles to penetrate through the pores, increasing the wear of the membrane.

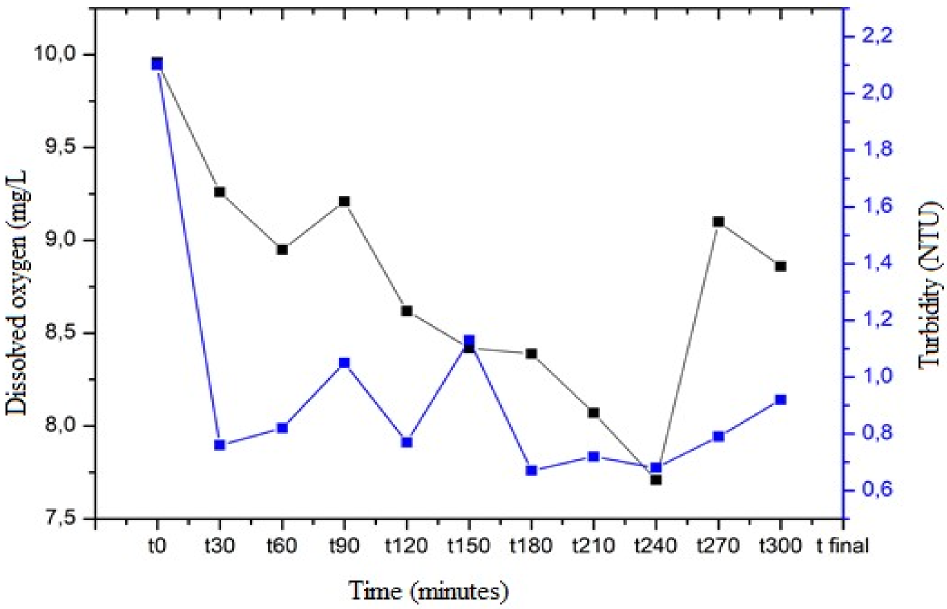

Figure 8. *Mutual influence of chemical indicators*

The turbidity in the feed and permeate samples was measured directly after samples collection. The continuous variation of turbidity demonstrates that the membrane functioned continuously without disturbing, since the ultrafiltration process did not require operation at high pressures for the entire experiment. Otherwise, the membrane would have been much more mechanically stressed and its wear and fouling would alter the smooth functioning of the physico-chemical parameters of the ultrafiltration plant.

Effectiveness of PAN membrane for peptone retention

Figure 9 shows the evolution over time of peptone retention on the membrane.
As shown in Figure 9, the membrane presented in the study registers a continuous increase in its effectiveness, but in a rather small percentage. Clearly, there is no decline in peptone retention, its slightly increasing trend demonstrates that the membrane can still function in good conditions and that it does not have any physical damage.

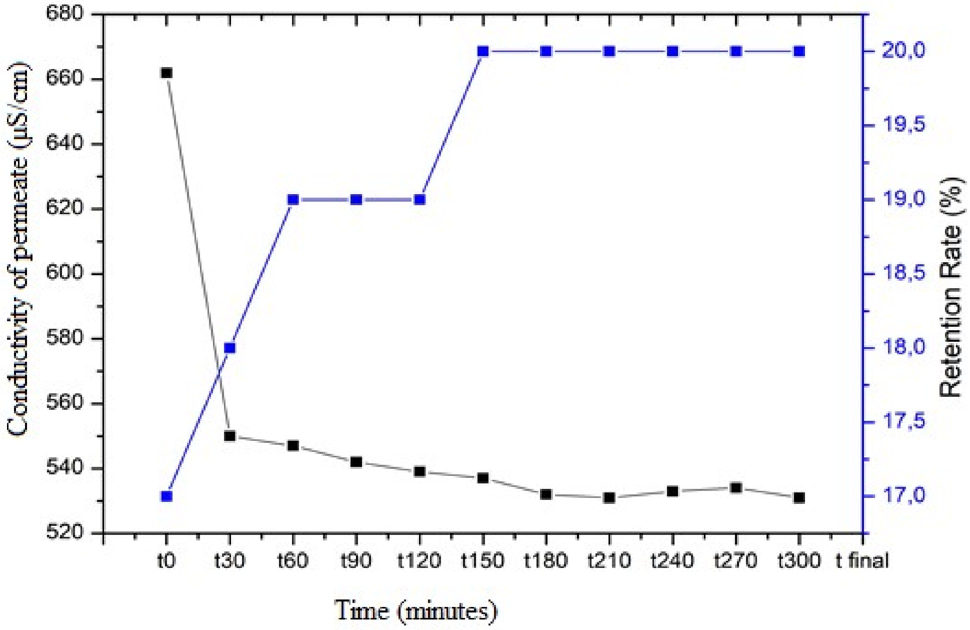

Figure 9. *Peptone retention rate by PAN membrane*

However, the recorded values of peptone retention on the membrane are small, barely reaching 20 % to the end.
As stated above, with regard to choosing the time of the 300 minute experiment, Figure 9 confirmed that membrane operation was not required for a longer time. From minutes 150 to the end of the experiment, the retention rate was virtually the same. In this case, due to the fine particles and the peptone properties, the PAN membrane organic membrane cannot largely retain this substance, but perhaps its combination with another type of substance can be prevented from passing through the membrane pores.

CONCLUSIONS

In this study, an organic hollow membrane of polyacrylonitrile was used for monitoring over time the peptone retention on its surface. The time for the study was 300 minutes because it was initially considered that this interval is enough to see if the membrane is forced or not during the experiment as a result of the peptone particle deposition on the filter surface thereof.
In fact, the relationship between the main parameters that were followed throughout the experiment showed that operating the system at a relatively low pressure ensures good membrane functioning and a prolonged life cycle.
The membrane filtration surface in this experiment was 0.6 m^2 (according to the installation manual) and the transmembrane operating pressure was 0.8 - 0.9 bar, with slight fluctuations.
It was expected that the peptone would not be largely retained because the small particles obtained after dissolution were readily passed through the membrane pores, making its retention efficiency consistent and not increasing, this being retained in only 20 %.

As with any membrane filtration process, the volume of permeate has decreased over time as the membrane deposition of the substance has been increasingly pronounced, the permeate volume recorded a decrease of about 2.5 liters over 300 minutes. In the present case, the peptone deposition was not emphasized because the system pressures did not change visibly, but the peptone was present in a fairly high concentration in the permeate samples that were collected after every 30 minutes.
The data recorded in this study showed that ultrafiltration of water containing only peptone is not very effective. However, wastewater from industries such as pharmaceuticals, food, etc. will not only contain peptone particles, and the combination of several substances may mean perhaps a better solution by which agglomeration of the particles is easier to retain on the effective filtration surface of any type of the membrane.

REFERENCES

1. Barsan, N., Nedeff, V., Temea, A., Mosnegutu, E., Chitimus, A. D., Tomozei, C.: A perspective for poor wastewater infrastructure regions: a small-scale sequencing batch reactor treatment system, *Chemistry Journal of Moldova,* **2017**, **12** (1), 39-46;
2. Barsan, N., Joita, I., Stanila, M., Radu, C., Dascalu, M.: Modelling wastewater treatment process in a small plant using a sequencing batch reactor (SBR), *Environmental Engineering and Management Journal,* **2014**, **13** (7), 1561-1566;
3. Acero, J.L., Benitez, F.J., Leal, A.I., Real, F.J., Teva, F.: Membrane filtration technologies applied to municipal secondary effluents for potential reuse, *Journal of Hazardous Materials*, **2010**, **177** (1-3), 390-398;
4. Tataru, L., Nedeff, V., Barsan N., Panainte-Lehadus, M., Mosnegutu, E.F.: Efficiency studies of hollow fiber organic membranes in restraining yeast from wastewater subjected to ultrafiltration, hydrology and water resources, *Proceeding of the International Multidisciplinary Scientific GeoConference Surveying Geology and Mining Ecology Management, SGEM 2018*, **2018**, **18** (3.1), 235-242;
5. Tataru, L., Nedeff V., Barsan N., Panainte-Lehadus, M., Mosnegutu E, Chitimus D.A.: Studies on the application of hollow fiber membranes in the field of ultrafiltration*, Journal Engineering Study Research*, **2016**, **22** (4), 42-53;
6. Tataru L., Nedeff, V, Barsan, N., Panainte Lehadus, M., Chițimuș, D.A.: The importance of using membranes in seawater desalination as a result of excessive exploitation of water sources, *The Annals of "Dunarea de Jos" University of Galati Fascicle IX. Metallurgy and Materials Science*, **2017**, **3**, 39-46;
7. Tataru, L., Nedeff, V., Barsan, N., Panainte-Lehadus, M., Mosnegutu, E. F., Chitimus, D.A., Fabian F.: A comparative study between water-gap and air-gap distillation membranes for removal of salts from saline water by desalination method, *Journal of Engineering Studies and Research*, **2018**, **24** (2), 46-57;
8. Fan, L., Harris, J.L., Roddick, F.A., Booker, N.A.: Influence of the characteristics of natural organic matter on the fouling of microfiltration membranes, *Water Research*, **2001**, **35** (18), 4455-4463;
9. Rojas, J.C., Perez, J.,Garralon, G., Plaza, F., Moreno, B., Gómez, M.A.: Humic acids removal by aerated spiral-wound ultrafiltration membrane combined with coagulation-hydraulic flocculation, *Desalination*, **2011**, **266** (1-3), 128-133;
10. Listiarini, K., Sun, D.D., Leckie, J.O.: Organic fouling of nanofiltration membranes: Evaluating the effects of humic acid, calcium, alum coagulant and their combinations on the specific cake resistance, *Journal of Membrane Science*, **2009**, **332** (1-2), 56-62;
11. Peiris, R.H., Jaklewicz, M., Budman, H., Legge, R.L., Moresoli, C.: Assessing the role of feed water constituents in irreversible membrane fouling of pilot-scale ultrafiltration drinking water treatment systems, *Water Research*, **2013**, **47** (10), 3364-3374;

12. Bârsan, N., Nedeff, V., Moşneguţu, E.F., Panainte, M.: Heat balance components of a small sequencing batch reactor applied for municipal wastewater treatment, *Environmental Engineering and Management Journal*, **2012**, **11** (12), 2131-2138;
13. Boyer, T.H., Singer, P.C.: Bench-scale testing of a magnetic ion exchange resin for removal of disinfection by-product precursors, *Water Research*, **2005**, **39** (7), 1265-1276;
14. Mergen, M.R.D., Jefferson, B., Parsons, S.A., Jarvis, P.: Magnetic ion-exchange resin treatment: Impact of water type and resin use, *Water Research*, **2008**, **42** (8-9), 1977-1988;
15. Zheng, X., Ernst, M., Jekel, M.: Stabilizing the performance of ultrafiltration in filtering tertiary effluent-Technical choices and economic comparisons, *Journal of Membrane Science*, **2011**, **366** (1-2), 82-91;
16. Huang, H., Young, T.A., Schwab, K.J., Jacangelo, J.G.: Mechanisms of virus removal from secondary wastewater effluent by low pressure membrane filtration, *Journal of Membrane Science*, **2012**, **409-410**, 1-8;
17. Löwenberg, J., Zenker, A., Baggenstos, M., Koch, G., Kazner, C., Wintgens, T.: Comparison of two PAC/UF processes for the removal of micropollutants from wastewater treatment plant effluent: Process performance and removal efficiency, *Water Research*, **2014**, **56**, 26-36;
18. Tang, S., Zhang, Z., Zhang, X.: New insight into the effect of mixed liquor properties changed by pre-ozonation on ceramic UF membrane fouling in wastewater treatment, *Chemical Engineering Journal*, **2016**, **314**, 670-680;
19. Tataru, L., Nedeff, V., Barsan, N., Mosnegutu, E., Panainte Lehadus, M., Sandu, I., Chitimus, D.: Studies of humic acid removal from aqueous systems by using polymeric membrane ultrafiltration process, *Materiale Plastice*, **2018**, **55** (4), 680-685;
20. http://ro.sp-samehealth.com/news/what-do-you-know-about-peptone-made-from-soybe-10330136.html, accessed November 14, **2018**;
21. http://ro.sp-samehealth.com/news/peptone-manufacturers-introduce-its-role-in-th-10345029.html, accessed November 14, **2018**;
22. http://himedialabs.com/TD/RM001.pdf, accessed November 14, **2018**;
23. Gumbi, N.N., Hu, M., Mamba, B.B., Li, J., Edward, N., Nxumalo, E.N.: Macrovoid-free PES/SPSf/O-MWCNT ultrafiltration membranes with improved mechanical strength, antifouling and antibacterial properties, *Journal of Membrane Science*, **2018**, **566** (15), 288-300;
24. ****Ultrafiltration pilot MP 90, version AM-03/2014, Technical Book*, DELTALAB SMT France.