

Elimination of Penicillin V by Membrane Process

Messaouda Samai^a, Mustapha Chikhi*^a, Ferhat Bouzerara^b

^aLaboratoire de l'Ingénierie des Procédés de l'Environnement,
 Département de Génie de l'Environnement, faculté de Génie des Procédés Pharmaceutiques, Université Constantine 3.
 Constantine, Algérie.

^bDépartement de physique, faculté des sciences exactes et informatique, Université de Jijel, Jijel, Algérie.
 chikhi_mustapha@yahoo.fr

Recovery of penicillin V in wastewater from a membrane microfiltration process, aims to minimize the pharmaceutical pollution in our discharges. This study is carried out using a synthetic solution of penicillin V, at various concentrations, in a tubular microfiltration ceramic membrane; the tangential filtration is very convenient, it reduces the accumulation of particles on the membrane surface. Different parameters were studied, the change in permeate flux versus time for different pH values of solution, the volumes of the retentate and the permeate as a function of the concentration of the penicillin V, variation of the turbidity of the permeate and the retentate as a function of time; the obtained results show that the permeate flux is important for a low concentration of penicillin V (0.01 g/L), the volume of the permeate decreases and of the retentate increases with increasing concentration of penicillin V. basic pH are very favorable, they can get best yields microfiltration of the solution. The turbidity of the retentate increases, while that in the permeate it decreases as a function of time, showing that the penicillin V is recovered by the microfiltration membrane.

1. Introduction

It is necessary to advance knowledge on the various water contaminants, including drug induced pollution in wastewater, in view of the implementation of new laws by the authorities (Kangmin et al., 2014). Moreover, these products act on biomass treatment plants, causing a decrease in performance of these systems, particularly in terms of the elimination of phosphorus and nitrogen; where the risk of eutrophication of aquatic environments (Shimiao et al., 2014). Antibiotics are essential elements of human and veterinary medicine that improve our quality of life, but so far there was little information on the levels of antibiotic residues in the environment (Byung et al., 2007). It should also be noted that the release pharmaceutical industries: 1% of the production is in the effluent (Yasemin et al., 2013). Two different types of pharmaceutical pollution can be cited for wastewater. Those found in the pharmaceutical plants and other that are found in the water purification plant. It must be very careful about the influence of pharmaceuticals and antibiotics especially on the water surface. The membrane processes can be used as one of methods for the removal of substances by ultrafiltration or microfiltration (Jardel et al., 2011). This technique has been chosen to focus the molecules or ionic species in solution, or for separating particles or microorganisms in suspension in a liquid (Jing et al., 2013). In this study, the ceramic membrane was used in a tubular module, this geometry allows the flow of liquid at high speed while minimizing the accumulation of the substance on the surface of the membrane (Mohammad et al., 2013). The purpose of this study is to concentrate the solution containing penicillin V and decontaminate the water by using a microfiltration tangential flow.

2. Materials and method

The experimental work consists of placing a solution of the synthetic penicillin V, previously prepared at different concentrations in a feed vessel; the pump ensures the circulation of liquid through the cylindrical microfiltration module, pressure is controlled by the two pressure gauges before and after the filtration; it is

increased by the introduction of the compressed air within the feed vessel; a graduated cylinder allows to recover the retentate, while the concentrated solution (concentrate) is recirculated to the feed tank; the monitoring of the volume of the retentate as a function of time allows to determine the flow knowing the surface of the membrane; the membrane used is made of ceramic zirconium dioxide ZrO_2 . It should be noted that the clogging phenomenon still exists, though it is less important than the frontal filtration, membrane cleaning is essential, it consists to use a basic solution (NaOH 0.1 mol/L) then a solution of distilled water for 30 min, and finally an acid solution (HCl: 0.1 mol / l) for 60 min followed by washing with distilled water for 30 min before tackling new experiences. A turbidimeter was used in this study to determine the turbidity of the permeate and retentate solutions; the experimental setup is shown in Figure 1.

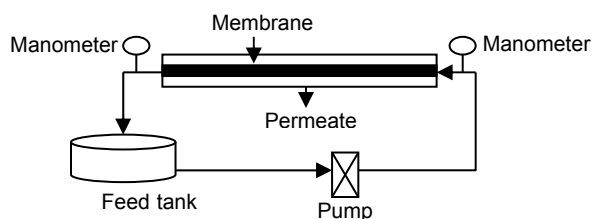


Figure 1: Microfiltration apparatus

The experimental assembly is mainly composed of a ceramic membrane filter, the pump used to circulate the liquid to be filtered from the feed tank to the membrane, the filtrate passes through the membrane and the solution is recirculated to the concentrate feed tank, a manometer controls the liquid pressure.

3. Results

The obtained results in this study are presented in this section, namely the change in permeate flux as a function of time for various initial concentrations; determining volumes of the retentate and the permeate as a function of time, the effect of pH ; monitoring turbidity of the retentate and permeate with time for various initial concentrations of penicillin V.

3.1 Variation of permeate flux

The permeate flux reflecting the amount of water containing the particles which could pass through the microfiltration membrane, its variation as a function of time indicates that there is a deposition of substances on the surface of the membrane. Filtration is carried out for a constant rate and a temperature of 24 °C, the pH of the initial solution of penicillin V varies between 3 and 11.

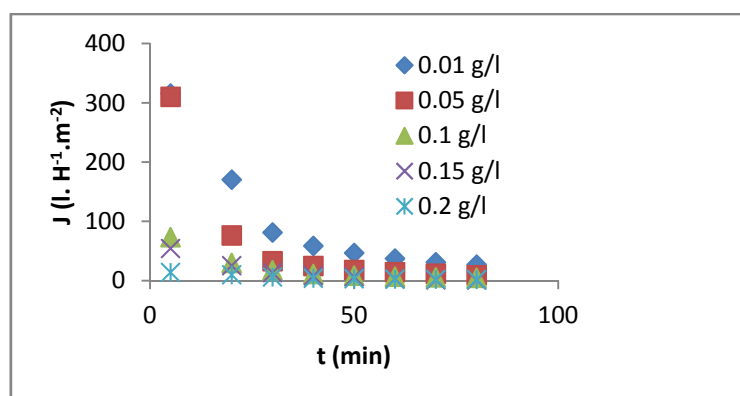


Figure.2: Variation of the volume flow of the penicillin V as a function of time at pH = 3

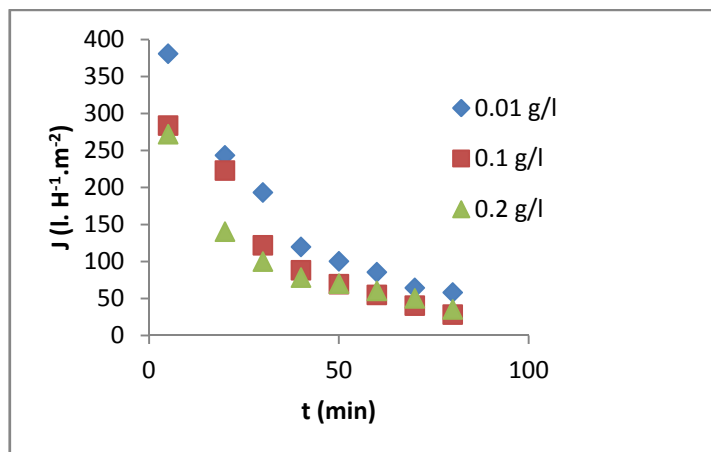


Figure 3: Variation of the volume flow of the penicillin V as a function of time at pH = 9

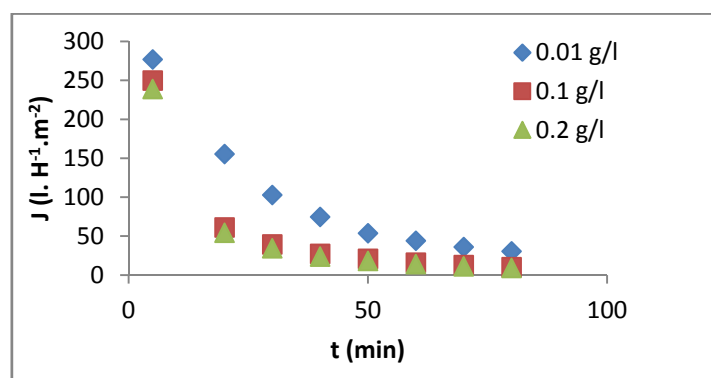


Figure 4: Variation of the volume flow of the penicillin V as a function of time at pH = 7

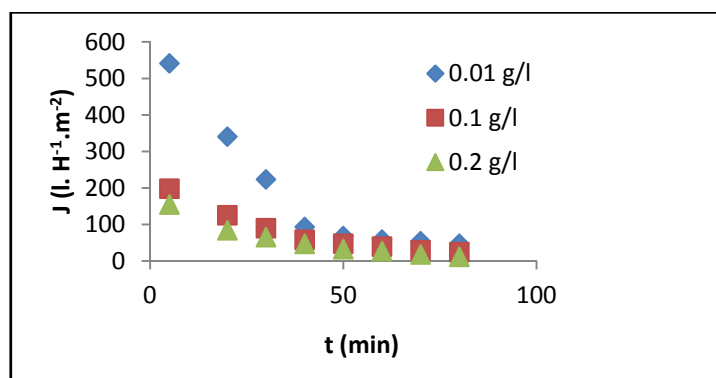


Figure 5: Variation of the volume flow of the penicillin V as a function of time at pH = 11

According to Figures 2, 3, 4 and 5, we note that the permeate flux decreases with time, it decreases quite rapidly when the initial concentration of penicillin increases, we see that this flux can tend towards zero at high time (70 and 80 minutes of pH 11 and for some low time when the pH is equal to 3), this tendency toward zero of the permeate flux indicates that we are in the presence of a serious clogging problem. It should be mentioned that the pH of 9 correspond to the better filtration giving greater fluxes.

3.2 Change in volume of permeate and retentate

The change in volume of permeate and retentate, depending on the initial concentration of penicillin V and for a definite time filtration, is shown in Figure 6, with the following operating conditions: pH=3.18, T=24 °C.

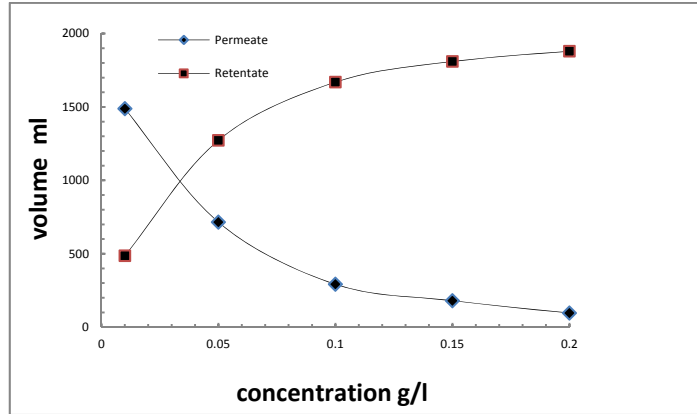


Figure 6: Variation of permeate and retentate volumes with the initial concentration of penicillin V

It is found that the volume of the permeate decreases with the initial concentration of penicillin V, for against that of the retentate increases toward the volume of the original solution, this observation is the same as the variation of the permeate flux, which explains that the microfiltration membrane stops penicillin V for forming a deposit on its surface, and this phenomenon is more significant when the initial concentration increases.

3.3 Influence of pH

The influence of pH on the change in permeate flux as a function of time is presented in this part, and this for different initial concentrations of the penicillin V.

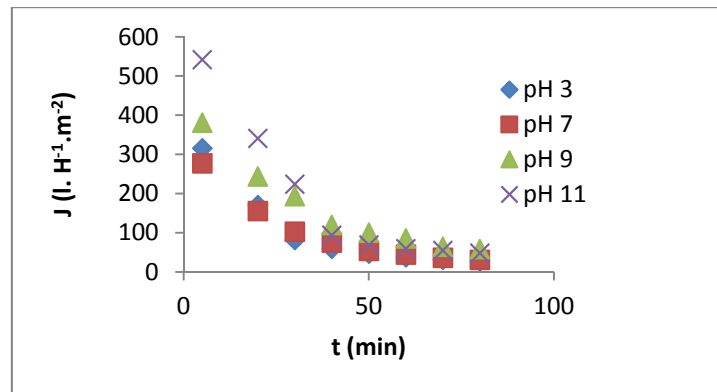


Figure 7: Variation of the permeate flux of penicillin V with time at C = 0.01 g/L (for different pH)

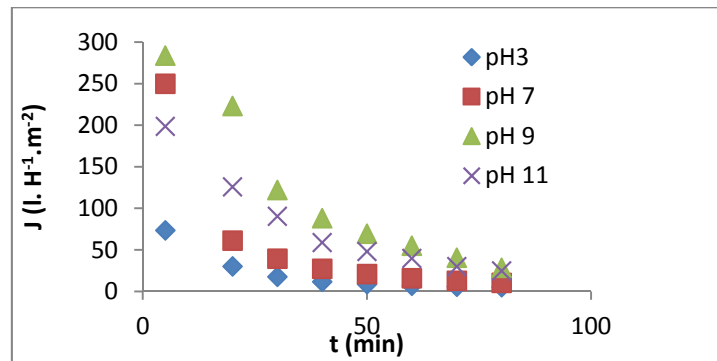


Figure 8: Variation of the permeate flux of penicillin V with time at C = 0.1 g/L (for different pH)

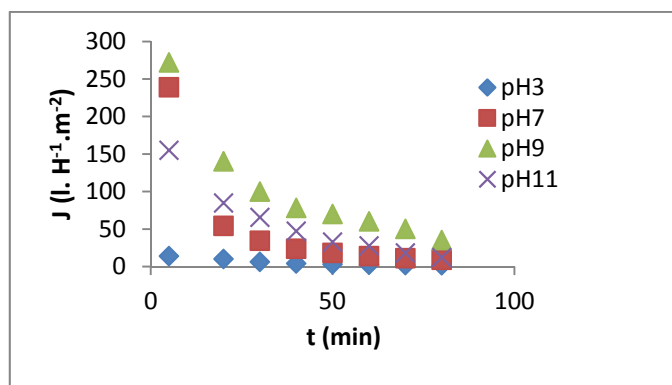


Figure 9: Variation of the permeate flux of penicillin V with time at $C = 0.2$ g/L (for different pH)

It is observed from Figures 7, 8 and 9 that the permeate flux always decreases with time, it is important when the solutions are basic at pH 9 and 11, this variation may be due to the nature of the membrane (inorganic), so we can say that the clogging of the membrane is delayed when the solution becomes basic.

3.4 Monitoring of turbidity

Figures 10 and 11 show the variation of the turbidity of the retentate and the permeate as a function of filtration time.

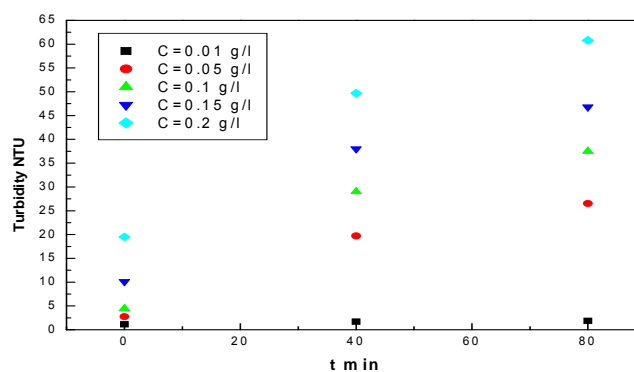


Figure 10: Changes in the turbidity of the retentate as a function of time

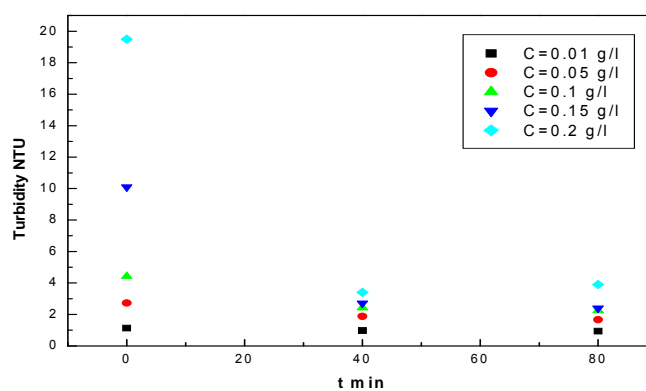


Figure 11: Changes in the turbidity of the permeate as a function of time

This parameter allows to know the presence of undissolved and dissolved penicillin V in the solvent before and after filtration, the pH of the solution is 3 (without addition of acid or base). It is found that the turbidity increases over time and with the initial concentration of penicillin for the retentate, because it will be concentrated at the end of the filtration; against the turbidity of the permeate decreases with time and with the initial concentration of penicillin, this confirms that the microfiltration membrane retains penicillin V initially contained in the solution, the permeate is rich in water.

4. Conclusion

Because of their ability to perform the separation, which is very specific and without phase change at very high or ambient temperatures, the microfiltration is therefore an effective method for the removal of penicillin V. The parameters monitored in this study show that the permeate flux decreases with time, and especially when the concentration of penicillin V increases, basic solutions (pH 9 and 11) show better removal by microfiltration, because in these cases the permeate flux is more important for the same operating conditions. The determination of the turbidity of the permeate and retentate confirms that there is significant elimination of penicillin V. The clogging problem of the membrane remains the major disadvantage of this technique; it causes a significant reduction in the permeability of the membrane.

References

- Byung S. O., Ha Y. J., Tae M. H., Joon-Wun K., 2007, Role of ozone for reducing fouling due to pharmaceuticals in MF (microfiltration) process, *Journal of Membrane Science*, 289(1–2), 178-186.
- Jardel P., Araujo P., Peres L., Application of ultrafiltration membranes in the separation of Ethylic Route Biodiesel, *Chemical Engineering Transactions*, 24, 769-774, DOI: 10.3303/CET1124129.
- Jing F., Guotong Q., Wei W., Xinqing Z., Lei J., 2013, Elaboration of new ceramic membrane from spherical fly ash for microfiltration of rigid particle suspension and oil-in-water emulsion, *Desalination*, 311, 113-126.
- Kangmin C., Jaeweon C., Seung J. K., Am J., 2014, The role of a combined coagulation and disk filtration process as a pre-treatment to microfiltration and reverse osmosis membranes in a municipal wastewater pilot plant, *Chemosphere*, 117, 20-26.
- Mohammad A. K., Mohammad S., Mehdi Y., 2013, Mathematical modeling of crossflow microfiltration of diluted malt extract suspension by tubular ceramic membranes, *Journal of Food Engineering*, 116(4), 926-933.
- Shimiao D., Eun-Sik K., Alla A., Hiroshi N., Yang L., Mohamed G. E.-D., 2014, Treatment of oil sands process-affected water by submerged ceramic membrane microfiltration system, *Separation and Purification Technology*, 138, 198-209.
- Yasemin K., Gamze E., Ilda V., Z. Beril G., Gulsum Y., Nadir D., Coskun A., 2013, The treatment of pharmaceutical wastewater using in a submerged membrane bioreactor under different sludge retention times, *Journal of Membrane Science*, 442, 72-82.