NANOFILTRATION REJECTION OF PESTICIDES USED IN THE MEKONG DELTA AREA

D. L. Nguyen, S. Gassara, P. Sistat, A. Deratani

IEM (Institut Européen des Membranes), UMR 5635 (ENSCM-UM-CNRS), Université de Montpellier, Place E. Bataillon, F- 34095, Montpellier, France

Abstract

The pollution of surface water by recently used pesticides in Mekong Delta, a main drinking water source, is increasing both on the concentration residues and the polluted area [1,2]. Nanofiltration (NF) is arising as the most appropriate method for removal of pesticides with not only a good efficiency but also low energy consumption in comparison with other pressure-driven membrane processes [3]. The performance of NF membrane in removal of the three most frequent detected pesticides (fenobucarb, isoprothiolane and pretilachlor) in aqueous solution was investigated using NF90 and NF270 membranes (Dow Filmtec). The adsorption of pesticides on the NF membranes and their adsorption kinetic were studied in batch experiment in order to evaluate its effect on rejection of pesticides. The membrane separation experiments were carried out with synthetic model matrices using a crossflow filtration pilot. The influence of applied pressure and coexisting pesticides and salts like those present in real river water was investigated. It was found that the adsorption levels off after 48h, the amount of adsorbed pesticide being proportional to its hydrophobicity in a single pesticide system and also in a mixture of the three pesticides. The results of rejection showed that NF90 membrane is a crucial choice for removing pesticide with very high rejection. The NF270 membrane exhibited much higher permeate flux but lower selectivity. The rejection of each pesticide can be explained by the combined influence of their molecular weight and dipole moment, whereas the performance in the mixture of three pesticide solution seems to be influenced by a competitive adsorption of pesticides on the membrane surface. The transmembrane pressure had no drastic effect on the pesticide rejection. The presence of salts tends to reduce the pesticide rejection in all cases, while the rejection of salts seems to be independent of the presence of pesticides.

1 Materials and methods

1.1 Membrane selection

Two kinds of flat-sheet-type nanofiltration membranes used in this study are NF270 and NF90. These are popular commercial membranes of Dow Filmtec and useful for removing organic compounds as well as salinity in water.

1.2 Target pesticides

Three target pesticides in this study are used at their standard chemical purity from Sigma Aldrich: fenobucarb (purity 97.0%), isoprothiolane (purity 98%) and pretilachlor (purity 98.7%). They are detected great frequency in surface water in Mekong Delta. They are representative for 3 main kinds of pesticide depending on their function: insecticide, fungicide and herbicide.

1.3 Synthesis Mekong river water

For the study of the influence of coexisting inorganic salts in feed solution on the rejection of pesticides, some synthesis surface water samples like the Mekong Delta are prepared. The composition of the synthesis surface water is chosen depending on occurrence of salts in real surface water in the Mekong River.

2 Results and discussion

2.1 Adsorption of pesticides on NF membrane

The amount of fenobucarb, isoprothiolane and pretilachlor adsorbed on membrane NF270 and NF90 in the single solute system and in the mixture of solutes system after 48 hours contact is shown in figure 1. These results obtain from the batch adsorption experiment without pressure, so they only reflect the interaction between membrane surface and pesticide compound. The error bars indicating the accuracy of experimental results are standard deviation of duplicate experiments. The results of adsorption on both membranes in the single solution as well as in the mixture solution are, in general, quite different for the three pesticides. The adsorption of pretilachlor is the highest, followed by isoprothiolane and fenobucarb. This is attributed to hydrophobicity of the three pesticides, pretilachlor being more hydrophobic than isoprothiolane and fenobucarb with log K_{ow} 4.08, 3.3 and 2.78, respectively.

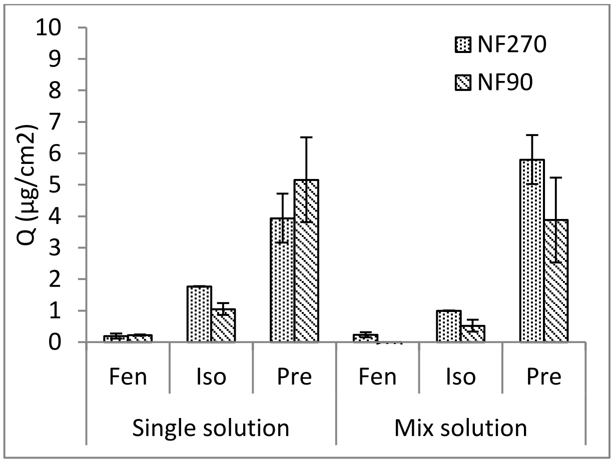

Figure 1. The adsorption of pesticides on NF membranes in single solute system and in mixture of solutes system (Fen: Fenobucarb; Iso: Isoprothiolane; Pre: Pretilachlor)

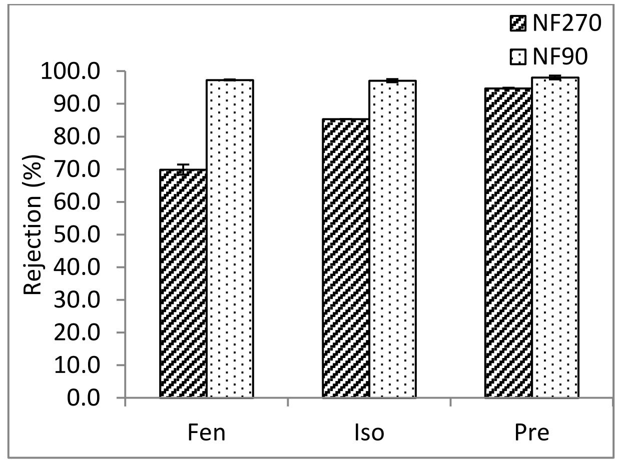

Figure 2. Rejection of the pesticides in pure solution (concentration 300 – 1000µg/l, pressure 11 bar)

2.2 Rejection of pesticides in pure water solution

2.2.1 Rejection of single solution

The rejection of fenobucarb, isoprothiolane and pretilachlor by membrane NF270 and NF90 at the applied pressure 11 bar is depicted in Figure 2. Generally, the rejection by the NF90 membrane is higher than the NF270 membrane. A similar rejection for all of pesticides used is observed in case of the NF90 membrane with the values more than 95%. This can be explained by the MWCO of the NF90 membrane, which is smaller than the molecular weight of the pesticides. This result is in agreement with the recommendation of its manufacturer. Since this membrane would have predominantly exhibited size exclusion mechanism.

In the case of the NF270 membrane, with relatively wide-pore size, the difference on rejection of the three pesticides is somewhat larger than that by the NF90 membrane, from 70% to 94%. The results show that the rejection of pesticides conforms to the trends of increased rejection with increasing molecular weight of the pesticides.

2.2.2 Effect of transmembrane pressure

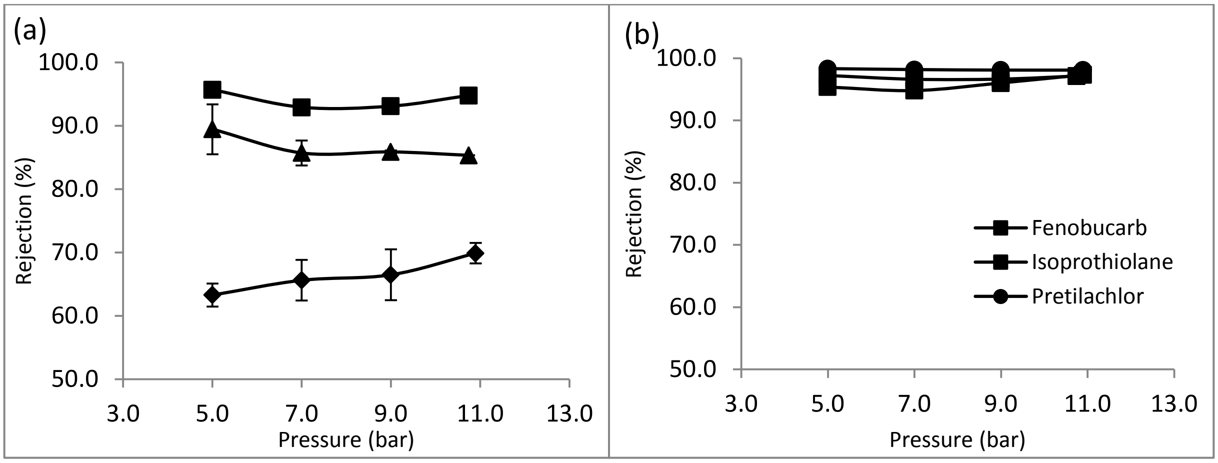

Figure 3. Pesticide rejection in different pressure applied in the case of (a) NF270 and (b) NF90 membrane.

The rejection results of the three pesticides in different transmembrane pressure are presented in figure 3. In summary, the transmembrane pressure seems have a little effect to rejection of pesticides, especially in the cases of NF90 membrane which has rejection higher than 95%. This no effect on transmembrane pressure demonstrates that the transport through this membrane cannot be explained by the solution-diffusion model. But this is a separation by steric hindrance.

2.3 Effect of coexisting pesticides

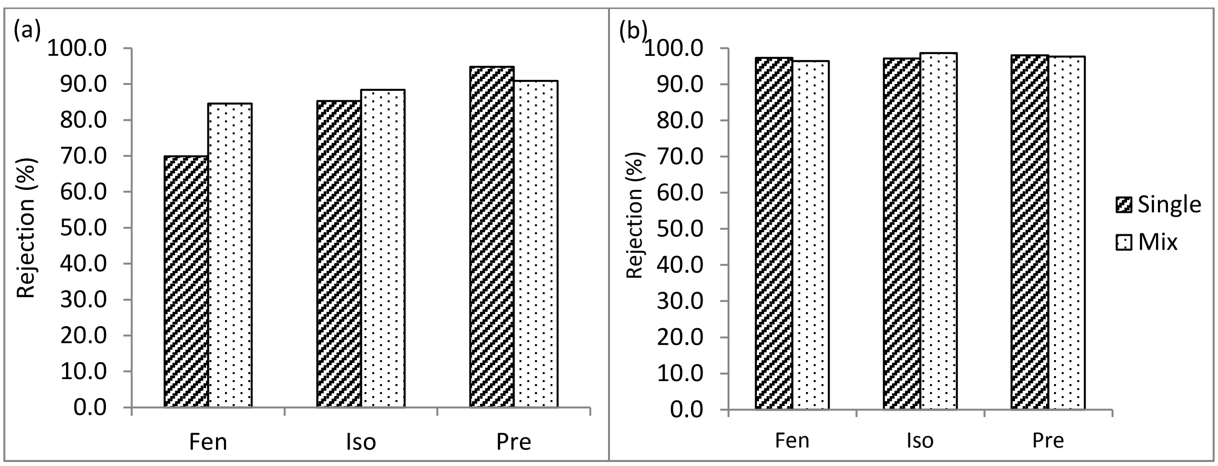

Figure 4. Rejection of pesticides in single solution and in mixture solution at pressure 11 bar by (a) NF270 and (b) NF90 membrane

The effect of coexisting pesticides on the rejection performance of the NF270 and NF90 membrane is evaluated by comparison removing efficiency in case of single pesticide solution and the mixture of the three pesticides solution. The experiment results are shown in figure 4. For the NF270 membrane, influence of coexisting pesticide is different depending on the competitive absorption of them on the surface of membrane as well as the hindrance effect together. For pretilachlor, rejection value of the mixture pesticide experiment is smaller than that of the single pesticide experiment. Conversely, the rejection of fenobucarb and isoprothiolane increases in case of the mixture pesticide experiment. These results might be explained by the increasing absorption of pretilachlor and the decreasing adsorption of isoprothiolane on the membrane NF270 in the mixture solution. In case of the membrane NF90, the pesticide rejection does not seem to be significantly influenced by the presence of other pesticides. It is attributed by the very high rejection by the membrane NF90.

2.4 Rejection of pesticides in synthetic representative river water

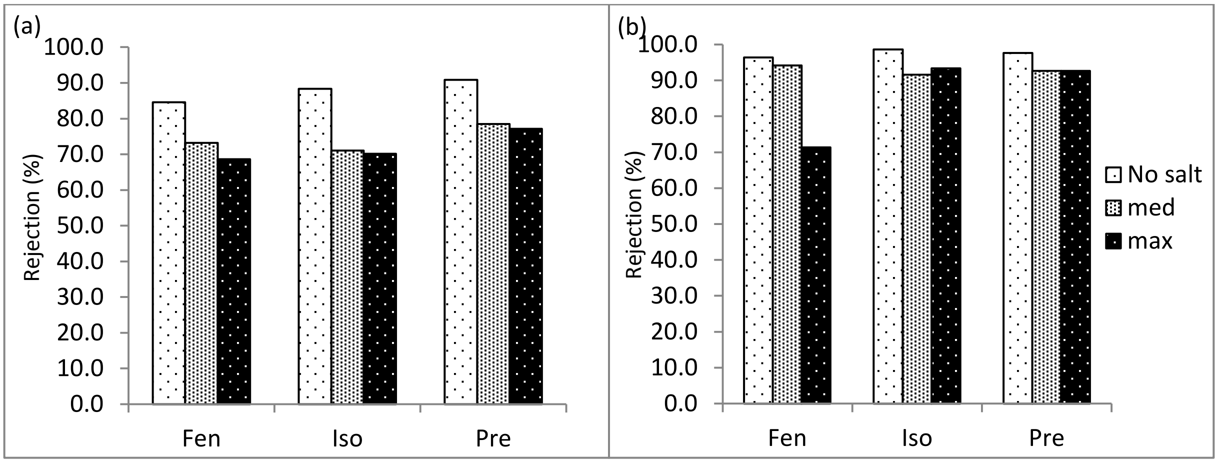

Figure 5. The rejection of pesticides from three water matrices by (a) NF270 and (b) NF90 membrane (no salt, from pure water without salt; Med, from the synthesis water with median concentration value of salts; Max, from the synthesis water with maximum concentration value of salts)

The rejection of fenoducarb, isoprothiolane and pretilachlor from three water matrices is shown in figure 5. When the comparing the different matrices, it is observed that the rejection from pure water is generally higher than those from the synthesis river water, for all pesticides and membranes used. In the case of our study, both the swelling of pores and the decreasing of pesticide Stokes radius are considerable reasons for the reduction in rejection of the pesticides when salt concentration increases. Because there is an increasing of pure water flux after each membrane operation observed.

3 Conclusions

Surface water in Mekong Delta is a main source for drinking water. But the pesticide pollution threatened the source as well as human health. This study shows that nanofitration process is a potential solution to remove pesticides in surface water. The NF90 membrane shows as a crucial choice for removing pesticide when its performance is better than that of the NF270 membrane. The very high rejection by the tight NF90 membrane induces the influence of pressure applied, coexisting pesticide and adsorption of pesticide on membrane performance becoming negligible.

In the single pesticide solution, the rejection of pesticides increases in a corresponding with increasing of molecular weight of pesticides. Among the three pesticides, pretilachlor display the greatest rejection, because of its greatest molecular size, followed by isoprothiolane and fenobucarb.

The absorption of pesticides influences the percentage of pesticides removal, especially in the mixture pesticide solution. For pretilachlor, the rejection value by the NF270 membrane form the mixture pesticide solution is smaller than that from the single pesticide solution because of the increasing absorption amount in the mixture case. Likewise, the rejection of isoprothiolane somewhat increases from the mixture when its absorption amount decreases.

In the presence of salts, the rejection tends to reduce for all pesticides and membranes used. Especially, there is an increasing of pure water flux after each experiment with the presence of salts in both cases of the two membranes. This shows that both the swelling of pores and the decreasing of pesticide Stokes radius are considerable reasons for the reduction in rejection of the pesticides when salt concentration increases.

In addition, the increasing of the transmembrane pressure posed no effect on the pesticide rejection. The hindrance effect of the big molecular from the mixture of pesticides solution also affects to the rejection of small one.

Reference

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[3] K. V. Plakas and A. J. Karabelas, “Removal of pesticides from water by NF and RO membranes — A review,” *Desalination*, vol. 287, pp. 255–265, Feb. 2012.