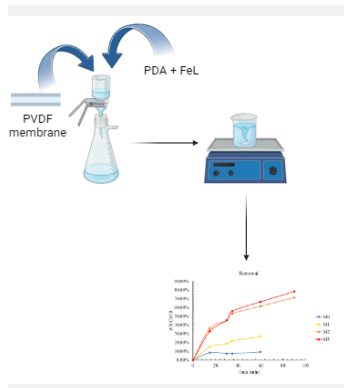


Promising pollutant removal using FeL-impregnated polymeric membrane

ORAL
Ph.D. Student: N
Journal: JECE

L.C. Souza¹, F.P. da Silva¹, A.M.F. Linhares¹, F.V. Fonseca¹. (1) Universidade Federal do Rio de Janeiro, Av. Athos da Silveira Ramos, 149 - Escola de Química, Rio de Janeiro, Brasil, larissacarreiro@eq.ufrj.br.



Coordination compounds are formed when ligands react with one or more metal centers. Catalytic membranes, which combine catalytic activity with conventional filtration membranes, have shown promise in degrading organic compounds. This study aimed to develop catalytic membranes for removing organic pollutants in a bench-scale system. Commercial membranes were impregnated with iron complex (FeL) catalyst, and removals of up to 78% were achieved for the model molecule used at pH 6.5. The membranes were functionalized with the catalyst, and a hydrogen peroxide concentration of 11.7 mmol/L was found to be effective. These results suggest that FeL impregnated membranes have potential for the degradation of pollutants in the present study.

Introduction

A membrane acts as a barrier that separates two phases and regulates the transport of chemical species in a specific way^[1]. Membrane separation processes (MSP) offer numerous advantages, such as energy conservation, ease of operation and scaling, and selectivity in separation. However, these processes still face the challenge of reduced flow and useful life of the membrane owing to fouling problems, which impose excessive costs on the process.

One promising solution to solve this problem is to combine MSP with advanced oxidative processes (AOP) utilizing catalytic membranes. This hybrid process merges catalytic activities with traditional filtration membranes, allowing for simultaneous physical separation and chemical oxidation in a combined system^[2]. This integration presents several advantages for membrane filtration processes. The objective of this study was to develop catalytic membranes for bench-scale tests involving the removal of organic compounds. The model molecule employed was the red dye Drimaren X-6BN.

Material and Methods

The complex (FeL) was synthesized under magnetic stirring for 2h by adding a methanolic solution of iron (III) chloride hexahydrate (1:2) to the ligand (L) obtained by the condensation of salicylaldehyde with ethanolamine (1:1)^[3]. FeL was then precipitated and filtered through a qualitative filter paper.

The membranes were obtained using a methodology described in the literature^[4] with some modifications. Commercial PVDF membranes were immersed in ethanol for 1 h to remove surface impurities, and then washed with distilled water for further use. A Tris-HCl buffer solution (40 mmol/L, pH 8.5) was prepared, and then 0.2 g of dopamine (PDA) was

added and dissolved in 40 mL of this solution in falcon tubes. PDA was cross-linked at 160 rpm for 1 h. The membrane was then immersed in cross-linked PDA for 3 h. Finally, the catalyst mass was added to the cross-linked PDA and filtered under vacuum by suction on the membrane. The functionalized membranes were characterized by contact angle, water permeance, X-ray Diffraction (XRD), and Scanning Electron Microscopy with Energy Dispersive Spectroscopy (SEM-EDS).

Finally, removal tests were performed on a shaking table in Erlenmeyer flasks with 250mL of 20 mg/L Drimaren Red (VDX) solution. The tests were carried out in duplicate, for one hour, at room temperature (25°C), 160 rpm, pH 6.5, 11.70 mmol/L of hydrogen peroxide (H₂O₂), and different catalyst concentrations (M0 = 0 mg/L; M1 = 0.28g/L; M2 = 0.56g/L; M3 = 0.84 g/L).

Results and Discussion

The membrane characterization results demonstrated that the impregnation technique was successful. The hydraulic permeability test revealed a substantial increase in water permeance for the M3 membrane, as compared to the commercial membrane. This result indicated that FeL loading was effective in enhancing water flux. This conclusion was further supported by the contact angle analysis, as the angle decreased from 56.02° for the commercial PVDF membrane to 27.28° for the M1 membrane, indicating a notable improvement in the hydrophilicity of the membrane. The SEM image in Figure 1 shows agglomerated FeL on the surface of the membrane, and EDS confirmed the presence of Fe in the functionalized membrane.

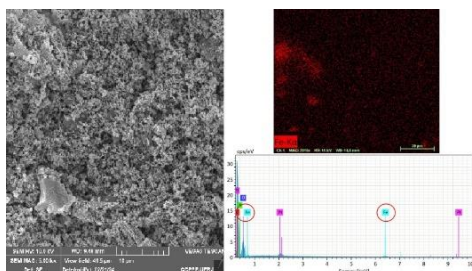


Figure 1. SEM and EDS of the M1 membrane

Figure 2 shows the decrease in the concentration of Drimaren Red dye after contact with the membranes functionalized with the FeL catalyst (M1, M2, and M3). In contrast, it is possible to observe a constant concentration of the solution in contact with the commercial PVDF membrane.

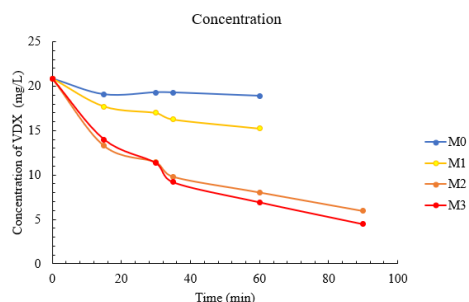


Figure 2. VDX concentration over time. Conditions: pH 6.5 and H_2O_2 concentration = 11.7 mmol/L.

Regarding the removal rate, it is possible to observe in Figure 3, at the end of 60 min, that the removal was greater in the membranes with the two highest

concentrations of FeL, with removals of 71 and 78% for M2 and M3 membranes, respectively. Such results suggest that membranes loaded with FeL are promising for degrading the pollutant used in the present study.

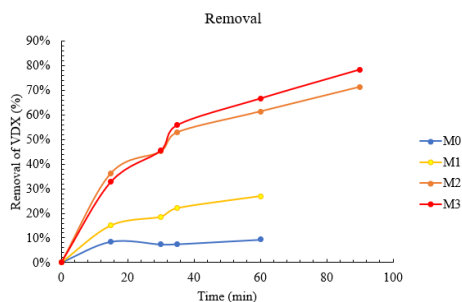


Figure 3. VDX removal (%) over time. Conditions: pH 6.5 and H_2O_2 concentration = 11.7 mmol/L.

Conclusions

Based on the results of the analysis, it can be concluded that the impregnation technique was successful. This implies that membranes impregnated with FeL have great potential for degrading the pollutants used in this study. Removals of up to 78% were obtained for the model molecule used at pH 6.5 with the membranes functionalized with the catalyst and a hydrogen peroxide concentration of 11.7 mmol/L. Therefore, the study can proceed with Diclofenac Potassium, which is a nonsteroidal anti-inflammatory drug that has recently gained attention.

Acknowledgments

The authors would like to thank the Coordenação de Aperfeiçoamento de Pessoal de Nível Superior (CAPES) and the Fundação de Amparo à Pesquisa do Estado do Rio de Janeiro (FAPERJ) (Grant number E-26/200.918/2021), Brazil, for financial support.

References

- [1] ROUT, Prangya R. et al. Treatment technologies for emerging contaminants in wastewater treatment plants: A review. *Science of the Total Environment*, v. 753, p. 141990, 2021.
- [2] QING, Weihua et al. Functional catalytic membrane development: A review of catalyst coating techniques. *Advances in Colloid and Interface Science*, v. 282, p. 102207, 2020.
- [3] SOUZA, L. C.; SILVA, F. P.; LINHARES, A. M. F.; FONSECA, F. V. . Degradação de compostos orgânicos em processo oxidativo mediado por um complexo de Ferro (III). In: Cleiseano Emanuel da Silva Paniagua. (Org.). *Soluções Inovadoras em Engenharia Sanitária e Ambiental*. v. 1, p. 261-273.
- [4] SUN, Xin et al. Fabrication of FeOCl/MoS₂ catalytic membranes for pollutant degradation and alleviating membrane fouling with peroxymonosulfate activation. *Journal of Environmental Chemical Engineering*, v. 10, n. 3, p. 107717, 2022.