

# BIOFOULING CONTROL BASED ON IRON NANOPARTICLE COATING FOR RO MEMBRANES

**Authors:** *María Magdalena Armendáriz-Ontiveros, Alejandra García-García, Leandris Argente-Martinez, Jesus Álvarez-Sánchez, Gustavo Fimbres-Weihs*

**Presenter:** *María Magdalena Armendariz-Ontiveros, MSc.*  
PhD Student - Instituto Tecnológico de Sonora - Mexico  
mcthe@hotmail.com

## **Abstract**

The effect of biofouling control measures, based on iron nanoparticle (FeNP) coating for RO membranes, was determined using accelerated biofouling experiments on a commercial polyamide. The zeta potential, XRD, roughness, contact angle, permeance and flux were determined for coated and uncoated membranes. Feed water was obtained from the Sea of Cortez, Mexico, which was pretreated, sterilized and inoculated with a high concentration ( $10^9$  CFU ml<sup>-1</sup>) of bacteria found naturally in the Sea of Cortez. In order to evaluate the anti-biofouling effect, the total organic carbon, total cell count and optical density were determined for the biofilm cake layer. The FeNP coating resulted in a significant reduction (32%) in the membrane permeance for pure water. Nonetheless, after the biofouling test the coated membranes showed 90% less biofilm cake layer, 67% lower total cell count and a 42% reduction in optical density. This highlights the biocide effect of the FeNPs, which could have a practical and important application in sea water desalination. Due to the easy synthesis of FeNPs, they have the potential to extend the useful life of RO membranes without significantly increasing membrane production costs, thus decreasing the total water cost.



## I. INTRODUCTION

Reverse Osmosis (RO) is presently the most commonly used technology in desalination worldwide, as it represents 65% of all installed capacity in the world [1]. However, RO membranes have fouling problems due to their physical and chemical surface properties that make them prone to organic and biological fouling (biofouling) [2].

Interest in RO membrane surface modifications using nanoparticles (NPs) as a measure to mitigate biofouling has been increasing, because NPs have been shown to present antimicrobial characteristics. Recent studies have coated membranes with Ag, Zn, and Cu NPs, as well as with other NP materials. It has been found that size, shape, high dispersion and strong adhesion of NPs on the surface of membrane significantly regulate the antibacterial activity [3].

However, most studies into the antibacterial properties of NPs have been carried out using AgNPs, which are particularly costly and thus increase membrane production costs. Iron nanoparticles (FeNPs) also possess antimicrobial properties and cost less than AgNPs. Hence, FeNP coatings are an attractive alternative for mitigating biofouling on RO membranes. This research aims to coat a commercial RO membrane with FeNPs in order to study their effect and potential to reduce biofouling problems in desalination plants.

## II. METHOD

FeNPs were synthesized following the methodology of Arancibia-Miranda et al. [4]. The surface of a polyamide (PA) composite membrane (Dow Filmtec SW30HR) was modified by depositing FeNPs onto the PA layer via immersion.

Prior to coating, the chemical structure and surface of the FeNPs were characterized by scanning electron microscopy (SEM) and X-ray Diffraction (XRD). The XRD analysis was carried out in a XRD diffractometer (Shimadzu, XRD-6000). SEM micrographs were collected from a field emission scanning electron microscope (FE-SEM) (Nova NanoSEM-200, FEI Company) to determine the superficial and structural changes to the Fe. The surface zeta potential ( $\zeta$ ) of the FeNPs was analyzed with an electrophoretic measurement device (Zetasizer Nano ZS, Malvern instruments).

For uncoated and coated membranes, the surface morphology was examined by SEM (FEI NanoSEM-200) and atomic force microscopy (AFM) (Asylum Research MFP3D-SA). Their hydrophilicity was evaluated by contact angle measurements (OCA, 15 plus). Biofouling tests of FeNPs coated and uncoated (control) PA membranes were carried out in a cross-flow cell (CF042, Sterlitech Corp.) with an effective membrane area of 0.0042 m<sup>2</sup>. Seawater with a high concentration (10<sup>9</sup> CFU ml<sup>-1</sup>) of Gram-positive bacteria (*data do not declared*) obtained from the Sea of Cortez in Mexico was used in order to accelerate biofouling. The test was running for 90 h at temperature of 29 °C.

## III. RESULTS

Figure 1 illustrates X-ray diffraction (XRD) patterns of FeNPs. The Fe exhibits typical diffraction peaks at  $2\theta = 44.5^\circ$ ,  $64.9^\circ$  and  $82.1^\circ$ , with crystallographic parameters representative of cubic Fe (96-411-3942) according to Woodward et al. [5]. The existence of cubic magnetite (Fe<sub>3</sub>O<sub>4</sub>) (96-900-5841) can also be observed, similar to the results reported by Nakagiri et al. [6]. The presence of a peak at



$2\theta = 18.5^\circ$  suggests the existence of residual material from the FeNPs synthesis process.

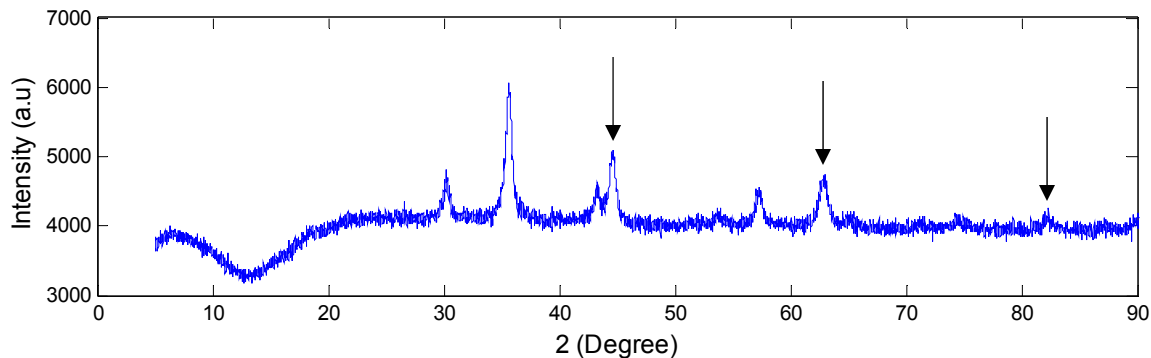


Figure 1. X-ray diffraction peaks associated with FeNPs.

Micrographs obtained by SEM show the diameters of synthesized FeNPs to be around 8–22 nm (Figure 2). The FeNPs show a negatively charged surface ( $\zeta = -7.58$  mV), which can explain their agglomeration tendency.

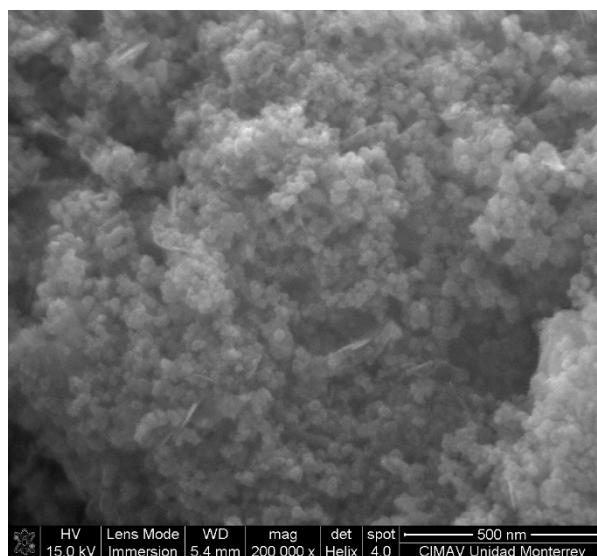


Figure 2. SEM micrographs of FeNPs.

AFM data (Figure 3) show that the uncoated membrane had a smoother surface than the FeNPs coated membrane, probably due to the FeNPs agglomeration tendency.

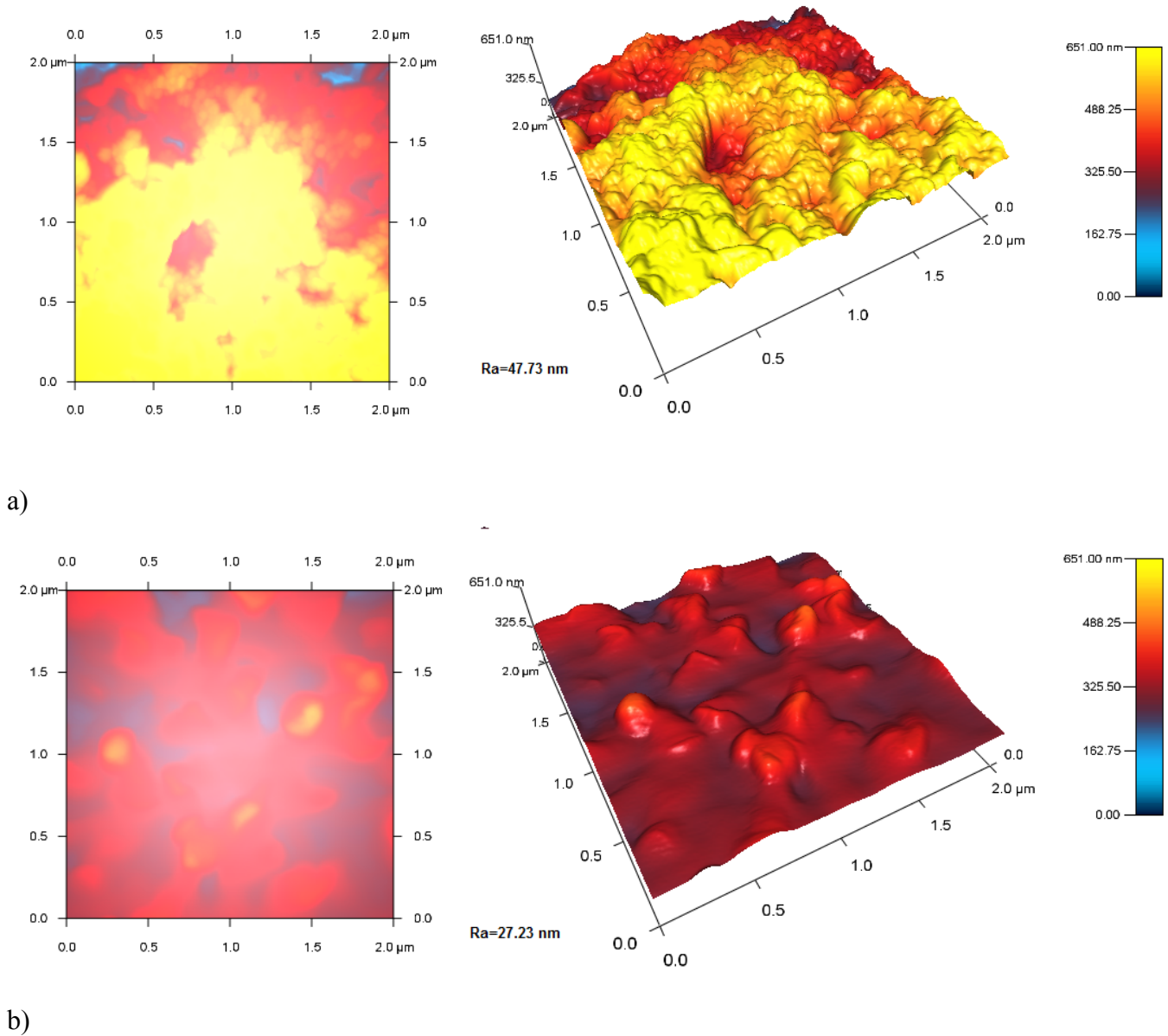


Figure 3. Results from AFM: a) FeNPs Coated; a) uncoated

Membrane permeance was affected significantly by the FeNP coating with respect to the uncoated membrane. This result alludes to the agglomeration tendency of the FeNPs [4]. The reduction in permeance due to the effect of coating was around 32% (Figure 4a), the contact angle results also predict that permeance would be reduced by the FeNP coating, since the mean value for the uncoated membrane was  $50.3^\circ$  and  $65.7^\circ$  was obtained for FeNP coated membranes, showing that FeNPs reduced hydrophilicity. However, the flux for the FeNPs coated membrane was not significantly different to uncoated membrane (figure 4b).

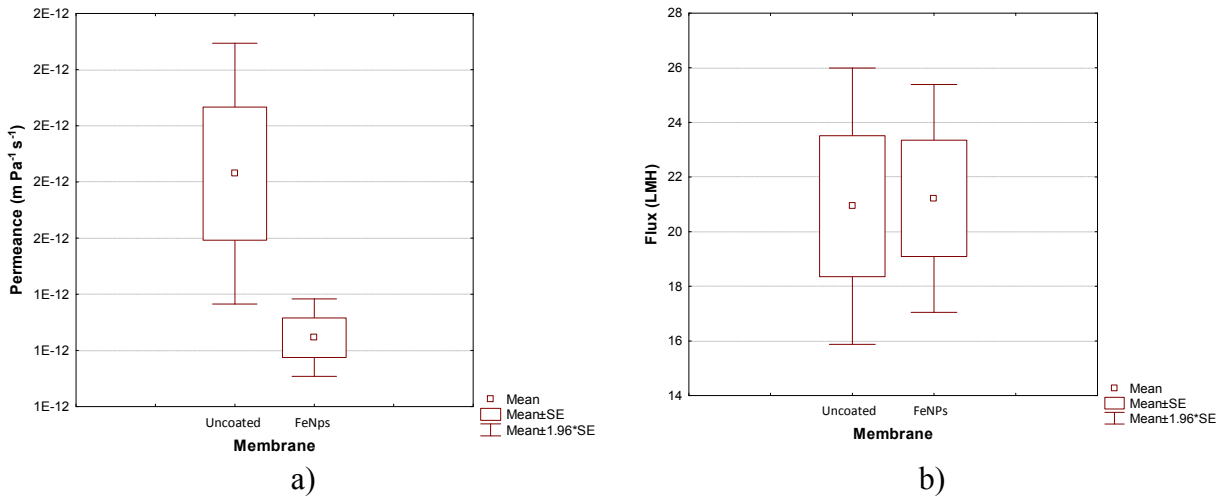


Figure 4. Membranes permeance: a) uncoated; b) FeNPs Coated.

Biofilm cake layer formation on the membrane surface shows a highly significant decrease for the FeNP coated membrane with respect to the uncoated membrane. This result evidences the antimicrobial effect of FeNPs, already demonstrated in the medical field [7]. The reduction on cake thickness due to the coating was of 90% (Figure 5a). This result is in agreement with the reduction in total organic carbon, since it was about 92% lower for the FeNP coated than for the uncoated membrane (Figure 5b).

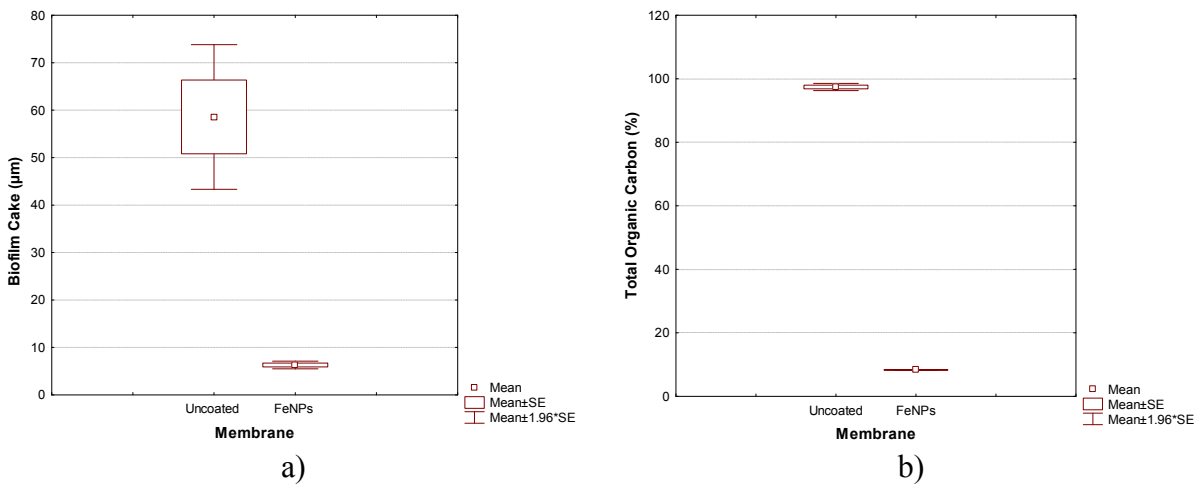


Figure 5. a) Biofilm cake layer thickness in membranes; b) total organic carbon in fouled membranes

The total cell count also showed a highly significant reduction due to the effect of the FeNPs, as it was 67% lower than for the uncoated membrane (Figure 6a). This agrees with the result from the analysis of optical density, since the latter also showed a highly significant reduction attributed to the FeNPs, with a reduction of 42% (Figure 6b).

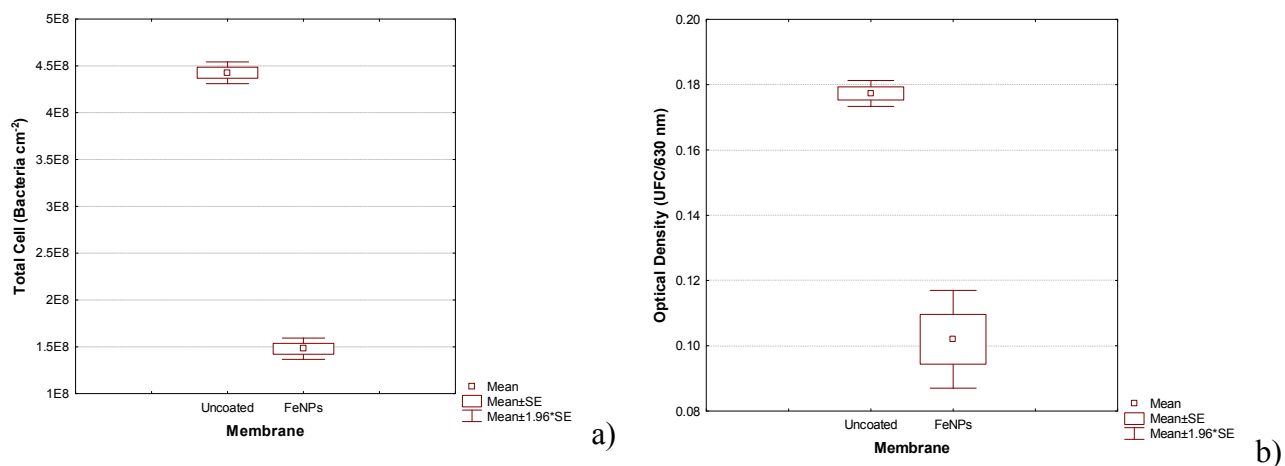


Figure 6. Fouled membranes. a) Total cell count; b) optical density

#### IV. CONCLUSIONS

The results of this study suggest that a FeNP coating has the potential to reduce the biofouling tendency for RO membranes in desalination plants, without significantly increasing membrane costs. However, it was also found that the FeNP coating reduces membrane permeance and decreases its hydrophilicity. This probably by FeNPs can block membrane pores. Nonetheless, FeNP coated RO membranes may offset this by reducing operational costs and problems for desalination processes related to biofouling, which would lead to long term increased operating pressure and more frequent cleaning and membrane replacement. Further studies are recommended to better understand the FeNPs leachate tests in permeate, long-term flux and economic effects of FeNO coatings on RO membranes.

#### V. ACKNOWLEDGEMENT

The authors acknowledge support by the Cátedras CONACYT Program (Project 2338), by the CONACYT Institutional Fund (FOINS) “Proyectos de Desarrollo Científico para Atender Problemas Nacionales” (Project 2015-1221), and by ITSON through the PROFAPI fund (Project 2017-0021).

#### VI. REFERENCES

1. GWI, *The 27th Worldwide Desalting Plant Inventory*, 2015, Global Water Intelligence.
2. Rahaman, M.S., et al., *Control of biofouling on reverse osmosis polyamide membranes modified with biocidal nanoparticles and antifouling polymer brushes*. Journal of Materials Chemistry B, 2014. **2**(12): p. 1724-1732.
3. Dinh, N.X., et al., *Water-dispersible silver nanoparticles-decorated carbon nanomaterials: synthesis and enhanced antibacterial activity*. Applied Physics A, 2015. **119**(1): p. 85-95.
4. Arancibia-Miranda, N., et al., *Nanoscale zero valent supported by Zeolite and Montmorillonite: Template effect of the removal of lead ion from an aqueous solution*. Journal of Hazardous Materials, 2016. **301**: p. 371-380.
5. Woodward, P.M., E. Suard, and P. Karen, *Structural tuning of charge, orbital, and spin ordering in double-cell perovskite series between NdBaFe2O5 and HoBaFe2O5*. Journal of the American Chemical Society, 2003. **125**(29): p. 8889-8899.
6. Nakagiri, N., et al., *Crystal structure of magnetite under pressure*. Physics and Chemistry of Minerals, 1986. **13**(4): p. 238-244.
7. Dinali, R., et al., *Iron oxide nanoparticles in modern microbiology and biotechnology*. Critical reviews in microbiology, 2017: p. 1-15.