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Removal of pollutants by polyamide microfiltration membrane impregnated with photocatalytic nanoparticles

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Abstract

Micropollutants or emerging contaminants (ECs) are organic compounds of anthropogenic origin that are challenging to degrade [1]. Those compounds can be from a wide variety of organic classes, such as pesticides, dyes, or pharmaceutical products [1]. As conventional effluent treatment processes do not treat them, the existence of these ECs in freshwater bodies is already a reality in several developing countries [2]. Therefore, there is a great demand for efficient techniques for their removal. Ultrafiltration and Nanofiltration Membrane Separation Processes (MSP) can remove such pollutants by size exclusion, but they do not have a high permeate flux in addition to sizeable fouling formation [3]. Although the Microfiltration process is inefficient in removing EC, it is a simple and low-cost process that presents a greater permeate flow, allowing the treatment of a greater volume of effluent [4].

To improve the EC removal efficiency of membranes, it is possible to synergistically integrate MSP with the Advanced Oxidative Process (AOP) through the impregnation of photocatalytic nanoparticles, such as zero-valent iron nanoparticles (nZVI) or TiO_2, with the help of a chelating agent, such as polyacrylic acid (PAA), to increase the affinity between the particles and the polymeric surface. AOP is a process capable of degrading emerging contaminants through the release of hydroxyl radicals, but it presents some challenges to be overcome, such as the agglomeration of catalytic nanoparticles, which reduces the contact area with the pollutant and consequently the efficiency of the process [5]. In this way, a hybrid MSP/AOP process can increase the efficiency of both processes because AOP controls the formation of scale on membranes, and MSP prevents the agglomeration of nanoparticles [5].

This work aimed to study the MSP/AOP hybrid process using a polyamide membrane impregnated with nZVI and TiO_2. A flat commercial nylon membrane with a porosity of 0.45 µm was chosen. PAA polymerization in situ on the membrane surface was performed according to the methodology described by Silva et al., [6]. The impregnation of nZVI nanoparticles also followed the methodology described by Silva et al. [6], while that of TiO_2 occurred by ultrasound.

The permeation system used has a cross-flow operation, with continuous recirculation of the permeate and retentate to the source reservoir, in addition to the possibility of removing aliquots of the permeate without stopping the process. The membrane cell had a chamber with an attached UV lamp that allowed radiation to be applied to the membrane surface when activated (Fig 1).

Figure 1: Cross-flow permeation system with recirculation of permeate and retantate.

A model contaminant was used for the micropollutant removal tests: Drimaren Red dye (DR, 5 mg/L). The initial conditions adopted were pH = 4.5, $[DR]_0$ = 5mg/L, P=1 bar and H_2O_2 = 10mM. The permeate was removed at predetermined intervals to follow the process of removing the dye.

When characterizing the polymeric surface by Scanning Electron Microscopy, it is possible to notice a reduction in the size of the pores due to the polymerization of PAA (Fig 2). In addition to this aspect, it is possible to observed that just adding the chelating agent (PAA) already generates an increase in the removal of the dye by 15% (Fig 3).

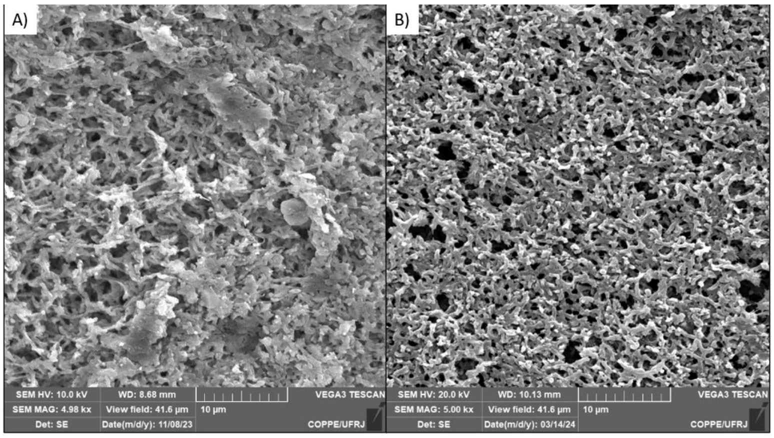

Figure 2: Scanning electron microscopy of (a) Nylon membrane and (b) Nylon/PAA membrane.

The presence of the nanoparticles strongly influenced the dye removal aspect to the point that the membrane with the TiO_2 nanoparticles achieved 85% dye removal in the final time (120 min), and the nZVI was 58% (Fig 3).

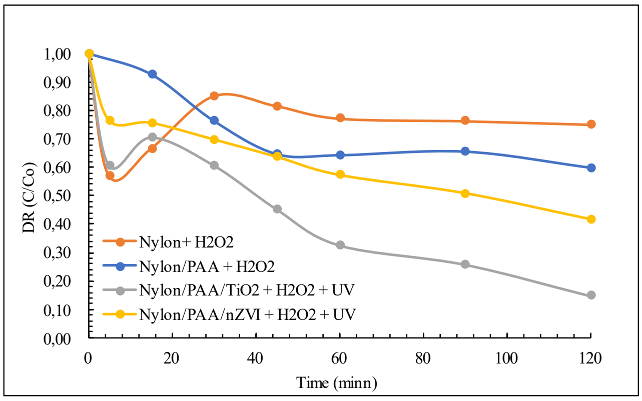

Figure 3: Removal of Drimaren Red Dye in the permeate during 120 min of the experiment using different membranes.

Therefore, the hybrid process of MSP-AOP is a possible form of efficient innovation in the removal of micropollutants because, in preliminary tests, it has already managed to remove up to 85% of the dye.

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