17th International Conference on Inorganic Membranes
New Frontiers for Inorganic Membranes
July 1 - 5, 2024
Oceania Convention Center - Florianópolis - Brasil

Innovative progress in hybrid alumina/dopamine/graphene oxide membrane for wastewater filtration

A. M. J. M. Cardoso[a], V. L. Cardoso[a], and M. H. M. Reis[a*]
[a] Department of Chemical Engineering, Federal University of Uberlândia, Uberlândia, Brazil
[*] miria@ufu.br

Introduction

Graphene oxide (GO) membranes, with a unique structure to provide high flux rate and selectivity, have been deeply investigated for the retention of target compounds [1]. Ultrathin GO membranes are usually deposited on a porous support that should be stable at harsh conditions, require mechanical resistance and present high permeation flux [2]. Ceramic hollow fibers offer a number of advantages in their use, such as the high surface area that enables space saving, greater productivity and cost reduction [3-4]. The proper GO deposition depends on the properties of the support surface and the pre-treatment of the surface of alumina hollow fibers can be an alternative to facilitate the effective deposition of GO membranes on the substrate. Dopamine is suggested to be used as an intermediate layer to promote the GO adhesion on the support surface [5].

Herein, we propose an innovative progress in the use of alumina hollow fibers, pre-treating their surface with dopamine for subsequent deposition of GO membranes and an application of this proposed system in the removal of pharmaceutical contained in a synthetic solution.

Material and methods

Alumina hollow fibers were produced by the phase inversion method followed by sintering at 1400 °C as suggested in the literature [6]. The following ceramic suspension (mass percent) was prepared: 60% of alumina (1.4 µm, 99.94%, surface area 3.1 m^2/g, Almatis), 33.6% of dimetilsulphoxide (DMSO, PA, ACS, Êxodo Científica), 6% of polyethersulfone (PESf, Veradel 3600P, Solvay), and 0.4% of polyvinylpyrrolidone (PVP, Sigma Aldrich). The alumina hollow fibers were immersed in a dopamine (≤100%, Sigma Aldrich) solution for 4 h and dried at room temperature before the GO deposition [7]. A GO suspension at 0.075 mg/mL was deposited on the ceramic fibers by the vacuum-assisted dip-coating method.

Atomic force microscopy (AFM, Shimatzu SPM 9600) was applied to visualize the morphological structure of the GO membrane and to evaluate the average roughness of the membrane outer surface. Membrane morphology was also evaluated using a scanning electron microscope (SEM, Tescan, model VEGA3).

For the application of the produced membranes, synthetic pharmaceutical solutions of rifampicin (≥97%, Sigma-Aldrich), propranolol hydrochloride (≥99%, Sigma-Aldrich) and methylene blue (ARS, Vetec) were filtered under the pressure of 5 bar. The concentrations of the pharmaceutical in aqueous solution before and after filtration were determined using the UV-VIS spectroscopy (Shimadzu, UV-1280).

Results and discussion

The roughness of the uncoated alumina hollow fiber was 122.5 ± 5.6 nm (Fig. 1a) and, after the dopamine pre-treatment, the roughness was reduced to 118.6 ± 10.7 nm (Fig. 1b), representing a slight decrease in the surface irregularity. The alumina hollow fiber pre-treated with dopamine and coated with GO presented a roughness of 42.32 ± 3.50 nm (Fig. 1c).

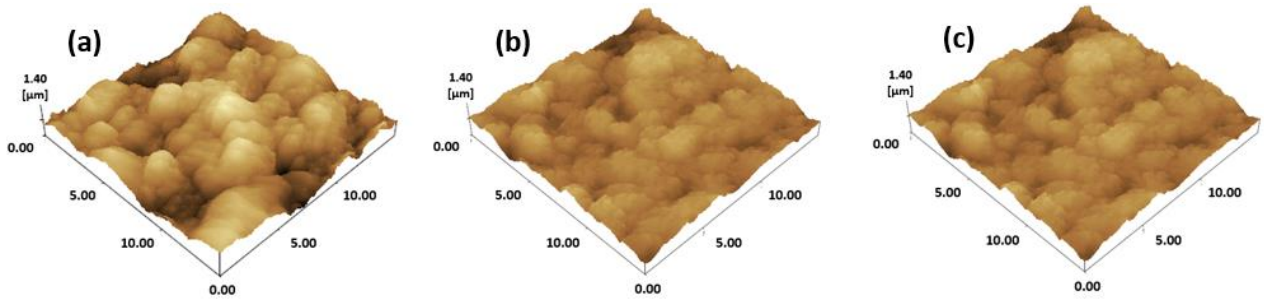

Fig. 1: AFM images of the surface of alumina hollow fibers: (a) uncoated; (b) pre-treated with dopamine; (c) pre-treated with dopamine and coated with GO.

The pristine hollow fiber presented outer and inner diameters of 2.1 ± 0.1 mm and 2.9 ± 0.2 mm, respectively (Fig. 2a). Micro voids were formed throughout the hollow fiber cross section due to the phase inversion process [4]. The porosity of the outer surface of the support was reduced after GO deposition, as shown in Fig. 2b and Fig. 2c. The measured coating thickness of the GO/EDA composite membrane was 0.27 ± 0.02 µm, as exhibited in Fig. 2d.

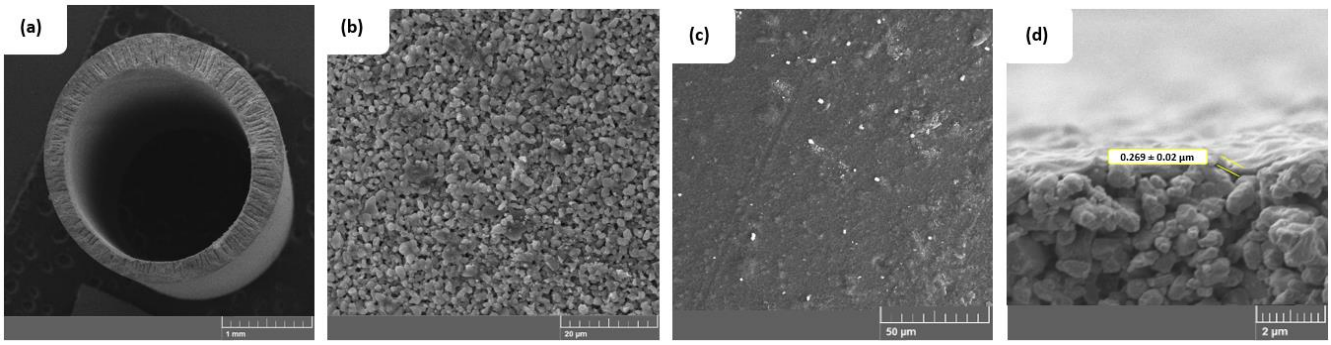

Fig. 2: Cross-section (a,d) and outer surface (b,c) SEM images of the alumina hollow fiber before (a,b) and after (c,d) GO deposition.

Regarding the filtrations of the pharmaceuticals, the hybrid alumina/graphene oxide membrane successfully removed 52% of the rifampicin contained in the synthetic solution, 38% of methylene blue and 34% of propranolol. Hence, the results obtained in this work are promising for the utilization and enhancement of the proposed system here in tertiary processes for treating aqueous effluents containing pharmaceuticals.

Conclusion

Alumina hollow fibers pre-treated with dopamine are suitable substrates for the deposition of GO membranes by the simple vacuum-assisted process. The dopamine layer decreases the roughness of the substrate outer surface and favors the adherence of the GO layer. The morphology of the asymmetric alumina hollow fibers, with micro voids surrounded by a densified porous ceramic structure, enables the membrane mechanical resistance and do not compromise the permeate flux. The hybrid alumina/graphene oxide membranes are suggested to the removal of pharmaceutical compounds from liquid solutions.

References

[1] Austria, H.F.M., J. Widakdo, O. Setiawan, T.M. Subrahmanya, W.S. Hung, C.F. Wang, J.Y. Lai. "Tailoring the specific crosslinking sites of graphene oxide framework nanosheets for controlled nanofiltration of salts and dyes" J. Clean. Prod., 395, Article 136280 (2023).

[2] Kim, S.J., D.W. Kim, K.M. Cho, K.M. Kang, J. Choi, D. Kim, H.T. Jung. "Ultrathin graphene oxide membranes on freestanding carbon nanotube supports for enhanced selective permeation in organic solvents" Sci. Rep., 8, p. 1959 (2018).

[3] Ferreira, E.S.B., R.S.B. Ferreira, C.B.B. Luna, E.M. Araújo, H.L. Lira. "Hollow fiber membranes of several materials and their applications". Res., Soc. and Dev., [S. l.], v. 10, n. 1, p. e55910111206 (2021).

[4] Terra, N.M., G.S. Madrona, F.B. Ferreira, V.L. Cardoso, M.H.M. Reis. "High Performance of asymmetric alumina hollow fiber membranes for the clarification of genipap (Genipa americana L.) fruit extract". Food Bio. Technol., 12, pp. 27-38 (2019).

[5] Lee, H., S.M. Dellatore, W.M. Miller, P.B. Messersmith. "Mussel-inspired surface chemistry for multifunctional coatings". Science, 318(5849), 426-430 (2007).

[6] Ferreira, E.P. "Produção de membranas cerâmicas do tipo fibra oca utilizando pentóxido de nióbio. Msc. dissertation, Universidade Federal de Uberlândia. (2018).

[7] Kong, F.X., Q. Liu, L.Q. Dong, T. Zhang, Y.B. Wei, J.F. Chen, C.M. Guo. "Rejection of pharmaceuticals by graphene oxide membranes: role of crosslinker and rejection mechanism". J. Membr. Sc., 612, 118338 (2020).

in crossflow microfiltration," J. Membr. Sci., 96, 1-58 (1994).