Mixed Matrix Membranes incorporated with Sonication-assisted ZIF-8 Nanofillersfor Industrial Wastewater Treatment

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Abstract: Attention to treat industrial wastewater using mixed matrix membranes (MMMs) has been increased rapidly to obtain high effluent qualities. MMMs containing zeolitic imidazolate framework-8 (ZIF-8) as filler in polydimethoxysilane (PDMS) matrix were synthesized. ZIF-8 was prepared using a modified recipe and characterized by different techniques to evaluate its morphology, thermal stability, surface area, pore volume, and other characteristics. The performance of membranes was evaluated for their applications in industrial dye-stuff wastewater treatment and solvent resistant nanofiltration. The results demonstrated that an increase in the percentage of ZIF-8 loading in PDMS led to a simultaneous increase in the solvent permeability as well as solute rejection from wastewater. The organic dye rejection was achieved more than 87% with MMMs incorporated with 20% loading of nanofillers. Rejection of the MMMs was significantly higher than that of unfilled PDMS membrane because of reduced polymer swelling and size exclusion of the nanofillers. Membrane swelling tests with toluene and isopropanol demonstrated that nanofillers amount has an inverse relation with membrane swelling; implied that nanofillers were in good interaction with polymer and allowed defect-free membranes with higher solute rejections and reduced membrane swelling.

*Keywords:*Industrial wastewater;Dyes removal;Mixed matrix membrane;ZIF-8 nanofillers; Solvent resistant nanofiltration; Membrane swelling

Introduction

Mixed matrix membranes (MMMs) are promising for industrial wastewater treatment because colloidal and suspended organic compounds are removed more efficiently with high permeance performance. Solvent resistant nanofiltration (SRNF) is one of the advancements in the field of MMMs. SRNF membranes are used for the separation of solutes through solvent streams. Small molecules of solutes with molecular weight ranges from 200 to 1000 *Da* are retained by SRNF membranes whereas solvent molecules are passed through them [1]. Recently, metal-organic frameworks (MOFs) gained significant attention as nanofillers in MMMs [2]. MOFs are porous crystalline materials that are composed of metal ions and organic ligands. They have a high surface area, good thermal stability and show strong interaction with the polymer in the matrix [3]. In this study, MOFs were selected as nanofillers material. The incorporation of ZIF-8 synthesized MMMs, as nanofillers, containing zinc ions as coordination centers are linked with 2-methylimidazole, and their performance in synthetic dyestuff industrial wastewater and SRNF applications were evaluated.

Materials and methods

ZIF-8 was synthesized using a modified approach of subsequent stirring and sonication to obtain nano-sized particles. Phase inversion technique was used to prepare P84 porous support layer. A polymer dope solution was prepared by dissolving 15 wt. % of P84 powder in a mixture of N-methyl-2-pyrrolidone (NMP) and tetrahydrofuran (THF) in a composition of 3:1. The dope solution was deposited on non-woven polypropylene support (Novatex 2471) with a casting knife using an automatic film applicator (Porometer, Belgium). Coating solutions with different filler loading (5, 10, 15 and 20 wt.%) were prepared by adding the fillers in a PDMS solution. The filtration

experiments were carried out using a stainless steel dead-end filtration cell (Sterlitech, USA) with 0.00146 m² active membrane area. The feed solution was poured into the cell and pressurized with nitrogen to the desired pressure. Permeate was collected under atmospheric pressure. A teflon stirred the feed solution lined magnetic stirrer at 700 rpm. All the experiments were carried out with a feed solution of synthetic industrial wastewater including Congo red dye and IPA where the selected concentration of solute was 17.5 μ mol/L.

Results and discussion

The results demonstrated that an increase in the percentage of ZIF-8 loading in PDMS led to a simultaneous increase in the solvent permeability as well as the rejection of Congo red dye from solution. The ultra-sonication approach during the synthesis of ZIF-8 particles resulted in particles with smaller sizes and improved dispersion in the polymer matrix. The high pore volume and specific surface area promoted the enhancement of flux through the synthesized MMMs. The high surface area of ZIF-8 and their incorporation in the matrix showed very good adsorption and sieving mechanism, leading to higher retention of dye molecules at higher loadings of filler. The achieved solute rejection was more than 87% with MMMs incorporated with 20% loading of nanofillers. The synthesized membranes showed good stability for a more extended period of time even under dead-end flow. The membranes showed a simultaneous increase in solvent flux and dye retention. The incorporation of filler not only increased the separation but also decreased the swelling of the membrane as tested in IPA and toluene solvents.

References

- 1. Vandezande P, Gevers L E, Vankelecom I F. Solvent resistant nanofiltration: separating on a molecular level. Chemical Society Reviews, 2008, 37(2): 365-405.
- 2. Li H, Eddaoudi M, O'Keeffe M, Yaghi O M. Design and synthesis of an exceptionally stable and highly porous metal-organic framework. Nature, 1999, 402(6759): 276-279.
- 3. Banerjee R, Phan A, Wang B, Knobler C, Furukawa H, O'Keeffe M, Yaghi O M. High-throughput synthesis of zeolitic imidazolate frameworks and application to CO2 capture. Science, 2008, 319(5865): 939-943.