

THE ENHANCEMENT OF GRAPHENE OXIDE MEMBRANES' WATER PURIFICATION PERFORMANCE AND STABILITY VIA COVALENT CROSSLINKING WITH P-PHENYLENEDIAMINE (PPD)

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Introduction

The increasing depletion of freshwater globally necessitates efficient re-use and purification of wastewaters. Among the existing separation membrane materials, graphene oxide (GO) is a promising next generation separation membrane candidate, owing to its tunable physicochemical properties [1]. However, the widening of GO membranes pore gap in aqueous environments is a major limitation [2]. Crosslinking agents can be incorporated to alleviate this problem. This study describes a comparative analysis of uncrosslinked and p-Phenylenediamine (PPD) crosslinked GO membranes' water purification performance. Dip-coating and dip-assisted layer by layer methods were used to fabricate the uncrosslinked and crosslinked membranes respectively.

Consequently, this study aimed at the enhancement of the stability and separation abilities of GO membranes through the introduction of a small, subnanometer-sized covalent crosslinker, p-phenylenediamine (PPD), on a dip-assisted layer-by-layer basis. This should bond adjacent layers of GO plates together, thus stabilizing the structure and fixing the pore gap during membrane operation. Two sets of GO based membranes were fabricated, uncrosslinked and those crosslinked with PPD. Dip coating fabrication was used for the uncrosslinked membranes while the crosslinked membranes were fabricated by the dip-assisted layer by layer assembly method.

Materials and Methods

GO powder was purchased from Graphenea (product code: C28/GOB02/Pw, Gipuzkoa, Spain). The membranes were supported by fibrous 0.2 μm pore sized, 47 mm diameter poly (acrylonitrile) (PAN) filter substrates from Sterlitech Corporation (Washington, USA). The crosslinker, p-phenylenediamine (PPD powder, product code: P6001) and methylene blue (MB, $\text{C}_{16}\text{H}_{18}\text{ClN}_3 \cdot 3\text{H}_2\text{O}$, >99% purity; product code: M9140) were all purchased from Sigma Aldrich (Haverhill, UK). A bath type sonicator (Fisherbrand FB1505, Elmasonic S30H) enhanced the dispersion of the GO water suspensions through a 2 hour sonication and the membranes were fabricated with a rotary dip-coater device (Nadotech Innovations, Navarra, Spain).

Following the PAN substrate pre-treatment, crosslinked and uncrosslinked membranes were fabricated using the schematic in figure 1 where the PPD step was omitted for the crosslinked membranes.

Membrane performance on the other hand was evaluated by the separation of 10 mg/l of methylene blue solution in a homemade lab scale nanofiltration cell.

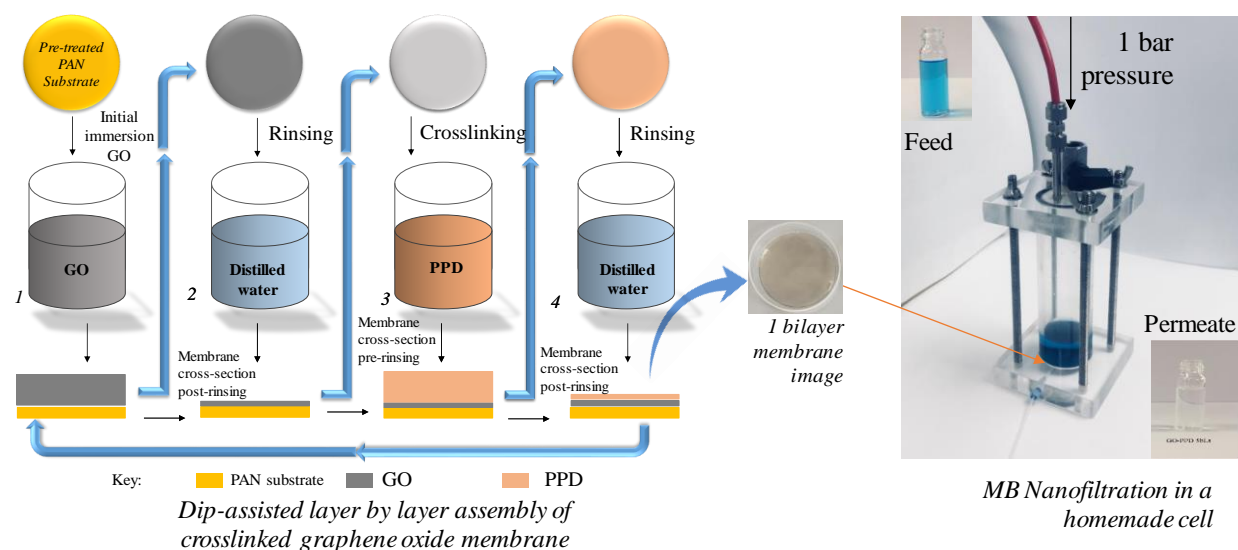


Figure 1. Dip-assisted layer by layer fabrication schematic and the nanofiltration cell

Results and Discussion

The improvement impact of the crosslinker was manifested on the enhancement of the stability and performance of the membranes during nanofiltration tests of aqueous solutions of methylene blue in a homemade nanofiltration cell operated at 1 bar. Higher rejection was achieved for the crosslinked membranes relative to the uncrosslinked ones at similar bi-layers (figure 2).

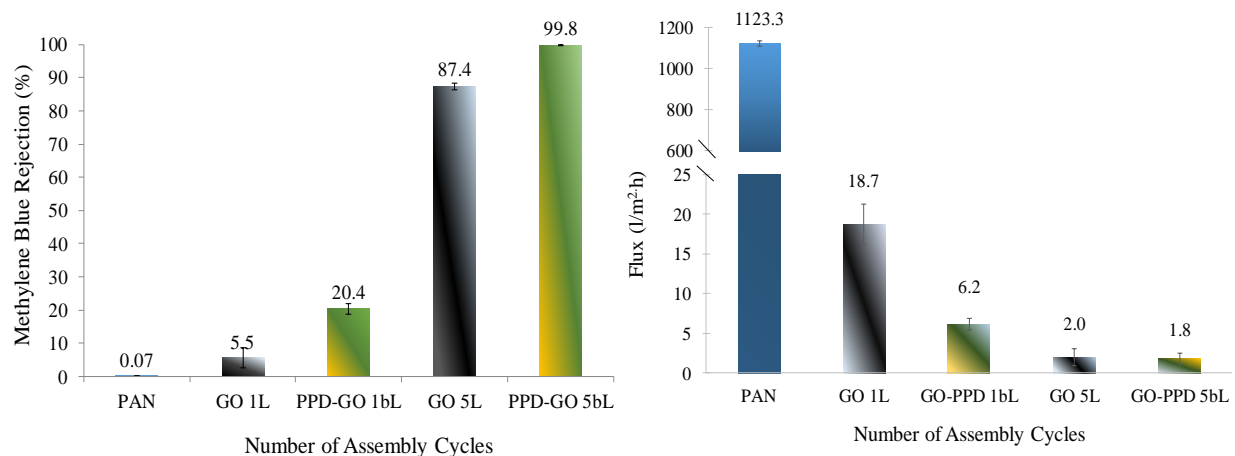


Figure 2. Crosslinked and uncrosslinked membranes rejection and flux

Conclusions

Through this study, the significance of covalent crosslinking was demonstrated, it was not only manifested in nanofiltration performance but also in enhanced stability and membrane intactness over time.

Acknowledgment

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References

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