

ABSTRACT

Water shortage has become a global problem due to the high human consumption, rapid industrialization and low availability of freshwater resources. Extensive efforts have been put forward by researchers to overcome the freshwater crisis by exploring new desalination techniques, and wastewater reuse without damaging the natural freshwater ecosystems. One such pivotal technology for purifying water was the membrane technology. Membrane technology is an emerging and advanced process that can provide a sustainable solution to desalination and wastewater treatment. Although pressure-driven membrane separation processes were used for producing freshwater, its high energy consumption prevent them from further employment. Hence, modification of existing membrane-based separation technology is required to overcome its demerits.

Forward osmosis (FO) is an emerging membrane-based separation technology that can replace the existing pressure-driven membrane-based separation processes without compromising the energy consumption. Compared to thermal processes and pressure-driven membrane separation processes, the FO allows the concentrating solution with higher concentration while maintaining the product quality. Advantages like low fouling propensity and availability of energy-efficient draw solution regeneration technology may enable FO as one of the energy-efficient technology for desalination and wastewater treatment. However, commercial and polymeric FO membranes still lack in some areas like providing high water flux, low reverse solute flux (RSF) and low anti-fouling properties. Hence, there was an apparent requirement for the commercial FO membranes to be modified with nanomaterials that can render anti-fouling properties together with attaining high water and low RSF. Therefore, the practical utility of the FO process in critical applications like

desalination, energy harvesting, wastewater treatment and resource recovery relies on the availability of robust and effective fouling-resistant FO membranes. Therefore, the thesis has addressed certain identified research gaps in the fundamental aspects of the existing membrane.

Based on the prior knowledge of the FO membranes and its working principle, this thesis focuses on the development of existing membranes with the incorporation of different compositions of two-dimensional (2D) nanomaterials as active layer for enhancing the performance in various applications in the FO process. The long-term stability of the fabricated membranes in ideal as well as real conditions was investigated in this thesis.

To enhance the membrane quality and performance in FO process, fabrication of FO membrane with reduced graphene oxide (rGO) nanosheets doped with polystyrene sulfonic acid (PSS) was done. The practical application of the FO process depends on high water permeability and low RSF. Therefore, we have reported the use of rGO and PSS to enhance the water permeation properties and selectivity of the as-prepared FO membranes. After the fabrication process, the membrane was tested in ideal conditions with 1M NaCl and DI water as draw and feed solutions, respectively. Subsequent addition of PSS layer onto rGO laminates improved the hydrophilicity and surface charge of the membrane rendering excellent water flux ($34 \text{ L m}^{-2} \text{ h}^{-1}$) and lower specific reverse solute flux (SRSF) (0.18 g/L), thus enhancing the FO performance for longer duration also.

Once the fabricated membrane showed promising result in ideal conditions, other fabricated membranes were tested in real conditions to validate the feasibility of the membranes to sustain in harsh conditions. Therefore, a novel FO membrane consisting of sericin and reduced graphene oxide nanosheets was fabricated for antibiotic recovery application using

membrane crystallization forward osmosis process. The use of hydrophilic natural protein named sericin was used to enhance the FO performance and resolve the trade-off between permeability and selectivity of FO membrane also improving the membrane stability during the antibiotic's recovery process. The sericin doped rGO membrane achieved an outstanding water flux of $54 \text{ L m}^{-2} \text{ h}^{-1}$ while maintaining the SRSF of 0.12 g/L as compared to commercial FO membranes ($0.5\text{-}6 \text{ g/L}$). The as-prepared membrane also showed higher stability FO performance for 30 hrs of continuous operation without compromising the water flux and RSF, thus confirming the anti-fouling properties of sericin-doped rGO membranes. Moreover, the application with batch FO process yielded continuous crystalline rifaximin antibiotics without consuming higher energy.

After analysing the performance of fabricated FO membranes in real conditions, the balance between the FO process and simultaneous energy harvesting using lamellar bilayer FO membrane was investigated to resolve water-energy nexus issues. Here, we have fabricated a novel amorphous silicon oxide crosslinked vanadium pentoxide (VO) and rGO membrane using a vacuum assisted filtration process. The ion-selective membrane showed superior nanofluidic properties in salinity gradient energy conversion technology with high powder output of 4.72 W.m^{-2} . Finally, the fabricated membrane was used for simultaneous energy harvesting and FO process and it was observed that with 1M KCl as draw solution and DI water as feed solution, the membrane achieved output voltage and current of 300 mV and $35 \mu\text{A}$, respectively, at lowest flow rate of 0.5 L/h . With the pumping power output of 0.1 W.m^{-2} at lowest flow rate, the extracted energy from FO process was able to save an overall 26% of energy. Hence, the ease of coupling FO and concentration gradient-driven energy

harvesting processes in novel platform can resolve highly entangled issues in water-energy nexus.

Overall, the results discussed in the given thesis provides a better insight into the development of forward osmosis membrane and its various applications.

Keywords: 2D nanomaterials, Forward osmosis, Membranes, Crystallization, Nanofluidic



