Fouling Subsequent Cleaning Performance of Spiral Wound Reverse Osmosis Membrane Module

H Rathore, C. Das, P Kushwaha, V Singh

Abstract

Fouling and subsequent cleaning study of spiral wound reverse osmosis (RO) membrane modules were done. The membrane modules consisted of polyamide (PA) with cross sectional area of 1 m². RO membrane fouled by polysaccharides equivalent to sodium alginate (SA), with different doses of SA (200, 300, 500 ppm) was and calcium ions (0.5 to 2 mM) at various operating conditions. Initial flux was the 1.132×10⁻⁵ m³/m² at 1379 kPa and it decline to 1.02×10⁻⁵ m³/m².s after 5 h. Fouled membrane modules were cleaned by using different chemical agents, such as, anionic surfactant, e.g., sodium dodecyl sulphate (SDS) and metal chelating agent e.g., ethylenediaminetetraacetic acid (EDTA). Flow rate of 9 lpm and transmembrane pressure drop of 69 kPa were used to fouled membrane modules. The cleaning performance of membrane module in terms of flux recovery with variation of cleaning agent dose was studied. It was observed that with increase of operating time and cleaning agent dose, cleaning efficiency of membrane module increased. The optimum doses of SDS and EDTA as cleaning agents were 8 and 1 mM, respectively. It was noted that EDTA had a better cleaning efficiency than SDS.

Keywords: Reverse Osmosis, Membrane Fouling, EDTA, SDS, Polyamide.

1. Introduction

Spiral wound membrane module such as reverse osmosis (RO) membrane processes are becoming increasingly popular as a result of the impressive improvements of membrane properties and the corresponding reduction in treatment cost, the significant stress to meet the ever increasing water demand, and the increasingly stringent water quality regulations. Reverse osmosis is the key technology for desalination and wastewater reclamation, In membrane filtration process; spiral wound membrane module fouling depends on the foulant available in the feed stream. Wastewater filtration process, wastewater contains three type of membrane fouling categories, microbial (bacteria, viruses, etc.), organic (NOM) and inorganic (minerals) contents [1]. Depending on the physicochemical properties of the membrane, composition of feed solution and process conditions, membrane loses its performance with time. Fouling not only decreases the flux but also changes the rejection of solutes [2]. Fouling layer are general terms for deposits on or into the membrane that unfavourably affect filtration.

In this study Spiral wound membrane module fouled by Nature organic matter (NOM) such as sodium alginate to model of polysaccharides.

Alginate is a well-known polysaccharide that can be mainly obtained from seaweeds. In the presence of some cations such as calcium alginate can form a three dimensional network which contains high amount of water within its structure. One of the most ubiquitous components of extracellular polymeric substances in the secondary wastewater effluent. The presence of calcium ions, alginate makes a highly compacted gel network of fouling layer [5]. In membrane filtration, the presence of calcium as one with alginate results in different outcomes in terms of permeate flux decline.

Membrane cleaning is more direct and quick to restore the permeability of spiral wound membrane module. The cleaning is usually performed in four forms: physical, chemical, biological and enzymatic. Membrane chemical cleaning occurs through chemical reactions between cleaning chemicals and membranes or organic foulant to remove the fouling layer partially or completely [3]. In general, chemical cleaning can be broadly classified based on cleaning agents into six categories: acids (such as citric acid, HCl), alkaline (such as NaOH), Alkaline solutions remove organic foulants on membrane by hydrolysis and solubilisation of
the fouling layer. Alkaline solution also increases the solution pH and there for increase the negative charges and solubility of the organic foulant \[6\]. Chelating agents (such as EDTA, polyacrylates), metal chelating agents remove divalent cations from the complexed organic molecules and weaken the structure of the fouling layer matrix.

2. Experimental

2.1 Materials

Sodium alginate (MW 75,000 Da), cleaning agents, namely, sodium dodecyl sulphate (SDS) and ethylenediaminetetraacetic acid (EDTA) were purchased from Loba Chemie Pvt. Ltd., Mumbai India. All the analytical grade chemicals were used without further treatment. Calcium carbonate was used as divalent calcium source.

2.2 Methods

2.2.1Experimental set-up

The schematic flow diagram of the Perma Pilot Plant (a spiral wound module for research purpose) was shown in Fig. 2.1. The experimental setup was supplied by M/s Permionics Membranes Pvt. Ltd., Gujarat, India. Spiral wound membrane module was connected with a feed tank (20 L). A cooling tank (20 L) was provided to control the temperature of retentate. Two rotameter were connected with retentate and permeate line to measure flow rates of retentate and permeate, respectively. To manipulate feed flow rate and pressure in membrane module, a part of feed was recycled through a bypass line. High pressure (up to 4136 kPa) triplex plunger pump was attached with the pilot plant to circulate the feed in RO membrane module. Spiral wound RO polyamide membrane module was procured from M/s Permionics Membranes Pvt. Ltd., Gujarat, India. The RO hydraulic membrane permeability (Lp) was measured as \(3.24 \times 10^{-11} \text{ m} \cdot \text{Pa} \cdot \text{s} \). Polyamide RO membrane module with effective membrane cross sectional area of 1 m\(^2\), length of 510 mm and diameter of 73 mm was housed in stainless steel (SS 316) cylindrical shell. Flow rate (up to 10 lpm) of the feed was controlled with the help of rotameter and control valve. One control valve was attached at the outlet of the retentate line to maintain the desired pressure inside the membrane module. To measure the electrical conductivity of permeate; one conductivity meter was attached with the module. Temperature had influence in the permeate flux. Therefore, a cooling tank was provided to maintain the temperature of feed tank. Before experiment, RO membrane module was compacted with deionised water at 100L for 1 hr.

2.2.2 Fouling operations

Organic fouling tests conducted using the small lab scale RO membrane modules. RO membrane module fouled by sodium alginate, solution were preparer 200, 300, 500 ppm by using stirred at 200 rpm for 30 min duration, magnesium and calcium ions different concentration added in feed and pH adjusted by NaOH, Hcl. Prepared feed was put in feed tank. RO membrane experiment was conducted 1379 kPa, and flowrate 9 lpm. Constant feed flow rate was maintained by adjusting the control valves. Permeate samples were collected at a definite time interval for analysis of purpose and the remaining amount of permeates was recycled to feed tank. Total time to run these experiments was 5 hr and 90 min. To maintain the transmembrane pressures (TMP) inside the membrane module, feed and retentate outlet control valves were adjusted. The temperature of the feed tank was measured periodically and maintained at 30± 1 °C with the help of cooling tank by circulating coolant.

2.2.3 Cleaning operations

Membrane cleaning is carried out to recover the permeate flux. The cleaning process started with the circulation of selected cleaning solution for 30 and 60 min at 69kPa and cross flow velocity of 9 lpm. DI water, SDS and EDTA used cleaning of fouled membrane module. Cleaning agent used with different physical and chemical operating conditions. Make a Cleaning solution and adjusted pH with used NaOH and Hcl and fixed feed temperature. Feed put in tank after started set up and measured the flux. Flux is recovered at cleaning process. The restored flux using organic-free test solution at identical operating conditions used in the fouling experiment determined the efficiency of the membrane cleaning. All the operations were carried out at a temperature of 30±1 °C.

3. Results and discussion

3.1 Sodium alginate Rejection

Foulant (sodium alginate), different concentrations (200 to 500 ppm) at TMP drops 1379 kPa were used to foul the spiral wound RO membrane module. With increased of sodium alginate concentration from 200 to 500 ppm, rejection will increase from 77 to 87.2%. Rejection percentage with different concentration shown in table 3.1 the result is at high concentration rejection rate high. Because at high concentration sodium alginate molecules will pass less from membrane, maximum alginate particles deposited on membrane surface and make a gel layer. These deposited particles created some problems in filtration process; membrane fouled on after some time.

<table>
<thead>
<tr>
<th>Feed (ppm)</th>
<th>Permeate (ppm)</th>
<th>Rejection %</th>
</tr>
</thead>
<tbody>
<tr>
<td>200</td>
<td>46</td>
<td>77</td>
</tr>
<tr>
<td>300</td>
<td>55</td>
<td>81</td>
</tr>
<tr>
<td>500</td>
<td>64</td>
<td>87.2</td>
</tr>
</tbody>
</table>

3.1.1 RO membrane module fouled by sodium alginate

The normalized flux \((J/J_0)\) profiles for RO Polyamide membrane fouled by individual organic foulant sodium
alginate (200 ppm). Shown in Fig. 3.1 the permeate flux during fouling runs decreased dramatically since alginate fouling of the RO membrane. Initial permeate flux $1.1314 \times 10^{-5}$ m$^3$/m$^2$.s after 5 hr flux $1.0204 \times 10^{-5}$ m$^3$/m$^2$.s at 1379 kPa, is steady state because alginate molecules deposited of membrane surface. Alginate molecules in a highly cooperative manner shaped gel network. In fact, visual inspection of the membrane surface at the end of the fouling runs confirmed the presence of a thick alginate gel layer.

![Sodium alginate (200 ppm) pH 8.5, Flowrate 9 lpm TMP: 1379 Kpa Temp. 30±2ºC $J_0 = 1.1314 \times 10^{-5}$ (m$^3$/m$^2$.s)](image)

**Fig 3.1:** Flux behaviour during the organic fouling of RO membrane

### 3.1.2 Effect of sodium alginate concentration on RO fouling
Alginate fouling was also investigated with different sodium alginate concentrations (200, 300, and 500 ppm) in the absence of calcium concentration. Changes in permeate flux due to alginate fouling at three different sodium alginate concentrations over about 5 hr of operation. Initial fouling rates were observed to correspond with the sodium alginate concentration, i.e., increasing sodium alginate concentration resulting in higher fouling rate shown in Fig. 3.2. This observation contrasted greatly to the flux curves shown in Fig. 3.2 which showed similar initial fouling rate. It seemed that the alginate concentration, rather than the calcium concentration, affected the initial fouling rate. Near the end of the experimental runs, the stabilized normalized fluxes at three different sodium alginate concentrations Fig. 3.2 were less different as compared to normalized fluxes for varying calcium concentration but with constant sodium alginate concentration Fig. 3.2. Hence calcium concentration seemed to have greater influence on the long term flux than the sodium alginate concentration.

![Flux Decline with different concentration of sodium alginate.](image)

**Fig 3.2:** Flux Decline with different concentration of sodium alginate.

### 3.1.3 Effect of cleaning dose
To optimize the dose of SDS (pH = 11) and EDTA (pH = 11) in terms of original flux recovery SDS concentration was varied from 2 to 10 mM whereas, EDTA concentration was varied from 0.5 to 2 mM. Pure water flux at 69 kPa was $16.67 \times 10^{-6}$ m$^3$/m$^2$.s and after membrane fouling with calcium ions flux was reduced to $5.26 \times 10^{-6}$ m$^3$/m$^2$.s. The variation of permeate flux values with SDS concentration was shown...
Fig. 3.3 (a). It was observed from the figure that with the increase of SDS dose, permeate flux was found to increase. When SDS concentration was increased permeate flux recovery was increased. Flux recovery was observed when SDS concentration was further increased to 8 mM. Beyond this concentration, recovery of permeate flux was marginal. Hence 8 mM SDS concentration was taken as optimum surfactant dose. With EDTA, the recovery of permeate flux from 5.26×10⁻⁶ m³/m²·s to 14.28×10⁻⁶ m³/m²·s was increased when dose was 1 mM. The results were shown Fig. 3.3 (b). Like SDS dose, further increase in permeate flux was observed with increase in EDTA dose. When EDTA dose was 1 mM at pH 11 the recovery of flux was 92%. Further increase in EDTA dose to 2 mM could not increase the permeate flux recovery. So 1 mM EDTA was considered as the optimum dose.

4. Conclusion

- In this present work organic pollutants (Sodium alginate) were treated with the help of RO membrane modules. The maximum removal efficiency of alginate is 87% (RO module) after it reaches its steady state value after 5 h of operation.
- For cleaning of Fouled spiral bound membrane modules, we took different doses of SDS and EDTA and we found that 8 mM SDS and 1mM EDTA gives maximum cleaning efficiency.
- EDTA was shown to be a better cleaning agent as compared to SDS for alginate fouling in the presence of calcium because of its ability to form soluble complexes with calcium from the sodium alginate calcium gel layer.

5. Reference