

A Short Review on Reverse Osmosis Membranes: Fouling and Control

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Abstract

Reverse Osmosis (RO) is the process of separating dissolved salts from water with the help of semipermeable membranes. Membrane based solution are now widely accepted technology to combat safe drinking water shortage. Reverse osmosis has increasing market shares due to reduced cost and improvements in the process. This paper reviews the major issue of fouling that is faced during operation of RO and ways to regulate them. Fouling is categorized into many classes and the control is discussed respectively. It also discusses basics of RO, modular arrangements for RO membranes as well as multiple options for pretreatment which is a mandatory requirement of the process. Final discussion is the ways to consider while disposing of brine.

Keywords: Reverse Osmosis; Modules; Fouling; Pretreatment

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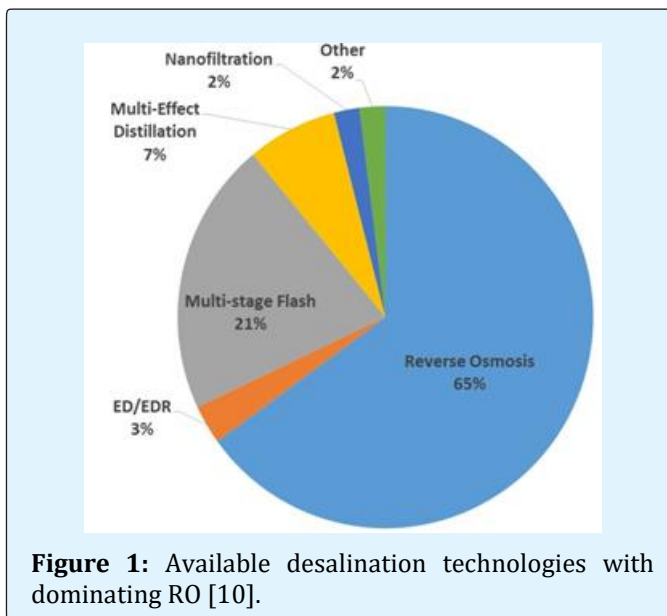
Introduction

It is believed that 70 % of the earth is covered with water. This makes a common person assumes that water is abundantly available on earth crust so no need to pay specific attention to the control on utility and reducing wastage of the water. However, it must be kept in mind that all water is not available for utilization. According to USGS, About 97% of the water is available in oceans and seas with high total dissolved solids (> 30,000 mg/L) and only 3% is fresh water. Out of this 3%, only 0.8% is readily available fresh water resource due to location, physical state and natural distribution of hydrologic waters. The only water

available for human consumption is about 0.77% which is approximately 10.7M Km³ [1,2].

Water requirement and consumption rates are increasing with each passing days due to enormous increase in world population especially in developing countries, developing growth and economies, changing climatic conditions, and lifestyles. Global water demand is increasing with an increment of 1% per year and is expected to grow dramatically in the future due to above mentioned reasons. Industrial and domestic water requirements are increasing at faster rates, putting more pressures on existing fresh water resources [3]. Relying on water conservation and storage strategies alone cannot serve the purpose. This is resulting in search for new methods to obtain fresh water resources to combat with the emerging water scarcity issues [4]. Desalination has been used for centuries to extract fresh drinking water out of saline sea water with the help of thermal distillation processes. Desalination removes dissolved salts from saline water turning it into fresh water. According to USEPA, drinking water should have < 500 mg/L total dissolved solids. However these criteria varies from region to region and country to country [5].

World is now looking toward seawater as a source of fresh water. Membrane based Reverse Osmosis (RO) has dominated over all available technologies since the past 60 years. Membrane based processes are not only used for sea water desalination but for treatment of brackish water and wastewater treatment too. The overall market has outraged other desalination technologies by more than 60% [6-9].



The rapid shift toward RO from other thermal based processes for desalination is due to technological advancements happening in membrane technology and its application. Starting from the development of asymmetric Leob Sourirajan membranes, then, thin film composite membranes, graphene, Carbon Nanotubes (CNT) and other novel material based membranes have played their role toward advancements. Moreover improvements in membranes packing, modules and operation has also resulted in reduced energy consumption. The overall trend of shift from thermal based technologies to RO can be seen in Figure 1. The overall benefits don't mean that they are no challenges left. To date there are numerous bottlenecks that are still needed to be improved. These include fouling of membranes, pretreatment requirements, membrane cleaning, cost of membranes and membrane based processes [8,9,11]. This paper covers basics of RO process, review of recent advances in modular arrangements, membrane fouling, controlling strategies, pretreatment options and brine disposal. At the end, future directions to be look into are also discussed.

RO Basics, Process etc

Reverse osmosis (RO) is the process of separating dissolved salts from water with the help of semi permeable membrane that allows water to pass through but reject salt passage. Saline water at high pressure is fed to the membranes. This pressurized water is allowed to pass through membranes and salt free permeate is gathered across the low pressure side. The schematic representation of reverse osmosis process is shown in Figure 2. This phenomenon is century's old but gained attention in twentieth century with the discovery of Leob Sourirajan membranes. These membranes had more than 98% rejection with ten times more flux than previous membranes. These properties made them commercially viable [11]. The principle of reverse osmosis is based on difference between water flux and salts flux. The equations for calculating water flux are:

$$J_w = A (\Delta P - \Delta \pi)$$

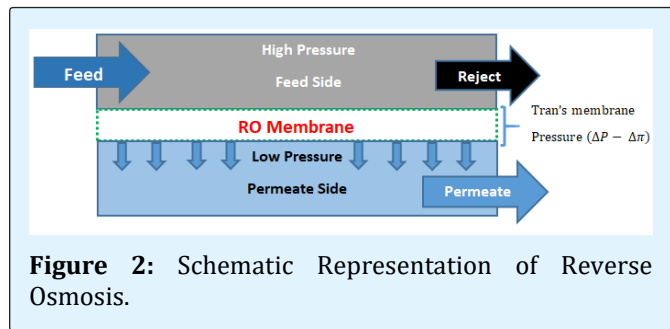
Where A = permeability coefficient, ΔP is transmembrane pressure and $\Delta \pi$ is the osmotic pressure of feed water. The equation for calculating solute flux is as follows:

$$J_s = B (C_f - C_p)$$

Where B = Salt permeability coefficient, C_f is feed concentration and C_p is permeate concentration.

These equations demonstrate that as applied pressure is increased, water flux increases and so does the selectivity of membrane. Selectivity is measured in terms of salt rejection. Equation for rejection is:

$$R = \left(1 - \frac{C_p}{C_f} \right) \times 100\%$$

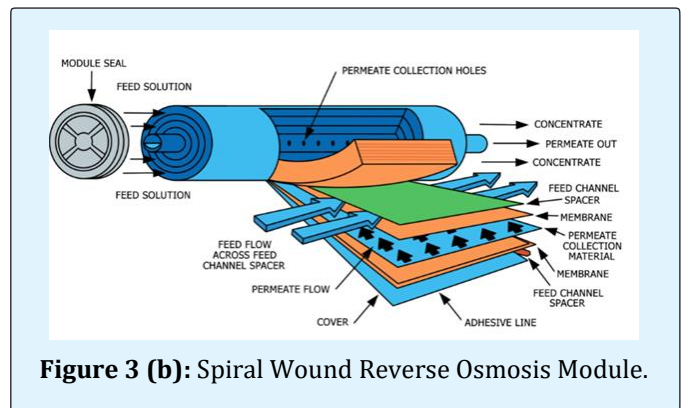
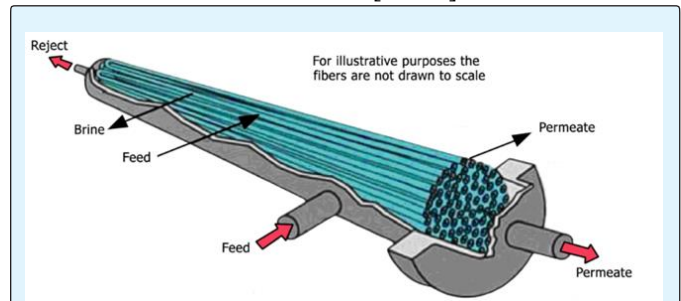


Membrane Modules

For a functional setup, membranes need to be placed in a housing that provides support to the membrane and effective fluid management. The module selection determines the area of membranes covered, cost of membranes, ease of replacement and cleaning, trans membrane pressure and resistance provided against fouling. Different modules can be formed from two main types of membranes i.e. flat and tubular. Based on inner diameter tubular can be divided into fine hollow fiber, capillary and tubular modules. Spiral wound and plate and frame modules can be made using flat sheet membranes [8].

For RO, spiral wound and hollow fiber modules have been used extensively. The hollow fiber modules were initially made in Dow chemicals in 1966 but further used by many like DuPont, Dow and Monsanto. They have diameter of about 50-200 micrometer. They have highest packing density ($500-5000 \text{ m}^2/\text{m}^3$) as compared to all other modules. Thousands of bundles are packed together to make a module. This module is then placed inside a pressure vessel. For RO, Feed enters from the shell side and permeate enters in the bore side. (Figure 3(a)). The total flux produced from these modules is approximately ten times of spiral wound membranes. Although they have least manufacturing cost but due to the closely packed nature, the effective hydrodynamic conditions are not met. These results in high concentration polarization, increased fouling, high permeate side pressure drop, reducing permeate flux and increasing cleaning cost.

There is now only one commercial user of hollow fiber membrane for reverse osmosis [12-14].



The second and widely used universal option for module is spiral wound membrane shown in figure 03(b). Flat sheets of membranes with spacers are rolled around a central perforated pipe. The feed passes outside the membrane tangentially and permeate moves axially until reaches to the central collected pipe. To achieve high recovery of permeate multiple elements of spiral wound are placed in a single pressure vessel. Traditionally 8 in diameter spiral wound membranes have been used. Recently 16 in diameter spiral wound membranes have been tested. They have three times the membrane area thus requiring less elements, pressure vessels, pipes and joints etc. but providing the similar productivity [12].

Membrane Fouling and Control

Membrane fouling is the inevitable issues in the membranes. After commercialization of membranes, fouling is the domain in which maximum research work is carried out. Fouling is basically the deposition of material (salts and/or solids) on the external of internal surface of the membranes. This deposition reduces the efficiency of transmembrane pressure, thereby reducing permeate flux [11,13]. Unless water and ions permeation through RO membranes are not well understood, fouling cannot be

studied. Water passes through these membranes via random Brownian movement, flush and jump diffusion [15]. Operating pressure temperature and membrane surface properties also varies fouling propensity [16]. Fouling can be broadly divided into two categories namely surface and internal fouling. For MF and UF, pore flow model is being followed so clogging also occurs within the pores due to depth filtration. In NF and RO, filtration is done by surface filtration. Permeates

transports using solution diffusion model, so fouling also occurs on the external surface of the membrane [8,11]. External fouling can be due to deposition of minerals, cake layer and biofilm development on the surface of the membranes. Fouling is mainly caused due to organic, inorganic, colloidal and bio foulants [9,17-20]. Figure 4 represents these fouling on surface of membranes individually [19].

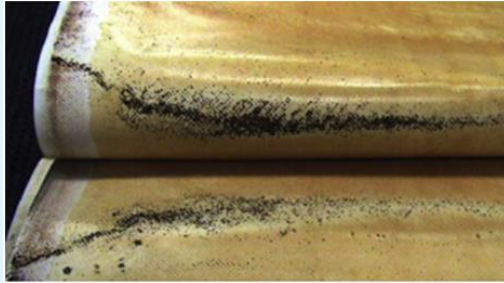


Figure 4 (a): Accumulation of Organics on Membrane Surface

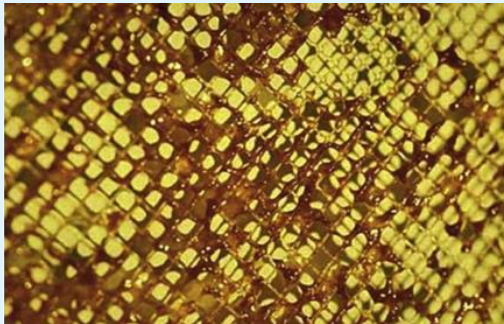


Figure 4 (b): Accumulation of Biofilm on Brine Spacer.



Figure 4 (c): Accumulation of Scale on Brine Spacer.

Biofouling

Microbial fouling is due to attachment of microorganisms on the membrane's surface. After attachment, they start to grow, release Extracellular Polymeric Substances (EPS) during their metabolism, traps other microorganisms and the film growth. This layer consists of dead and alive microorganisms [21, 22].

Biofouling is more complex phenomenon than all other fouling types. Moreover it is challenging to exterminate using pretreatment unless all the present microorganism was killed [9]. The stage for biofilm development is shown in Figure 5.

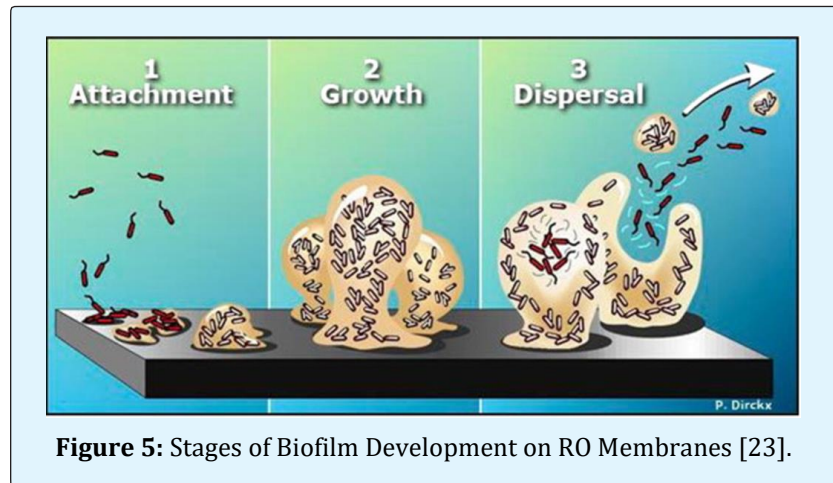


Figure 5: Stages of Biofilm Development on RO Membranes [23].

Biofilm development can be controlled by pretreating the feed solution with commonly used disinfectants. These include chlorine, ultraviolet radiations, ozone, monochloramines etc. However direct dosing of chlorine results in subsequent damage to the membrane [9,22]. Despite the facts that lots of research is being done in the biofouling region, yet this is the most challenging issue in terms of permeate flux reduction. After extensive pretreatment, there is no surety that no biodegradable nutrients are extracted out of feed water. Hence a small amount of microbes and nutrients can aid in developing and growth of biofilm.

Recently many developments have been made to reduce biofouling. These include use of quorum quenching bacteria on reducing the communication between bacteria and hence less biofilm development. Their practical application is not yet done on commercial scales [24,25]. Chemical enhanced backwashing (CEB) and Chemical cleaning also reduces the biofilm developments and retrieves the flux on initial level but at the cost of membrane's life [22].

Organic Fouling

Organic fouling is caused by organic matter present in the feed water. The organic matter adsorbs on the surface of the membrane and forms irreversible fouling.

Monolayer adsorption occurs on the surface of the membranes following Langmuir adsorption isotherm [26]. Organic fouling can be divided into three types Natural Organic Matter (NOM), Algal Organic Matter (AOM) and Effluent Organic Matter (EfOM). AOM = Extracellular macromolecules + Intracellular macromolecules. FfOM = NOM + Soluble Microbial Products (SMPs)

NOM consists of complex macromolecules after degradation of plant and animal biomass. This is present in ground water, surface water and sea water as well. Mainly it is composed of humic acid, which serves as complexation macromolecule for inorganics [18,27]. Fouling due to NOM is affected by feed water chemistry, its temperature, pH and Ionic strength (Figure 6).

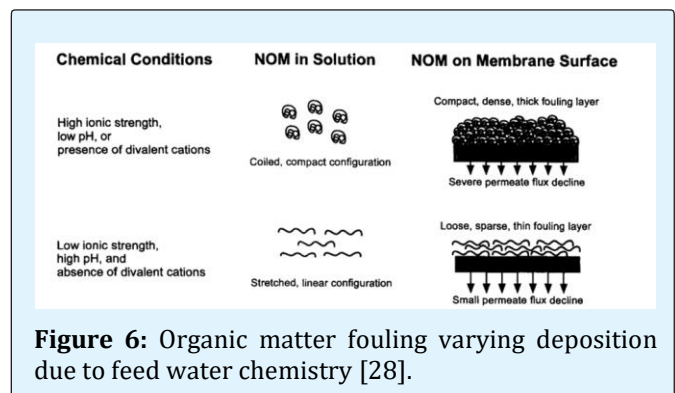


Figure 6: Organic matter fouling varying deposition due to feed water chemistry [28].

Organic fouling can be controlled by variety of options including pretreatment and cleaning of the reverse osmosis membranes. Coagulation/flocculation reduces the adverse charge on the surface of the molecule resulting in fast agglomeration of the molecules. The agglomerates then convert these smaller flocs to visible range for further removal by sedimentation/Micro Filtration (MF)/ Ultra Filtration (UF). Coagulation can be chemical or electrical. These result in reduced cake formation and its compressibility. Granular media filtration using activated carbon, sand, coal etc. also serves the purpose. Adsorption of organic matter on the surface of media plays the primary role to control the exit of organic matter. However, reliable feed quality can be achieved using MF/UF as pretreatment technology to control organic matter [9]. Direct hydraulic backwashing of RO membranes is not in practice due to their nonporous structure. So forward flushing is in practice. Recently osmotic back washing is suggested to remove organic fouling on the surface of membranes. Osmotic backwashing with 96g/L of NaCl resulted in 100% flux recovery. Hence this method proved to be a replacement for chemical cleaning [29].

Inorganic Fouling

Precipitation of the salts over the surface of the membrane cause scaling/inorganic fouling. Inorganic

foulants consists of calcium carbonate, calcium sulfate, calcium phosphate, barium silicate, aluminum silicates and silica. When feed water comes in contact with membrane, water passes through the membrane, leaving salts behind. The concentration of the salts keeps on increasing at the feed side near the membrane surface as compared to the bulk feed solution. When the salts concentration increases than the solubility product, salts starts to precipitate on the surface of the membrane and scaling forms. This scale deposition along with cake layer does not allow water to get in contact with the membrane, thereby reducing flux, membrane degradation, loss of production and elevated operating costs [9,18,30]. Scaling potential of various feed waters can be determined with the help of software's designed by anti-scalant providers including Nalco, Avista, Genesys, etc. Scaling can be reduced by physical and chemical processes. The flow hydrodynamics, stirring speeds and cross flow velocity controls scale deposition on the surface of the membranes. The details of the physical and chemical methods are available [20,31,32]. The antiscalants are widely used to reduce inorganic scaling. Most commonly used antiscalant includes surfactants, organic phosphates and polymers organic in nature [14]. Details of some sparingly soluble salts in given in Table 1 [20].

Salt	Name	Solubility [g/L]	Solubility Product K	Solubility reduced by
CaCO_3	Calcium carbonate (calcite)	0.24	3.3×10^{-9}	High pH, high temperature
$\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$	Calcium sulfate dehydrate (gypsum)	2.0 (as CaSO_4)	2.6×10^{-5}	High temperature (>60 °C)
SiO_2	Amorphous silica	0.12	1.9×10^{-3}	Low temperature

Table 1: Details of Sparingly Soluble Salts.

Colloidal Fouling

Colloids can be organic or inorganic in nature for example silica, iron and proteins. Their size ranges from few micrometers to some nanometer [33]. They easily pass through pretreatment and get enter into RO system. Size of particle plays an important role on disposition of colloidal fouling. Large size colloids do not get attached to membrane's surface in cross flow velocity mode. Deposition on membrane can be determined by charge present on the colloid for example cations have more probability to clog polyamide membranes [9,18]. A new cake enhanced osmotic fouling mechanism is explained by Hoak. The colloids forms a cake layer on the surface of membrane enhance concentration polarization, dropping

the diffusion of particles from boundary layer to bulk. This results in more salts concentration and reduced flux [34,35]. Colloidal fouling when combined with organic and inorganic foulants enhances the fouling propensity [36]. Silica, colloidal iron, and manganese are measured by standard laboratory tests. Colloidal fouling can be prevented by operating the membrane module below threshold velocity of 5×10^{-4} cm/s [37]. Pretreatment with Coagulation/Flocculation and UF/MF can reduce the colloidal fouling. Coagulation helps in neutralizing the charges on the colloid's surface and flocculation aids in flocs development. After flocs formation, these aggregated particles can be removed by sedimentation or MF/UF membranes [9]. Details and various factors affecting fouling caused by colloids are given in Table 2.

Foulants	Caused by	Maximum Permissible Limit For RO	Increasing Factors
Colloids	Iron (Reduced)	2.0 mg/L	Increase by High TDS, Temp, pH and high system's recovery
	Iron (Oxidized)	< 0.05 mg/L	
	Silica	< 100 mg/L	
	Total Hydrocarbons	< 0.02 mg/L	
Scalants	Barium Sulfate	< 6000% in concentrate	Microorganism species present, available temp, available nutrients,
	Calcium Sulfate	< 23% in concentrate	
	Calcium Fluoride	< 12000% in concentrate	
Organics	TOC	< 0.5 mg/L	Microorganism species present, available temp, available nutrients,
Micro-organisms	TOC:TN:TP	1:01:01	
	Chlorophyll	< 0.5µg/L	

Table 2: Permissible Limits and Factors for Various Foulants [38].

Pretreatment

Any type of reverse osmosis process is incomplete without pretreatment. All the foulants can be majorly controlled with pre-treating the feed solution with appropriate chemical/physical/biological treatment methods. The level and complexity of the pretreatment requirement depends upon feed water characteristics, efficiency of RO membrane process and product quality required. Pretreatment not only increase the performance of RO but also increase the life, quality of product and reduces physical and chemical cleaning requirements. Feed water quality is directly dependent upon source of water. For sea water desalination, water can be taken from two sources namely open sea water and beach wells. Beach wells resulted in less fouling as compared to open sea water intake [8,38].

Pretreatment includes following options:

- The first step in pretreatment includes coarse screens, fine screens, microscreens and cartridge filters. Their purpose is to reject large particles to subsequent pretreatment facilities [38,39].
- Conditioning of Saline water:
 - a. Second step to pretreatment includes conditioning of saline water using chemicals. Coagulation is done prior to sedimentation/dissolved air floatation/filtration. Commonly used coagulants are alum, ferric chlorides and polymers. Dose of coagulant is critically dependent upon source water quality (surface water, ground water, river water etc.). Overdosing may result in rapid fouling of the membrane. In addition to coagulation, flocculation is done to improve pretreatment [9,18,40].
 - b. Precipitation of calcium carbonate, calcium sulfate can be reduced by anti scalants or by removal of scalants with the help of nano-Filtration (NF) or softening. Some of the compounds naturally contained in seawater (such as humic acids) serve as natural chelating agents and scale inhibitors. Therefore, acidification of

seawater prior to membrane salt separation is not usually needed and commonly practiced.

- c. For boron removal, acidic pH will have a negative impact. A high pH results in precipitation of Boron. However, addition of acid lowers the carbonate concentration by converting bicarbonate to carbon dioxide. The carbon dioxide passes through the RO membranes and is removed or used in the post treatment system [8,38]. Chlorination can also be done to oxidize microbes present.
- Absence of sand and silt content is absolutely necessary. These materials can be removed by strainers, silting canals and basins, cyclone separators, lamellar sedimentation tanks [41].
 - Dissolved air floatation can be used to remove oil, algae, grease and other contaminants which are hard to remove with sedimentation. This methods lowers the turbidity of feed water to less than 0.5NTU. This method diffuses air from the bottom of the tank. Air brings foulants on the top which can be then scraped away [38].
 - Granular media filtration can be done using anthracite, sand, gravel in single or multiple layers. These filters can be used in single stage of multiple stages. They can be operated in up-flow or downward flow modes by inducing pressure or under the effect of gravity respectively [42].
 - Membrane filtration is the most modern and efficient type of pretreatment. Particulate, colloidal inorganic, and some of the solid and colloidal organic foulants contained in the saline source water can be removed successfully using MF or UF membrane pretreatment. Although at present less than 10% of all existing desalination plants worldwide have UF or MF pretreatment, application of membrane filtration for saline water pretreatment is gaining a wider acceptance over the past 10 years [43,44]. The number of medium and large desalination plants with

membrane pretreatment has increased from less than a half a dozen in 2002 to over 40 in 2011. Membrane systems can be divided in two main groups depending on the type of membrane elements they apply: pressure-driven (pressurized) and submerged. Pressurized membrane systems use membrane elements installed in pressure vessels or housings and the membrane separation process in these systems is driven by of 0.2-2.5 bar of pressure. Submerged systems use membrane modules/cassettes, which are immersed in tanks and operate under a slight negative pressure (vacuum) of typically 0.2-0.8 bar. The following issues are recommended to be considered when choosing between submerged and pressurized type of membrane pretreatment system [44,45]. Their removal efficiency for various pollutants is as follows:

- a. for organics ranges from 20% to 40% of the soluble organics
- b. For solids, up to 90% with effluent turbidity < 0.1 NTU
- c. For bacteria and viruses: 99.99% removal

Brine Disposal

The reject stream coming out of reverse osmosis system is known as brine. Proper disposal of brine is much needed with environmental and economic point of view. Improper disposal leads to negative impacts on marine life, ground and surface water contamination [46]. Following options are considered for brine disposals:

- Disposal into surface water bodies: this is the least cost solution. Brine can be discharged into surface water bodies like river, lakes etc. but they may result in exceeding the national environmental quality standards within short duration. Moreover this also puts a burden on water treatment cost for those who are taking raw water from these water bodies. But for sea water brine is generally disposed of back into water body. However, the intake port and brine disposal ports are very far to have minimal effect on feed water quality [8].
- Waste minimization by evaporating the water out of brine in specially designed lined evaporation ponds. Evaporation seems to be a non-practical solution because of high land requirements. However some researchers have come up with cultivation of brine shrimps in these ponds.
- Brine can be disposed of into sewer system where wastewater treatment takes its care.
- Resources like salts can be recovered from brine. These include minerals, salts and caustic soda etc.
- Brine can be provided as irrigation water to high salt tolerant crops. But this method needs proper lining of soil to reduce leaching of salts toward ground water.

Future Directions

Membrane technology is growing vastly from last 60 years. The cost and energy requirements are on the decreasing trend as more and more advancement are being made. The basics of RO process remains the same, nonetheless this process is highly dependent on raw water quality. Membrane surface modifications are also playing a pertinent role in the development of this process. Further improvements in membrane materials, the cost and energy involved is needed to be looked into in detail. Moreover the energy and resources recovery can play a major role in balancing the cost of the system.

Conflict of Interest

There is no conflict of interest between authors.

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