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Research Article

INDUSTRIAL WASTE WATER TREATMENT BY MEMBRANE SYSTEMS

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ABSTRACT
One method of treating industrial effluents is membrane processes. In addition to the simplicity of the process and lower cost of the initial investment, the membrane methods produce products with uniform and similar high purity not related to the inlet wastewater. This paper investigates the industrial and oil wastewater treatment by Ro, MF, UF, NF methods, and Hybrid Membrane Systems and the Membrane Bio- Reactor methods will be discussed. Among the disadvantages of investigated produced water treatment technologies, the followings can be pointed out: High cost of the treatment, the use of toxic materials, large space required for the installations, side pollutions, etc. Given the lack of such disadvantages in membrane methods, these methods have more utility in industries, especially oil and gas industries. Membrane is a porous medium made of polymeric, ceramic or metal material. The fluid passage through the membrane depends on its physical and chemical properties. These membrane environments or media perform the separation operation by selective passage of particles through themselves. Membrane systems can compete with more advanced technologies of wastewater treatment having a large volume of petroleum compounds. However, these membranes have also some defects, such as inability to separate dissolved gases, organic compounds with low molecular weight from the water. The membranes depending on the size of permeable particles are divided into four types: Microfiltration membranes (MF), Ultra filtration membranes (UF), Nano-filtration membranes (NF) & Reverse Osmosis membranes (RO).

***Keywords**: Water treatment; Industrial wastewaters; Membrane*

INTRODUCTION
About a hundred years ago, since the emergence of the relationship between the effect of pathogenic bacteria and microbes in outbreaks and transmission of diseases, the man thought to cleanse and purify the polluted waters. In other words, water and wastewater treatment technology in its current trend has arisen mostly due to advances in biology and medicine. This technology came to the consideration since gradual banning of wastewater discharge into the natural resources of water, primarily the rivers (Ahmed et al., 2001). Such preventions and restrictions led to the need for wastewater treatment and development of its methodologies. Over the time, with the development of cities and industries, the risk of environmental pollution and the resulting need for wastewater treatment increased with an unprecedented speed, and simultaneously, many techniques were provided, studied and used for wastewater treatment (Veil et al,. 2004).

Differences in industrial wastewater with domestic sewage
The properties of industrial wastewater and plants sewage totally depend on the type of the plants products. Considering this, the main differences between plants wastewater and domestic sewage may include:

A. The presence of chemical toxic compounds and substances in the plants sewage are more likely.

B. Higher corrosive properties
C. High alkaline or acidic properties
D. The presence of organisms in them is less likely.

Uncontrolled discharge of industrial effluents in surface waters leads to the mortality of aquatic animals, especially fish. It is interesting to note that the body decomposing of the same animals causes the conversion of the water bacteria activity from aerobic to anaerobic conditions (Parvini et al., 2012).The activity of anaerobic bacteria is combined with the emergence of unpleasant odor so CH4 has a stench smell and is flammable, while SH2 is malodorous and smells like rotten eggs. PH3 is toxic and dangerous and smells pungent garlic. Generally, most products resulting from activity of anaerobic bacteria are harmful for other living organisms, particularly aquatic organisms. Choosing the appropriate method for the purification and treatment of industrial waste water, reduced costs and increased quality of the product (water) can be guaranteed. In this regard, membrane processes having are of great importance due to having advantages such as reduced energy consumption, mass transfer, high efficiency and ease of use (Ghasemi et al, 2013).

The objectives of wastewater treatment include:
A. Stabilization of organic matter
B. Producing dischargeable wastewater in the environment and environmental protection
C. Reuse of water and solid materials resulting from wastewater treatment

DEFINITIONOF THE MEMBRANE

In general, a membrane is a barrier that separates the two phases and controls the transmission of different chemical components in a certain approach. In fact, the membrane is separator barrier that adjust the leakage and seepage rate of its adjacent chemical components. There are generally two phases in a membrane process that are physically separated from each other by the third phase (membrane). The phases are composed of components that one of the mixture components is transferred more than the others. In other words, the membrane has selectivity to one of the components. Hence, the transition of that component from one phase into another will be carried out by the membrane. Thus, one phase is enriched of that component and the other is depleted from it. Such a functioning can be seen in Figure 1.

Figure 1: Schematic model of the membrane functioning

In general, the membrane performs two main actions:
1. Permeability
2. Selectivity

For the membrane process, the selectivity is usually defined by disposal factor.

Formula 1 : the membrane selectivity

$R=1-C_p/C_r$

Where:
C_p: Concentrations of the components in the permeated flow
C_r: Concentration of the components in the non-passed (remained) flow through the membrane

The disposal rate, indeed, defines percentage of disposed components through the membrane. For a given driving force, the passing flux per unit area of membrane is inversely proportionate with its thickness. Then, due to economic reasons, the membranes must be thin as possible.

MEMBRANE PROCESS

Membrane separation processes commercially developed include:

Microfiltration

It is a filtration process in which porous membranes are used for the separation of suspension particles with diameters between 0.9-90 microns from the flow of gases and fluids. Therefore, the scope of application of microfiltration membranes occurs between ultrafiltration membranes and common filtration.

Ultrafiltration

Ultrafiltration is known as a process between microfiltration and nanofiltration. Ultrafiltration membranes usually have pores in the range of 10 to 1000 Å. With smaller pores size, the larger particles pass less through the membrane. Accordingly, the rate of fluid filtration is determined according to the pores size of the membrane. These membranes are also capable of disposing particles in the molecular weight range of 300 to 500000. Materials and particles normally excreted by these membranes include sucrose, bio-molecules and polymers and colloidal particles.

Nanofiltration

Nanofiltration is known as a process between ultrafiltration and reverse osmosis. The membranes used in nanofiltration contain pores close to or lower than nanometer that the size is usually around 0.5 to 1.5 nm. Some researchers believe the reverse osmosis membranes and nanofiltration membranes have no holes (pores), and instead, there are empty cavities between polymeric chains.

Reverse osmosis

As described in the introduction, reverse osmosis refers to a process in which the minerals are separated from water by membranes permeable to water and impermeable to the minerals. The water containing minerals are entered into the membrane by applying pressure from the feeding side, and the solute -free water is collected as the leaked flow. The application area of the water separation processes regarding membranes acting based on pressure gradient (reverse osmosis, ultrafiltration and microfiltration) is shown in Figure 2 (S.S. Madaeni., 1999).

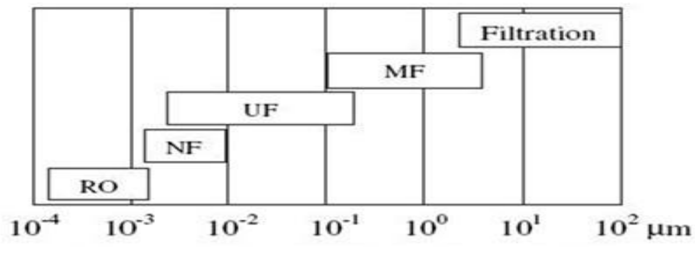

Figure 2: Basic principles of processes of reverse osmosis, ultrafiltration, microfiltration and filtration

Disposal rate of reverse osmosis membranes, nanofiltration membranes and ultrafiltration membranes for certain polluting substances are given in Table 1.

Table 1: Disposal rate of some substances in some NF, UF, RO membranes

	RO	NF	UF
Sodium chloride	99%	0-7%	0%
Sodium sulphate	99%	99%	0%
Calcium chloride	99%	0-90%	0%
Sulfuric acid	98%	0-5%	0%
Hydrochloric acid	90%	0-5%	0%
Fructose	>99%	20-99%	0%
Sucrose	>99%	>99%	0%
Virus	99/99%	99/99%	99%
Protein	99/99%	99/99%	99%
Bacteria	99/99%	99/99%	99%

SELECTINGTHE MEMBRANEMATERIAL

Proper selection of the membrane material for a specific process is very difficult and requires extensive data regarding the process environment to make a good choice.

Table 2: Propulsion (driving force), transition solution, and the permeated component for separation processes (Madaeni, 2003)

Membrane process	Ability to separation	Driving force provided by	Preferable permeable component
Osmosis	Aqueous solution	Concentration difference	Solvent
Reverse osmosis	Aqueous solution with low molecular weight - Aqueous-organic solutions	Pressure difference (100 bar)	Solvent
Ultrafiltration	Macromolecules solutions – Emulsion	Pressure difference (10 bar)	Solvent
Nanofiltration	All mentioned in RO & UF	Pressure difference (15 bar)	Solvent
Microfiltration	Suspension, Emulsion	Pressure difference (5 bar)	Solvent

In a very simple model of fluid movement within the membrane, the membrane is considered as a collection of cylindrical capillary tubes with a diameter of d. The flow of fluid within a cavity (q) is calculated by Paisley law as following:

Formula No. 2: Fluid flow inside a cavity (q)

$$q = \frac{\pi d^4}{128 \mu l} \Delta P$$

Where:
ΔP: Pressure difference inside the pore
μ: Viscosity of liquid
l: Length of the pore
The flux or flow rate per unit membrane area is the sum of all flows passing through the separated pores, which is as the following:

Formula No. 3: Flow rate per unit membrane area

$$J = N.\frac{\pi d^4}{128\mu l}\Delta P$$

Where

N is the number of pores per centimeter of membrane, which is inversely proportionate with squared diameter in case of membranes with similar pores number and same porosity.

Formula No. 4: the number of pores per centimeter of membrane

$$N = \varepsilon.\frac{4}{\pi d^2}$$

As a result, the flux rate is calculated according to the above equations as:

Formula No. 5: Flux rate

$$J = \frac{\Delta P \varepsilon}{32\mu l} d^2$$

INDUSTRIALWASTEWATER TREATMENT BY REVERSE OSMOSIS

The most advanced industrial water treatment method is the reverse osmosis desalination approach. In this process, water is passed through a series of semi -permeable membranes under pressure. The external pressure is higher than normal osmotic pressure. As a result, the smaller molecules pass through the pores of the membrane, while the larger molecules cannot pass through the membrane. Then, the larger molecules are passed in a lateral flow along the membrane and flushed. In RO Desalination systems, water is injected by high pressure pump into the semi-permeable membranes with tiny pores with a diameter of about 42 microns. These pores prevent the passage of molecules larger than water molecules, and thus, the permeate water is flowed from one side and the concentrated water (Brine) from the other side. Reverse osmosis can remove about 90 to 99% of all water T.D.S and colloidal material. These salts include sodium, potassium, sulfate, bicarbonate, and silicate salts, bacteria, viruses and other water soluble salts. The advantages of using a reverse osmosis system compared to other methods include the following: Economy, low power consumption, non- polluting the environment and ease of work with this device and reduced repairs and maintenance costs (Jerard et al., 1997).

The parts used in industrial water treatment by reverse osmosis method;

Industrial water treatment process by reverse osmosis or desalination method includes the following parts:

1. Sand Filter
2. Carbon Filter
3. Micron Filter
4. Infusion Pump
5. Layered Pump
6. Membrane
7. Electrical Panel
8. Production Flow-meter
9. Wastewater Flow-meter
10. Resistant chassis
11. Pressure switch
12. SOL

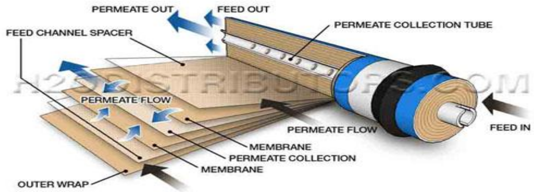

Figure 3: Sample of RO compartment

INDUSTRIAL WASTEWATER TREATMENT BY ULTRA FILTRATION METHOD

Like, reverse osmosis, nanofiltration and microfiltration, pressure is the driving force for mass transfer in the ultrafiltration. This pressure difference for ultra-filtration process is in a range between 2 to 5 bars, which is usually applied to the system through the pump and from the feed solution. Under these conditions, the solvent molecules and small dissolved solids pass through the membrane, while larger molecules and colloidal particles are not allowed to cross the membrane and remain behind it. Generally, the ultrafiltration units act as cross flow. In this method, the feed flows pressurized over the surface of the membrane and in parallel. In this case, only a little part of it is processed by the membrane. Therefore, in all practical cases of ultrafiltration use, the whole or part of the unprocessed fluid is returned back to the system. The reason to choose this method is generally to confront with the phenomena of concentration polarization and membrane fouling.

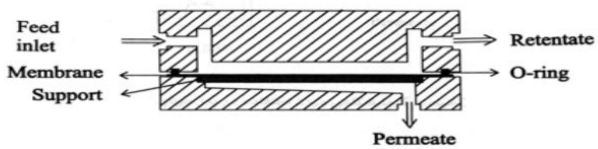

Figure 4: The cross-section of ultrafiltration cell

The system is designed in such a way that with avoidance of unnecessary complexities, all important operating parameters in the ultrafiltration process can be controlled. The experimental cell used in the laboratory system is composed of two separate pieces made of stainless steel according to the above Figure. These two pieces are connected to each other by bolts, and the space between them is completely sealed by an O-ring. The membrane plane is placed between the two pieces.Given that the membranes used in the structure are not perfectly homogeneous, the rate of pure water permeate for each piece of the membrane should be determined at the beginning of each test, and then used for feed processing. In all experiments, according to the requirements of each test, an accurate control was done on the feed flow rate control, operating pressure as well as the temperature. Using the liquid volume seeped in ten minutes and having the membrane area, the leakage flux was calculated and reported based on conventional unit:

Formula No. 6: Leakage flux

$lit/m^2.hr$

INDUSTRIAL WASTEWATER TREATMENT BY MICROFILTRATIONMETHOD

In this study, the effluent treatment of Tehran Refinery API unit was examined in a pilot microfiltration with cross-flow .The experiments show that the variables of pressure, feed cross-flow velocity, temperature and concentration are factors affecting volumetric permeate flux and the rate of rejection of organic hydrocarbons over the time.System performance at the optimum operating conditions prevents the rapid decline in permeate flux with time and will reduce the membrane fouling to a minimum. The membrane in the polyether sulfone microfiltration system is selected with pores size of 0.2 micron. Within the optimum operating conditions (pressure of 1.5 bar, cross-flow velocity of 1.25 m/s, and 35 ° C), the rate of oil separation was as 97.1% , and the treated effluent with the concentration of 209 mg/lit

could be discharged into the environment and used in agricultural applications well as. This methodology is also used in wastewater treatment of petrochemical units to separate organic pollutants. Preliminary studies show that the MF membranes can reduce the amount of petroleum materials of refining waste in the output to the permitted rates provided that the operating conditions in use would be optimized.
Doing the set of experiments, two types of feed were evaluated:

1. Synthetic Feed (water / gas oil/ surfactant) (a)
2. The API unit outlet effluent of Tehran refinery wastewater treatment plant (b)

Results of microfiltration use in wastewater treatment
According to studies, the hydrophobic membrane has a lower level of rejection than hydrophilic membranes due to the ease of formation of the gel layer on the membrane surface. Therefore, according to the results obtained for the feed (b), the low rejection at the beginning of the filtration system is justified. However, in continuing of filtration, with increased layer specific resistance and reduced porosity coefficient, the rejection rate increased up to 96.6%.
According to the mentioned, one can say that the membrane filtration in case of this type of wastewater is an acceptable and feasible approach. The acceptable advantages of this method include the appropriate specification of the external product (output), easy operational conditions and the membrane process combination capability with some other processes (adsorption, chemical, biological characteristics) as well as the simpler control of the system. In addition, low dependence of the output feature compared to the inlet feed characteristics is considered as so important points of this method. The conducted experiments represent that the operating pressure of 1.5 bars, the cross-flow velocity of 1.25 m/s and the temperature of 35 C° are the optimum operating conditions for the studied feed processing. The rejection rate of the system compared to oil separation in the actual feed is equal to 97.1 %, and the oil rate in the effluent is equal to 2.9 mg/lit with no environmental problem to be discharged (Rkabdar et al., 2009).

MEMBRANEBIOREACTORES (MBR)
Combination of biological processes with membrane separation is one of the newest wastewater treatment technologies rapidly growing in this industry. Biological reactors (MBRs) composed of a biological reactor (bioreactor) with suspended biomass and microfiltration membranes with pores diameter of 4-9 microns for separating solids have many applications in the wastewater treatment. These systems may be used in combination with bioreactors having aerobic or anaerobic suspended biomass.
The wastewater membrane systems can increase the quality of outlet wastewater to the quality of effluent resulting from incorporating secondary sedimentation and microfiltration. They may be replaced the sedimentation units, sand filtration and disinfection, which were used in Conventional CAS Activated Sludge methods to separate suspended solids (Asadi et al., 2007).

Simplifying figure to describe the MBR processes
In fact, the MBR systems perform microfiltration operations and biological treatment in a single processing unit. Thus, they can be considered as an auxiliary unit for secondary sedimentation and filtration, or totally eliminate the need for these units. Ability to remove the secondary sedimentation and performance at higher MLSS concentrations has the following advantages:

1. Higher volumetric loading up, and therefore the possibility to apply shorter hydraulic retention time
2. Occurring higher cellular retention time (SRT) with reduced sludge production
3. Performance at low Dos and the potential for simultaneous nitrification –denitrification in designs with high SRT
4. High quality effluent in parameters of turbidity, bacteria, BOD and TSS-
5. Less required space for waste water treatment

6. Less processing steps to achieve higher quality in output
7. Functionality in Higher MLSS compared to the traditional methods
8. Reduced amount of producing sludge
9. Eliminating concerns about sludge capability of sedimentation
10. Treatment capability of high BODs compared with traditional methods
11. Very low turbidity levels in the treated effluent
12. Lower cost of operation due to stable performance

In the first type, microfiltration membrane is directly located within the activated sludge reactor. The submerged MBR modules are placed in the biological tank of (activated sludge) or in the side tanks. Under vacuuming operation, while biomass remains behind the membrane, water passes through the membrane and module and exits.

Membrane modules contain membranes, their support, inlet and outlet feed connections and a major support. The membranes are exposed to a vacuum (less than 50 kPa) that pulls out the filtered water from inside the membranes, while the solids remain in the reactor. To maintain the TSS inside the reactor and to keep clean the outer surface of the membranes, the compressed air is distributed on the basis of membrane modules. As the air bubbles are moving to the surface, the membrane surfaces are washed and the air provides enough oxygen to supply the aerobic conditions.

In second type MBRs, the activated sludge is pumped from the bioreactor to the membrane; the solids remain inside the membrane and the filtered wastewater exits. The driving force is the pressure caused by high velocity inside the membrane. The solids remaining in the membrane are returned into the activated sludge reactor. The membranes are back washed periodically for separating the solids and chemically cleaned to avoid pressure drop.

Replacing the secondary sedimentation with this type of separation, the membranes overcome the sludge swelling problems and other problems related to gravity sedimentations. The MBR systems can operate at much higher MLSS than activated sludge reactors (15000-25000) mg per liter. Although these concentrations are available, but in practice the concentrations higher than 10,000 to 80,000 mg per liter would be very costly. The membrane flux, which is the volume or mass velocity of the passing flow from the membrane surface (liter per square meters per hour), is an important factor in designing that also strongly influences the design economy. Lower fluxes lead to higher concentrations of MLSS (Moslehi Moslehabadi et al., 2010).

INDUSTRIALWASTEWATERTREATMENTSYSTEMUSINGHYBRIDMEMBRANESYSTEMS

In combinatory or hybrid systems, different chemical, physical and biological technologies are used for pre-filtration of the membrane unit. Prior to 1980, the use of (Microfiltration / Ultrafiltration) MF / UF membranes for pretreatment was not common (Sharifi&Behzadi, 2013).

Separation process by hybrid MF-UF membranes

Microfiltration membranes are capable of separating particles with size ranging from 0.1 to 10 microns. Therefore, suspension and colloidal particles as well as big drops of oil present in the wastewater are removed by this process (Babapour et al., 2010)

Considering that the salts pass though the microfiltration membranes, the osmotic pressure difference between the two sides of the membrane is low, and the separation procedure requires a relatively small pressure difference approximately about 0.5 to 1 bar.

The ultrafiltration membranes pores are in the range of 1 to 100 nm, and are able to prohibit the passage of particles with molecular weight ranging from 300 to 500,000 g /mol (Madaeni, 2003).

Therefore, the smaller drops of oil present in water as emulsions or dissolved in the water can be removed by this process.

In ultra filtration, as microfiltration process, the osmotic pressure difference between the two sides of the membrane is low due to the passage of salt; thus, there is no need to high pressure for separation. The normal pressure required for ultrafiltration procedure is about 2 to 5 bar.
Due to separation of particles by size in these membranes, a layer of the remaining components is formed on the membrane surface over the time, which is the most important factor in reducing the membranes efficiency. The regulation of operating conditions and selecting the right membrane can delay this phenomenon (flux reduction) (Babapour et al., 2006, 2010).
An overview of the advantages and disadvantages of each of the membrane filtration methods

Table 3: Advantages and disadvantages of each membrane method

Process	Target pollutants	Separation efficiency	Advantages	Disadvantages
Microfiltration (MF)	- Suspended materials - Q & G - Suspended solids	- Very good - >5 mg/ l - >1 mg/l	Generally for membrane methods: • Lower energy consumption • Separation with no need to use material • Performing separation at ambient temperature • Low weight and size of separation equipment • Simple installation and operation • Minimum need for control, inspection and maintenance • Ease of access and possible use of separate phases (Chen at al., 1991)	Generally for membrane methods: • Membranes becoming dirty and decreased flow rate • Sensitivity to operating parameters and feed characteristics • Requiring constant cleaning and parts replacement (Liangxiong at al., 2003)
Ultrafiltration (Judd and Jefferson, 2003)	- COD - TOC - Polymers - Benzene, Toluene and Xylene - Heavy metals (copper, etc.) - Solids - Petroleum compounds	- 90% - 98% - - - 54% - 95% - 73% - 54%		
Nanofiltration (NF)	- COD - TOC - Oil	- 90% - 98% - Remained < 1%		
Reverse osmosis (RO)	- Separation of all compounds - Water - Ions (K^+, Na^+, Mg^{2+}, Ca^{2+})	-		
Biological Reactor Membrane	- COD - Activated Sludge	- 96% - -		In particular, in this case: • Higher membrane fouling • High fixed costs

DISCUSSION & CONCLUSION

Expansion of industry and technology development has caused a large variety of transformations and changes in supplying water for industry. Industry today requires water with special treatment due to using

very specific and expensive devices. In this regard, particular methods other than old and conventional methods, such as precipitation and ion exchange using resins should be used.
The most important and effective of these methods is the use of very thin membranes with pores with a diameter less than one micrometer. These membranes are capable of preventing the passage of soluble salts, and hereby, purify and treat water with a considerable efficiency.
Among the disadvantages of produced water treatment technologies that have been investigated, the followings can be mentioned

- High costs of treatment
- The use of toxic substances
- Too much space needed for installations
- Side pollutions

According to the absence of these flaws in membrane methods, such methods have more utility in industrial filtration.

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