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Membrane Distillation Review and Flux prediction in Direct contact Membrane distillation Process

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Abstract - *Membrane distillation (MD) is a process which utilizes differences in vapour pressure to pass water through a porous hydrophobic membrane and rejects other non-volatile constituents present in the feed aqueous solution. The Driving force employed for MD process is vapour pressure difference, induced by the difference in temperature across the hydrophobic membrane.*

This paper presents a review of MD and Flux prediction in direct contact membrane distillation (DCMD) process based on the published literatures till date and on preliminary analysis. The review covers the concepts such as membranes and modules design, configurations, Transport Mechanism, the terminology and fundamental concepts associated with MD, Commercially used membrane materials, MD membrane characterization, performance of MD process as well as an historical review of MD development, the heat and mass transfer phenomena, appropriate membrane properties, and applications of MD in different areas, and fouling in Membrane distillation.

There are four basic models for diffusion in MD; Knudsen number gives guidelines for identification of type of the model which can be the basis for further modelling. The DCMD theoretical mathematical modeling for a flat sheet membrane module is developed from the fundamental equation of mass and heat transfer. The Prediction of permeate flux can be done by using this model with the help of developed MATLAB codes and its results can be further compared with experimental process results.

Keywords: Membrane distillation (MD), direct contact Membrane distillation (DCMD), Heat & mass Transfer, Module configurations, Permeate flux, MATLAB.

1. INTRODUCTION

Membrane distillation (MD) is a thermally driven, membrane separation process [1-2], in which only volatile vapour molecules are transported through porous hydrophobic membranes. The liquid feed to be treated by MD is kept in direct contact with one side of the membrane and doesn't penetrate inside the pores of the membranes because hydrophobic nature of the membrane prevents liquid from entering its pores due to the surface tension forces. The driving force for the MD processes is quite different unlike other membrane processes, having the vapour pressure difference across the membrane rather than an applied absolute pressure difference, a concentration gradient or an electrical potential gradient, which drives mass transfer through a membrane [1, 3].The benefits of MD compared to other popular conventional separation processes are:

1) 100% rejection of ions, macromolecules, colloids, cells, and other non-volatile components.
2) Lower operating temperature than conventional distillation processes.
3) Lower operating pressure than conventional pressure-driven membrane separation processes.
4) Reduced chemical interaction between membrane and process solutions.
5) Less demanding membrane mechanical property requirements as it operating on lower operating conditions
6) Low vapour spaces compared to conventional distillation processes.

MD has also several limitations which must be overcome in order to this process become commercially successful for being a viable separation technology that includes:

1) Relatively lower permeate flux compared to other established pressure driven separation techniques.
2) Permeate flux decay with time due to concentration and temperature polarization effects, membrane fouling and total or partial pore wetting.
3) Desired Membrane availability and synthesis and difficulty in module design for MD.
4) Uncertain energy and economic costs for each MD configuration and its application.

2. MD TERMINOLOGY

The Terminology was defined at round table in Rome for Membrane Distillation separation process according to that [4-5] the MD process should have the following characteristics:

1) The membrane should be porous.
2) The membrane should not be wetted by process liquids.
3) No capillary condensation inside the pores of the membranes.
4) Only vapour should be passed through the pores of the membrane
5) The membrane must not change the vapour-Liquid equilibrium of the different components in the process liquids in any circumstances.
6) At least one side of the membrane is kept in direct contact with the process liquid.

The other terms commonly used to often define membrane separation process are,

1) *Thickness of the membrane*: - It gives information on both the mechanical strength of the membrane and the fluxes to be expected.
2) *Porosity of the membrane*: - The porosity of the membrane is defined as the volume of the pores divided by the total volume of the membrane in module.
3) *Liquid entry pressure*: - Pressure at which the liquid goes through a porous membrane. It is important in determining membrane wetting conditions.
4) *Fouling:* - The deposition of foreign material on the membrane surface or in its pores, which may lead to change in the membrane performance.
5) *Membrane Module*: - The smallest practical unit containing one or more membranes and supporting materials.
6) *Permeate*: - The portion of the feed as a desired useful product pass through the membrane.
7) *Pore size*: - This gives idea about size of the openings in a membrane; this term is used as an alternative to pore diameter and pore radius. For calculations uniform pore size is taken in consideration.
8) *Retentate:* - The portion of the feed is not passing through the membrane pores. Generally this stream is recycled back for higher efficiency.
9) *Flux*: - Amount of permeate, or components in the permeates, that is transported through a membrane per unit of membrane area and per unit of time.

3. THE HISTORY AND EVOLUTION OF MD

Researchers are working on membrane distillation processes since 60th decade. On 3 June 1963, Bodell filed the first MD patent [7] and in 1967, after few years Findley published the first MD paper in the international journal Industrial and Engineering Chemistry Process Design Development [8].Findley used the DCMD configuration using various types of membrane materials for his research. Based on his obtained experimental results, he outlined the most suitable membrane characteristics needed for a MD membrane. The same year1967 another U.S. scientist Weyl filed patent on 14 May 1964 [9].He claimed an improved method and apparatus for the recovery of demineralized water from saline waters using DCMD. In his patent, the use of suitable hydrophobic membranes made of Polytetrafluroethylene (PTFE), polyethylene (PE), polypropylene (PP) and polyvinyl chloride (PVC) was also suggested [9].In 1968, a year later a second U.S. patent was made by Bodell [10], in a continuation of his previous work on first U.S. patent [7].This patent provided novel apparatus and methods for desalting seawater and many other aqueous separations in an economical way. He described a system and a method to convert importable aqueous solutions to potable water using a parallel array of tubular silicone membranes having a 0.3 mm inner and 0.64 mm outer diameters, without presenting pore size and porosity Parameters. Bodell also work on other configurations of MD. For SGMD system he recommended the water vapour pressure in the air side to be at least 4 KPa below that of the aqueous medium. He also invented vacuum membrane distillation configuration and for the first time, an alternative means of providing low water vapour pressure in the tubes by applying vacuum emerged out[7,10].

A second MD paper was published by Findley and co-authors [7, 9-11]. An empirical correction related to the possible internal condensation and diffusion along the surfaces has been considered to perform their calculations [11]. In Europe, seawater desalination by using Swing gas membrane distillation process using dry air was also invented by Van Haute and Henderyckx [12, 13], another scientist Rodgers [14, 15] filled back to back two patents and presented a system and a method of desalination by use of a stack of flat sheet membranes separated by non-permeable corrugated heat transfer films with DCMD configuration. After these 5-6 years of continuous work on MD, interest in the MD process gone down quickly losing its brightness due to the observed lower MD production i.e. lower flux compared to that of well-established those times pressure driven processes.

MD process recovered once again interest within the academic communities in the early 1980s when novel membranes with good characteristics and modules became available [16, 17]. A Gore-Tex membrane, which is an expanded PTFE membrane having a thickness of 50 mm and 0.5 mm pore size, porosity up to 80% was used first by Esato and Eiseman [17-18].

Cheng proposed other types of MD membranes, method and apparatuses and Wiersma in a series of U.S. patents [19-22].The object of his patent [19], filed in 14

February 1979, was to provide an improved thermal membrane distillation process with continuous distillate production with maximum flux for a long period of time. The back to back filed patents [20-22] by Cheng and Wiersma claimed the usefulness of composite membranes having a thin micro porous hydrophobic layer and one or two thin hydrophilic layers. In the fourth patent, [22] filed by Cheng and Wiersma on 4 March 1982, he came with improved apparatus and method for MD using a composite membrane comprising of a micro porous hydrophobic layer having deposited on an non-porous hydrophilic coating.

The Swedish National Development Co. developed the plate and frame membrane module which can be applied in the AGMD configuration [23-24].On similar lines German company Enka A Galso presented polypropylene (PP) hollow fiber membranes in tubular modules in DCMD configuration with heat recovery mechanism at the Europee-Japan Joint Congress on Membranes and Membrane Processes, held at Stresa (Italy) in 1984 [25-27]. In the same congress, other papers on MD also have been communicated [28-29].This renewed interest resulted in the development of various types of porous hydrophobic membranes used for different MD configurations [30-34].

Recently, interest in MD has increased significantly as can be seen in Fig.1, which represents the number of published papers in journals per year in the MD field.

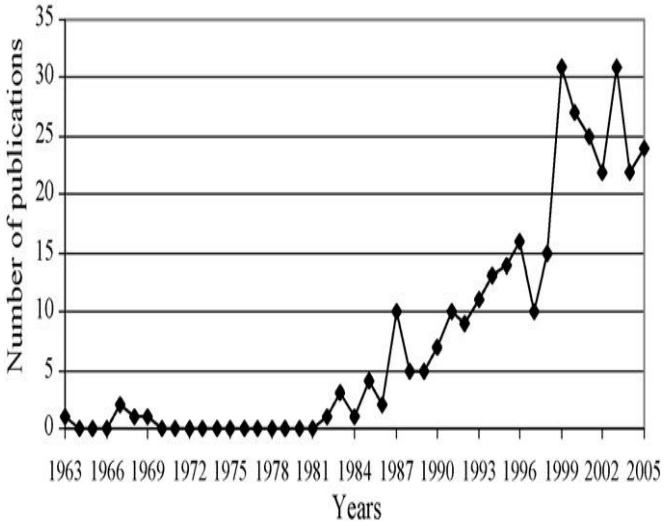

Fig-1: The work performed on MD process.

The number of total till date MD papers published in international journals are considered to be more than 2000.Within the published papers in international journals DCMD is the most studied MD configuration, as per Fig.2, 63.3% of the MD study is focused on DCMD. On the contrary SGMD is the least studied configuration, which contains only 4.5% of the MD published papers. This is because it requires external condensers to collect and store permeate and a source for gas circulation which may result into costly process. The majority of published papers are concerned with theoretical models of MD and experimental studies on the effects of the various operating conditions.50.4% of the MD publications dealt with theoretical models 40.5% for DCMD, 48.2% for AGMD, 40.8% for VMD and 72.5% for SGMD comparative to that only about 17% of the MD papers reported in journals are focused on the preparation of MD membranes.

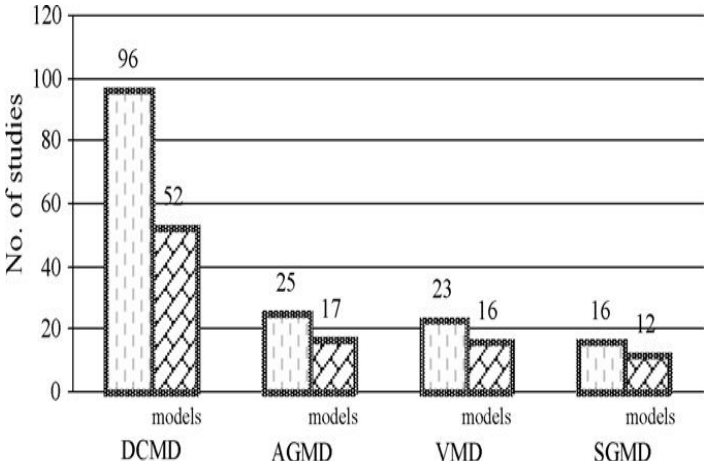

Fig-2: Bar chart for papers published in journals for different MD configurations and theoretical models.

Very few authors have considered the possibility of manufacturing novel membranes and membrane module designs specifically for MD applications [37].

4. THE MEMBRANE DISTILLATION PROCESS

MD phenomenon is very simple. A heated aqueous feed solution is brought in to the one side of hydrophobic micro-porous membrane interface. The hydrophobic nature of membrane prevents penetration of liquid resulting in vapours liquid interphase at each pore entrance. A variety of methods can be used to impose vapour pressure difference across the membrane to drive flux. The permeate transports through the membrane in three steps:

1) Vapour are formed at the hot feed solution– membrane Interface;
2) The same vapours are transported through the micro-Porous membrane;
3) Condensation of the vapour takes place at the other side Of the membrane

There are 04 types of MD configurations are available and can be used to drive flux. Each of the MD configurations has its own advantages and disadvantages for any given application.

1) Direct contact membrane distillation (DCMD)
2) Air gap membrane distillation(AGMD)
3) Sweeping gas membrane distillation(SGMD)
4) Vacuum membrane distillation(VMD

a) DCMD-Direct contact membrane distillation

In this configuration, the feed and permeate both liquids are in direct contact with the hydrophobic micro-porous membrane. This is most widely used configuration. Pure cold water is used as condensing fluid at permeates side in desalination.

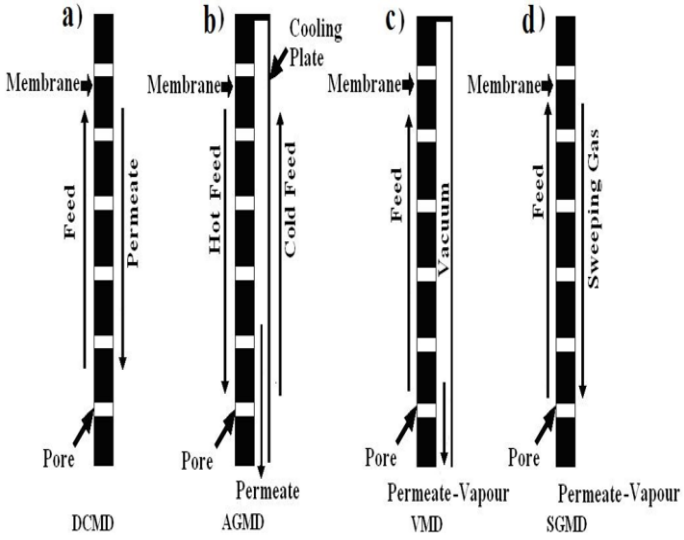

Fig.3 Different MD configurations: (a) DCMD; (b**)** AGMD; (c) VMD; (d) SGMD.

b) AGMD -Air gap membrane distillation

Water vapour is condensed at the permeate side separated from the membrane with an layer of air gap which can be in mm, thus heat losses are reduced in this configuration by addition of a layer of stagnant air gap between membrane and condensation surface at the cost of increase in mass transfer resistance by many fold.

c) SGMD -Sweeping gas membrane distillation

An inert cold gas is used in permeate side for sweeping the vapour molecules to outside the membrane module for condensation to take place. Here both heat and mass transfer resistance are reduced, but the operational cost is increased due to condensation system employed externally, SGMD is the least used configuration among all these configurations due to extra cost.

d) VMD -Vacuum membrane distillation

The driving force maintained by applying vacuum at the permeate side with the help of external agencies such as vacuum pump or steam jet ejector. One should take care that the applied vacuum pressure is lower than the equilibrium vapour pressure to avoid LEP.

Among the four configurations mentioned above, DCMD has the highest heat conduction losses because of the higher heat transfer coefficient on the permeate side for this configuration which results in relatively low thermal efficiency [39-40]. It is most commonly used in studies & commercial applications.

In AGMD, the air gap is usually controls the mass transfer process [41] because of its thermal and mass transfer resistances [42-43] AGMD has high heat efficiency than DCMD, due to the low temperature difference hence less driving force across the membrane is needed for same results [43-45].

SGMD has higher mass transfer rate than any of the MD type and has less heat loss through the membrane than DCMD, but, an external condenser which need be used for cooling and an air blower or compressed air to maintain operation. Energy use and operating costs causes SGMD less economical [46].

In VMD, permeate is continuously removed from the vacuum chamber to maintain the vapour pressure difference across the membrane surfaces. This provides the highest driving force at the same operating conditions. The cost of creating vacuum also makes this configuration less favorable.

DCMD is the most popular and mostly used for MD laboratory research, with more than half of the published references for membrane distillation based on DCMD [1-2, 45, 47] and AGMD is more popular in commercial applications, because of its high energy efficiency and capability for latent heat recovery [48].

5. MD TRANSPORT MECHANISM

In MD process, heat and mass transfer both takes place simultaneously through porous hydrophobic membrane surfaces. The mass transfer occurs through the pores of the membrane and heat is transferred through both the membrane matrix and in its pores. The heat transfer within the membrane is because of the latent heat accompanying vapour and the heat transferred by conduction across both the membrane material and the pores. There are a presence of fluid boundary layers adjoining both the feed and permeate membrane sides which creates temperature polarization and concentration polarization which is one type of resistance to MD process. Based on the kinetic theory of gases the transport of gases and vapours through porous media has been widely studied and many theoretical models have been developed.

The various types of mechanisms proposed for the mass transport are Knudsen flow model, Poiseuille flow model, ordinary molecular diffusion model or the

combination of any two or all three often called as the dusty gas model or transition flow model. In MD processes, the transport of molecules through the membrane matrix is just ignored because the diffusion area of the membrane matrix is so small that could not be compared with the pore area. The surface diffusion may affect MD performance in membranes with small pore sizes, for pore size less than 0.02 mm [49, 50].The main difference between per-vaporation and VMD is the characteristics of membrane [51]. It is observed that the heat loss by conduction through the membrane matrix becomes less important and significantly low when the MD system employed for high operating temperatures.

6. MEMBRANE MATERIALS

The commercially used micro-porous hydrophobic membranes made up of different polymers such as polypropylene(PP), polyvinylidene fluoride(PVDF), polytetrafluoroethylene (PTFE) and polyethylene (PE) available in tubular, capillary or flat sheet forms which have been used in MD experiments.

The porosity range of the membranes used is 0.50 – 0.90, pore size commonly used is in the range of 0.4 to 1.0μm, and the thickness is in the range of 0.04 to 0.25 mm [53-54].

The role of some commercial membranes in the MD process transport has been discussed by several publishers of scientific journals in details [54].

Table-1: Surface energy and thermal conductivity of commonly used membrane materials

Membrane Material	Thermal Conductivity (W m^{-1} K^{-1})	Surface Energy (×10^{-3} N/m)
PTFE	~0.25	9–20
PP	~0.17	30.0
PVDF	~0.19	30.3

Among all materials, PTFE has the superior properties like hydrophobicity, largest contact angle with water, good chemical and thermal stability and higher oxidation resistance compared to PP and PVDF. One has to study membrane properties since every material has its advantages and disadvantages for specific uses [47].Now days, new synthetic, composite materials, such as carbon nanotubes, fluorinated copolymer materials and surface modified PES [55–58], have been developed to make MD membranes with superior mechanical strength and high hydrophobicity and porosity. Sintering, stretching, and phase inversion are some of the methods used to fabricate MD membranes from these materials [59–60].Surface energy and thermal conductivity are properties to be considered while selecting membrane material for use [61-63].

7. CHARACTERISTICS OF MD MEMBRANE

MD membranes should be characterized by the following membrane parameters: [1- 2].

1) Liquid entry pressure (LEP):- It is the minimum Trans membrane hydrostatic pressure that must be applied before liquid solutions penetrate into the membrane pores.
2) An adequate thickness:- as membrane becomes thinner which tend to increase flux and tend to reduce heat efficiency or interface temperature difference
3) Pore size distribution:-Limited by the minimum Liquid Entry Pressure (LEP) of the membrane material. In MD, the hydrostatic pressure < LEP to avoid membrane wetting.
4) High hydrophobicity:- It helps for increasing flux, membranes made from more hydrophobic material will be applicable under higher pressures for a given pore size;
5) Low thermal conductivity:- High thermal conductivities increases sensible heat transfer and reduce vapour flux due to reduced interface temperature difference;
6) High porosity: - High porosity decreases both the thermal conductivity and increases the permeability of MD membranes, which results in increase in heat efficiency and flux, however, high porosity membranes bears lower mechanical strength and cracks or compress under mild pressure and are very susceptible for wetting, which results in the loss of membrane performance.
7) Good thermal stability: - Membrane should withstand temperatures as high as 100 C.
8) Excellent chemical resistance: - Should have excellent chemical resistance to various feed solutions and acids and bases especially if the membrane has to be cleaned.

The choice of a membrane for a given MD application is a compromise among a low thermal conductivity achieved by thicker membranes, a high permeate flux achieved by thinner membrane, a large pore size as well as a high porosity and a high separation factor [64].

8. MEMBRANES MODULES

Mostly used modules by MD researchers are Plate and frame, spiral wound, tubular, capillary and hollow fiber membrane modules. Flat sheet modules are most versatile and widely used than all modules. This model is easy to prepare, handle and mount.

Spiral-wound modules have a packing density more than flat sheet but these modules have disadvantage of being sensitive to fouling. These are normally shell and tube type modules. It is very compact module and one may face great difficulty in dismantling and packing for maintenance purpose.

Tubular membrane modules are considered one of the best modules with moderate a packing density. This model provides higher cross flow velocity and pressure drop they are generally employed for viscous solutions. Capillary modules are available with diameters of membranes 0.2-3 mm and with moderate packing densities. The advantage of this module is that Production costs are very low and membrane fouling can be controlled by maintaining proper feed flow and back-flushing of permeate in periodic time intervals, but limitation is that capillary membrane module requires low operating pressure.

9. MEMBRANE FOULING AND WETTING

Membrane fouling is one of the major constraints in the application of membrane technologies [65-66], as it causes flux to decline with time. Although membrane distillation is more resistant for fouling than other pressure driven processes, Dosing of antiscalants and anti-corrosion agents continuously can be used to prevent scaling and corrosion problems [67–69].Dow et al. showed that lower feed temperatures can substantially reduce the influence of fouling in MD [70].

Hydrophobic MD membrane is used as the barrier between the feed and permeate, membrane wetting reduces the rejection of the non-volatiles hence the efficiency of the process. Membrane wetting can occur under the following conditions,

1) When the hydraulic pressure applied on the surface of the membrane is greater than the Liquid Entry Pressure.
2) On long term operation or in treating high-concentration feeds for e.g. brine crystallization. The fouling on the membrane surface can effectively reduce the hydrophobicity of the membrane by lowering surface tension, [71,73]
3) When presence of high organic content or surfactant in the feed, this lowers the surface tension and reduce the hydrophobicity of the membrane & lead to membrane wetting [72].

10. FLUX DECAY

Different membrane types may exhibit different degree of fouling, for different period, as fouling depends on hydrophobicity, membrane surface structure, initial permeability and feed solution, [74]. Franken et al.[75] Observed 30% flux decay for 01 month of MD operation. He explained flux decay in terms of wetting; he hypothesized as time passes more and more pores become clogged and result in wetting. Another mechanism of flux decay is fouling that can cause pore clogging and thereby making available less area for vapour transport. There are three main common types of fouling [76] which lead to flux decay is,

1) Biological fouling
2) Crystallisation fouling
3) Particulate and corrosion fouling.

To fully understood flux decay and fouling phenomena more research need to be done in this area to find out the mechanism and real causes of fouling.

11. EFFECT OF OPERATING PARAMETERS ON MD FLUX.

There are several operating parameters affecting the permeation flux in membrane distillation process such as feed temperature, coolant temperature, feed flow rate and velocities, pore size, feed stirring rate and many more[77-79].The higher the feed temperature, the higher the flux due to the increase in vapour pressure gradient and vice versa for coolant temperature. In this way the increase in Reynolds number, induced by increasing recirculation rate, causes the decrease in temperature polarization and also concentration polarization, and consequently improves the mass flux. Flux also decreases with the increase in feed concentration because of the decrease in water activity and in heat and mass transfer coefficients in the boundary layer. The effect of vacuum on flux in VMD and effect of air gap in AGMD on flux is also important to study. The effect of different parameters is studied by several researchers for different types of configurations [77-79].

12. RESISTANCES IN MD

Heat transfer is achieved by two different ways, conduction through the membrane material and vapour within the membrane pore, and transfer of the heat of vaporization associated with the vapour flux. Conduction of heat across the membrane is considered as a loss of heat in the process which is large in DCMD, and should be as possible as minimized.

Dusty gas model is generally applied to describe mass transfer across the membrane. It consists of combination of Knudsen diffusion, molecular diffusion, surface diffusion, and viscous flow. Surface diffusion is always neglected in membrane distillation. Many researchers concluded that the DCMD process is molecular diffusion limited [80-81] and Knudsen-molecular diffusion transition [82-83] models were successfully applied to predict flux by using different software's in different platforms such as Aspen plus, Chemcad and MATLAB.

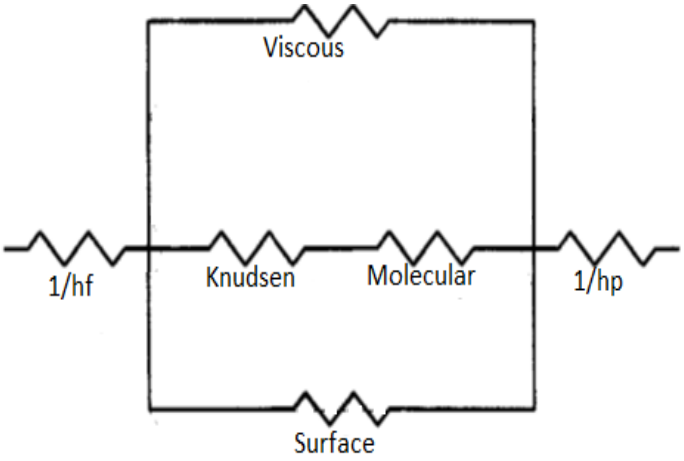

Fig- 4: The Mass transfer resistance Analog across the membrane in MD

The above fig.4 illustrates mass transfer resistances in MD analogical to electrical resistances. Boundary layers adjacent to membrane contribute to overall mass transfer resistance formation. There are mainly two types of resistances,

1) Knudsen resistance: - Resistance arises due to transfer of momentum and collisions of diffusing molecules with wall.
2) Molecular Resistances:-Resistance to mass transfer from collisions of diffusing molecules with other molecules.

As discussed above and concluded by many researchers Viscous or molecular diffusion is often rate limiting step in MD mass transfer [114].

Many Researchers studied resistances across DCMD and they studied that the transport resistance in the feed boundary layer plays important role at low cross flow velocity, and tend to decrease with increasing cross flow velocity. On comparison between systems operating at different feed temperature which showed that the higher is the feed operating temperature, the greater is the resistance in feed boundary layer [83].This shows that temperature polarization effect observed at high temperature. In contrast, the mass transfer coefficient across the membrane is proportional to the diffusivity, which is a function of the temperature and its equation can be given as

$$D_w = \frac{1.895 \times 10^{-5} T^{2.072}}{P} \qquad (1)$$

This contributed in the reduction of the resistance of the membrane with increasing operating temperature. DCMD system operating at high cross flow velocity shows minimum boundary layer resistance and thus the significantly higher flux. In novel DCMD processes efforts are being made to increase cross flow velocity by using spacers and maintaining optimum feed temperature to increase flux and reduce resistances across boundary layers

13. DIFFERENT TRANSPORT MECHANISMS AND POLARIZATION PHENOMENA IN MD.

13.1 Heat transfer

The mechanism of transport in MD involves simultaneously both heat and mass transfer processes. Heat transfer in all MD configurations is very important and is the rate controlling step in the MD process.

Heat transfer is carried out in four steps as given below:

1) Heat transferred from the feed solution to the membrane in the feed side of the membrane module. It is associated to the temperature polarization effect.

2) Heat transported by conduction across both the membrane matrix and the gas filled pores is considered as heat loss in MD;

3) Heat associated to the phase change i.e. latent heat of vaporization and therefore to the mass transfer through the membrane pores

4) Heat transfer from the membrane surface to the permeate solution across the thermally boundary layer in the permeate side also associated to the temperature polarization effect

The heat transfer boundary layer formed at each side of the membrane surface creates a resistance to heat transfer and makes the temperature difference at the membrane interfaces lower than that applied at the bulk phases. This is the real reason for low fluxes in MD process are due to the effect of temperature polarization. Up to 60-80% reduction in driving force is observed due to the temperature polarization effect [84-87]. The term temperature polarization coefficient (TPC) was defined as the fraction of the trans-membrane temperature to the bulk temperature difference and given by the formula below.

$$TPC = \frac{Tfm - Tpm}{Tfb - TPb} \qquad (2)$$

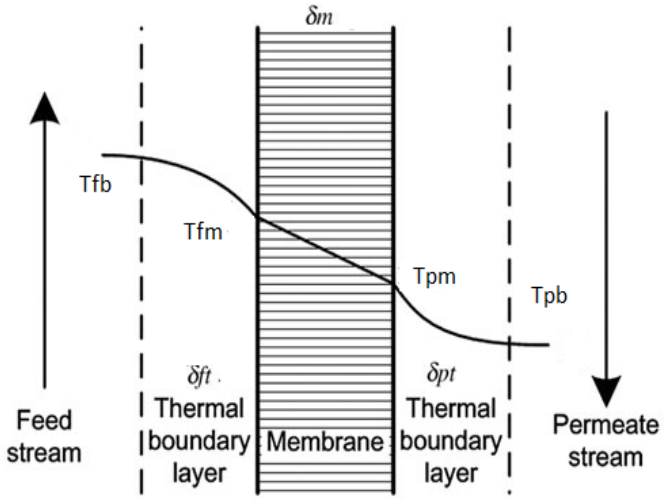

Fig-5: Temperature polarization Effect in MD.

Where T_{fm}, T_{pm}, T_{fb} and T_{pb} are membrane surface temperatures and fluid bulk temperatures at the feed and permeate side respectively. It can be seen from fig.5 that due to the temperature polarization effects, the bulk feed temperature T_{fb} is gradually lowered to T_{fm} across the developed thermal feed boundary layer of thickness δft, Similarly, at the permeate side the temperature at membrane surface (T_{pm}) is little higher than that of the (T_{pb}) permeate bulk phase temperature because of the development of thermal boundary layer of thickness δpt. Both T_{fm} and T_{pm} are function of fluid properties, operating conditions, and the hydrodynamic conditions. Optimum mixing conditions has to be maintained in order to diminish the temperature polarization effects.

Attempts are being made to enhance heat and mass transfer by improving the proper design of flow passage, membrane arrangement or by applying turbulence promoters like mesh spacers & making rough surface. High flow velocities is always an preferred option that is usually considered by MD investigators but it is associated with danger of membrane wetting, especially when membranes of large maximum pore size and organic aqueous solutions are employed, so flow velocities should be always increased with precautions.

The TPC has relation with heat transfer coefficients involved in the MD process [1, 44] as MD is depend on phase change for separation process, the latent heat of evaporation is continuously transferred from the bulk feed to the membrane surface. The supplied heat Qf, depends on the heat transfer coefficient (hf) and the difference between the feed bulk temperature and membrane surface temperature, if the heat loss is considered zero

$$Qf = hf(Tfb - Tfm) \quad (3)$$

The TPC increases with the T_{bf}, but higher T_{bf} usually results in an exponential increase in the flux rate [88-93].

13.1.1 Heat transport by conduction through the membrane.

In MD, the heat transferred by conduction through both the membrane matrix is considered as major heat loss and should be minimized to zero to minimize temperature polarization effects and to increase efficiency of the MD process. The equation is used for the heat flux transferred by conduction is given below,

$$Qm = hm(Tfm - Tpm) \quad (4)$$

Where, hm is the h.t.c, which can be evaluated as follows.

$$hm = \frac{km}{\delta m}$$

Where, km is the thermal conductivity of the membrane is calculated by,

$$km = \varepsilon ks(1 - \varepsilon)kg \quad (5)$$

The thermal conductivity of air is lower than that of the membrane material. Therefore, the heat lost by conduction is minimized by using membranes with high void volume.

13.1.2 Efficient Heat in MD

The heat that contributes to the evaporation step is considered the efficient heat. The efficiency of the MD process is best when temperature polarization effect and the internal heat loss by conduction through the membrane and the external heat lost to the environment are minimum. The heat flux can be estimated by the following expression.

$$Qv = N*H_v \quad (6)$$

The total heat transferred through the membrane whether it is considered efficient heat or heat lost by conduction Qt=Qm+Qv can be written as follows:

$$Qm + Qv = hm(Tfm - Tpm) + N * Hv \quad (7)$$

Heat Flux at permeate side which is also depend on temperature polarization but in case of DCMD it is considered to be very small as coolant liquid is in direct contact to permeate liquid and is given by equation.

$$Qp = hp(Tpm - Tpb) \quad (8)$$

Following equation is considered at steady state and considering no loss

$$Qf = Qm + v = Qp \quad (9)$$

The heat transfer coefficients of the boundary layers are usually estimated from the well-known heat transfer empirical correlations like Dittus Boelter equation or Sieder Tate equation which relates the dimensionless Nusselt number with the Reynolds and Prandtl numbers [94-95].

13.2 Mass Transfer

Fig . 5 Shows mass transport depend on the volatility of the solutes in the aqueous feed solutions. The mass transport of the volatile species is taking place in 03 steps given below.

1. The bulk feed to the membrane surface;
2) Mass transport through the membrane pores in gaseous phase;
3) From the membrane surface at the permeate side to the bulk permeate phase.

13.2.1 Concentration Polarization

As shown in fig.6 The increase in concentration of non-volatile compound next to the membrane surface layer at feed side results in reducing the trans-membrane flux due to the establishment of concentration polarization (CPC) of thickness, δfC, that acts as a one of the mass transfer resistance to the volatile molecule species. In other established pressure driven membrane separation processes such UF and RO, concentration polarization is usually considered a major cause for flux decline [96-98] but in MD process, with the help of moderate flow rates and high heat transfer coefficients it can be reduced to zero or can be maintained lower than that of the temperature polarization effect [97-98].

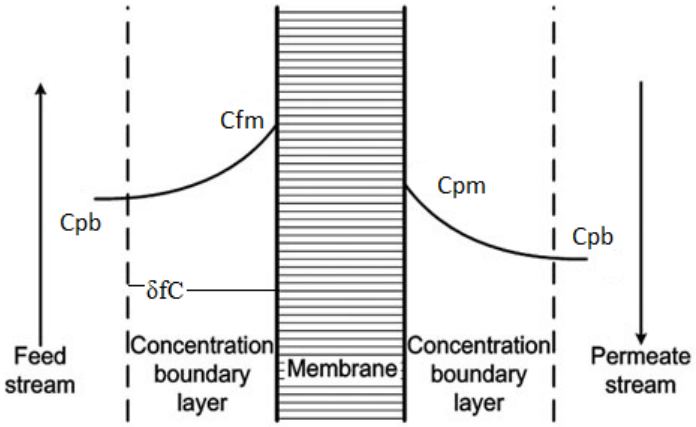

Fig-6: Concentration polarization in MD.

13.2.2 Mass Transport through the Membrane Pores.

The mass transfer process in MD is due to vapour pressure gradient between feed side and permeate side of the membrane. There are three basic mechanisms or models for mass transfer known as Knudsen-diffusion (K), Poiseuille-flow (P), Molecular-diffusion (M) or the combinations of them known as transition mechanism or dusty gas model [1,96,99-106]. According to every model molar flux N, is proportional to the vapour pressure difference across the membrane.

$$N = Cb[Pfm - Ppm] \quad (10)$$

Where, C_b is the Membrane Mass transfer coefficient or permeability. The membrane mass transfer coefficient C_b is function of both the operating conditions and the membrane structure and depends upon membrane distillation configuration employed.

P_{fm} and P_{pm} are the vapour pressures as function of temperature at both the feed and permeate at the membrane surface, respectively which can be determined by Antonie equation [113].

13.2.3 Rate controlling step in MD process

To improve the MD systems and to overcome the MD resistances the knowledge of the rate controlling steps in each MD configuration is important. It is observed that the rate limiting step is always heat transfer through boundary layers at low flow rates and membranes with small pore size and low porosity, and at higher flow rates the membrane resistance becomes very much predominant and becomes rate limiting step [1,79,108].Studies have been conducted by various scientists in determining the rate controlling steps in mass transport [107,109].Many researchers observed that that mass transfer within the liquid phase boundary layer dominates the mass transport process due to development of concentration polarization in that regime [107].The mass transfer resistances as well as the heat transfer resistances plays an important role in the transport processes, the contribution of which is different for different mixtures and is more important for the processes involved highly volatile components [109].

The rate controlling step may switch from being mass transfer controlled in the gas phase to mass transfer controlled in the liquid phase and vice versa which depends on the hydro-dynamic conditions and the membrane module design [111-112,105].The rate controlling step in MD process is differs from one MD configuration to configuration and also depend on the module type and its geometry, flow mode and operating condition, as well as membrane morphology and for which applications it is being used.

14. MATHEMATICAL MODEL DEVELOPMENT FOR PREDICTION OF PERMEATES FLUX (N)

Prediction of permeate flux can be done with the help of theoretical model for DCMD by using developed MATLAB codes. The following assumptions are considered in while developing the theoretical model:

1) The system is steady state

2) No pressure drop inside the air gap i.e. constant pressure is considered in the air gap
3) Membrane material has no selectivity toward any species.
4) Liquid entrance pressure (LEP) is greater than the pressure at the feed side of the Membrane.
5) No heat exchange between the system and the surrounding.
6) Film-wise condensation is considered in the cooling flat plate.

The following important major steps are considered in the heat transfer analysis such as:

1) Heat transfer by the convection from hot feed solution to the membrane surface,
2) Heat transfer by the conduction across the membrane material is the addition of latent heat and sensible heat,
3) Heat transfer by the condensation at the cold surface,
4) Heat transfer by the conduction through the cold plate,
5) Heat transfer by the convection from the cooling surface to the cooling water.

14.1 Procedure for calculation of permeate flux (N)

For the permeate flux prediction (N), following steps with iterative method has to be adopted.

1) Initially, the membrane surface temperature at the feed side (Tfm) and membrane surface temperature at the permeate side (Tpm) is assumed.
2) These assumed temperatures are less than bulk temperature in feed side & greater than coolant temperature at permeate side then utilized to estimate the Antoine equation and determine the vapour pressure at membrane surface (Pfm) and condensing film surface (Pfm) [113]

$$Pfm = \exp\left[23.1964 - \frac{3816.45}{T_{fm} - 46.13}\right] \quad (11)$$

$$Pfm = \exp\left[23.1964 - \frac{3816.45}{T_{pm} - 46.13}\right] \quad (12)$$

3) The flux (N) can be determined by using equation (6) [115-118] where C_b is Membrane Permeability and it is different for different type of diffusion, Knudsen number is depend on mean free path λ which is again depend on (KB) Boltzman constant.

$$Kn = \frac{\lambda}{dp} \quad (13)$$

Knudsen number provides the guideline in identifying the type of diffusion in mass transfer to be considered. Molecular diffusion, Knudsen diffusion and combination of this two called transition diffusion as the types of model.

(Kn >10 or dp < 0.1λ) –Knudsen type of Diffusion,
(Kn <0.01 or dp > 100λ)-Molecular type of Diffusion,,
0.01 < Kn < 10 (0.1λ < dp < 100 λ).Transition type of Diffusion,

Transition diffusion is considered here and equation for transition diffusion for permeability given as below [23-24],

$$Cb = \frac{M_w \varepsilon P D_w}{(RT\delta_m \tau)\left|P_a\right|_{ln,m}} \quad (14)$$

Where, $P_{aln,\,m}$ (Pa) is the log-mean air pressure across the membrane which can be calculated by following formula.

$$\left|P_a\right|_{ln} = \frac{Pfm - Ppm}{\ln\frac{Pfm}{Ppm}}$$

4) The heat transfer coefficients, h_f and h_c, are calculated by using dittus boelter equation for the laminar flow of feed (150 < Re < 3500) [121-123, 126]

$$Nu = 1.86\,(Re\,Pr\,\frac{d_h}{L})^{0.33} \quad (15)$$

For the turbulent flow of feed (2500 < Re < 1.25 x 10^5)

$$Nu = 0.023\,Re^{0.8}\,Pr^{1/3}\,(\frac{\mu}{\mu_w})^{0.14} \quad (16)$$

Where Nu, Re & Pr are calculated by below equations & largly depend on fluid properties at temperatures (Tfm & Tpm)

$$Nu = \frac{h\,d_h}{K_l} \quad (17)$$

$$Re = \frac{\rho V d_h}{\mu} \quad (18)$$

$$Pr = \frac{Cp\,\mu}{K_l} \quad (19)$$

5) From Heat Analysis equations and at steady state conditions, Qf = Qm+v = Qp, [124-125] So, from equations (4-8) on simplification we get the value of T_{mf} and T_{mp}

$$T_{mf} = \frac{\frac{k_m}{\delta_m}\left(T_{bp} - \frac{h_f}{h_p}T_{bf}\right) + T_{bf}h_f - NHv}{\frac{k_m}{\delta_m} + h_f\left(1 + \frac{k_m}{\delta_m h_p}\right)} \quad (20)$$

$$T_{mp} = \frac{\frac{k_m}{\delta_m}\left(T_{bf} - \frac{h_p}{h_f}T_{bp}\right) + T_{bp}h_p - NHv}{\frac{k_m}{\delta_m} + h_p\left(1 + \frac{k_m}{\delta_m h_f}\right)} \quad (21)$$

6) The obtained temperatures, Tfm and Tpm, are compared with the initial assumed temperatures in step 1.

The above procedure is repeated till the %difference between assumed temperature and calculated temperature is less than 0.1.

15. APPLICATIONS OF MEMBRANE DISTILLATION

Although The MD process is currently applied mostly only at the laboratory scale, there are several pilot plants currently under trial and testing The MD applications are very suitable for environmental, chemical, petrochemical, food, pharmaceutical and biotechnology, Biomedical and cosmetic industries. Attempts are being made in pilot plant applications which have been proposed for desalination and nuclear desalination but still they are under experimental tests and their results are yet not clear and hence its use is not fully extended [129-136].

MD is extremely is useful for some biomedical applications such as water removal from blood and treatment of protein solutions [1,142,159-167].It was proved that DCMD is feasible to process low and medium-level radioactive wastes by nuclear desalination [131,137-138].

MD has been tested successfully for the treatment of pharmaceutical wastewater containing taurine, textile wastewater contaminated with some special types of dyes such as methylene blue, aqueous solutions.

Recently study is being carried out using MD for treatment of water contaminated with boron, arsenic, and some heavy metals, ammonia, concentration of coolant liquid such as glycols, The chemical stability of the employed membranes is very high so MD is being used for Humic acid and acid solutions such as HCL and H2SO4 rich in specific compounds, oil-water emulsions, olive oil mill wastewater for polyphenols recovery and radioactive wastewater solutions [154-156].

MD can be applied for breaking azeotropic mixtures which have close boiling points was proposed and tested for the separation of acidic solutions such as hydrochloric acid/water, propionic acid/water and formic acid/water azeotrope mixtures [140-141], for the extraction of volatile organic compounds from dilute aqueous solutions and the concentration of aqueous solutions containing sodium hydroxide, strong mineral acid and sulfuric acid at various pH values [142].

MD is attractive in biotechnology field as well, for the removal of toxic products from culture broths.MD can used for temperature sensitive applications which lead to degradation of the process fluids at high temperature especially in food processing industries and concentration of milk and must [142].Recovery of volatile aromatic compounds from black currant juice [139] can be made with the help of MD. MD has found attractive for concentration of many types of juices such as orange juice, mandarin juice, apple juice, sugarcane juice which are taste sensitive for high temperature.[1,143,144–147,148–158,167-170]

16. CONCLUSIONS AND FUTURE PROSPECTS

As a promising alternative technology to replace other established separation processes, MD has gained much interest for its lower energy requirement and use of solar or geothermal energy, lower operating pressures and higher rejection factors compared to other pressure driven processes.

Still there some issues to be sorted out such as

1) Lack of commercially available MD units;.
2) Novel membranes specifically designed for MD applications should be fabricated in an economically feasible way.
3) Research on trans-membrane flux enhancement for large scale applications is required.
4) More focus on integration of one type of MD process with other type of MD Processes as well as conventional distillation techniques is necessary for improvement in the efficiency of the system and to make the process economical for industrial applications.

The ability to effectively operate at low temperature makes MD process promising for utilization of lower grade waste or alternative energy sources. In recent years, Emphasis is given on MD because there is great scope for coupling MD with solar, geothermal and waste energy systems to decrease energy consumption in desalination systems.

The DCMD theoretical mathematical modeling for a flat sheet membrane module is developed from the fundamental equations of mass and heat transfer. The Knudsen number is used as a guideline for identifying mechanism of diffusion. Out of 04 mechanisms for mass transfer, transition mechanism is fitted. The Prediction of permeate flux can be done by using this model with the help of developed MATLAB codes and results can be further compared with experimental process results.

17. NOMENCLATURE

A= Cross sectional area (m^2)
Cb= Permeability (mol/mSPa)
Cp= Specific heat capacity at constant pressure (J/(K kg)
CPC= Concentration polarization coefficient (dimensionless number)
dp= Pore size (μm)
dh= Hydraulic diameter (m)
Dw= Diffusion coefficient (m^2/s)
h= Heat transfer coefficient (W/m^2K)

H_v= Heat of vaporisation (kJ/kg)
Kn= Knudsen number (dimensionless number)
kl= Process Fluid thermal conductivity at specific temperature (W/mK]
km= Membrane thermal conductivity (W/mK)
ks = Thermal conductivity of the membrane material (W/mK)
kg= Thermal conductivity of gas filling the pores (W/mK)
L= Length of module (m)
N= Permeate flux (kg/m^{2}hr)
Nu = Nusselt Number (dimensionless number)
P = Total pressure (Pa)
Pfm= vapour pressure of feed (Pa)
Ppm= vapour pressure of permeate (Pa)
Plm= Logarithmic mean Pressure (Pa)
Pr = Prandtl Number (dimensionless number)
Qf= Feed heat flux (W/m^2)
Qv= Heat flux through latent heat transfer (W/m^2)
Qm= Heat flux through Conduction heat transfer (W/m^2)
Qp= Product heat flux (W/m^2)
R= Gas constant (J/Kmol)
Re= Reynolds number (dimensionless number)
T_{fm}= membrane surface temperatures at feed side (K)
T_{pm}= membrane surface temperatures at permeate side (K)
T_{fb}= fluid bulk temperatures at the feed and permeate side (K)
T_{pb}= fluid bulk temperatures at the permeate side (K)
T= Absolute temperature (K)
TPC= Temperature polarization coefficient (dimensionless number)
v = velocity of the fluid flow (m/s)

17.1 Subscripts and Superscripts used:

f= Feed
p= Permeate
m= Membrane
b= Bulk
mf= Feed side of membrane
mp= Coolant side of membrane
bf = Bulk feed
bp= Bulk permeate
c = Coolant side
h= Hot region
MD= Reference MD system

17.2 Greek Letters:

δ= Membrane thickness; film thickness (μm)
ε= Porosity in % (dimensionless number)
ρ= Density (kg/m3)
σ= Collision diameter of the molecule (m)
τ= Membrane tortuosity (dimensionless number)
θ= Membrane/liquid contact angle (° or rad)
λ= Mean free path of vapor (m)
μ= Viscosity of vapor (Pa s) or chemical potential (J/kg)

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