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Membrane Module Design and Dynamic Shear-Induced Techniques to Enhance Liquid Separation by Hollow Fiber Modules: A Review

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Abstract

Membrane-based separation processes have found numerous applications in various industries over the past decades. However, higher energy consumption, lower productivity and shorter membrane lifespan due to polarization and membrane fouling continue to present severe technical challenges to membrane-based separation. Improved membrane module design and novel hydrodynamics offer strategies to address these challenges.

This review focuses on hollow fiber membrane modules which are well-suited to membrane contactor separation processes. Attempts to improve membrane module design should begin with a better understanding of the mass transfer in the hollow fiber module, therefore this review provides a summary of prior studies on the mass transfer models related to both the shell-side and tube-side fluid dynamics. Based on the mass transfer analysis, two types of technique to enhance hollow fiber membrane module performance are discussed: (1) passive enhancement techniques that involve the design and fabrication of effective modules with optimized flow geometry; or (2) active enhancement techniques that uses external energy to induce a high shear regime to suppress the undesirable fouling and concentration polarization phenomena. This review covers the progress over the past five years on the most commonly proposed techniques such as bubbling, vibrations and ultrasound.

Both enhancement modes have their advantages and drawbacks. Generally, the passive enhancement techniques offer modest improvement of the system performance, while the active techniques, including bubbling, vibrating and ultrasound, are capable of providing as high as 3–15 times enhancement of the permeation flux. Fundamentally, the objectives of module design should include the minimization of the cost per amount of mass transferred (energy consumption

and module production cost) and the maximization of the system performance through optimizing the flow geometry and operating conditions of the module, scale-up potential and expansion of niche applications. It is expected that this review can provide inspiration for novel module development.

Keywords: membrane module design, passive and active enhancement modes, mass transfer, hydrodynamics, energy efficiency

1. Introduction

Membrane-based separation processes have found many applications in fields such as water, energy, chemical, petro-chemical and pharmaceutical industries. This growth has been primarily due to two developments: firstly, the ability to produce high permeability and essentially defect-free membranes on a large scale; and secondly, the ability to assemble these membranes into compact, efficient and economical membrane modules with a high membrane surface area [1-3].

Nevertheless, there are still several limitations hindering the application of membrane-based processes, including flux decline, concentration polarization and membrane fouling. These limitations can reduce productivity, increase energy consumption and shorten membrane lifespan. A sustainable flux depends not only on membrane permeation properties, but also on the fluid hydrodynamics within the membrane module. In recent decades, numerous attempts have been made to design and fabricate effective membrane modules with optimized geometries and/or shear-induced accessories to enhance permeation and suppress undesirable polarization and membrane fouling [4-9].

The performance improvement methods can be classified into two categories: passive enhancement techniques and active enhancement techniques. The passive techniques include modifying membrane layout or introducing spacers or baffles into the membrane modules to alter the flow geometries, by inducing secondary flows or eddies adjacent to the membrane or/and creating significant flow instabilities. The active techniques utilize external energy to enhance the relative motion between the fluid and the membrane. The induced high shear rate can facilitate mixing and reduce the thickness of the boundary layer on the membrane surface.

There is considerable evidence that properly designed and fabricated membrane modules can improve the fluid hydrodynamic conditions and enhance overall system performance dramatically. However, despite its importance, membrane module design and fabrication have received less attention than membrane materials and membrane process development. The literature in this field is relatively sparse in comparison with the rapidly increased amount of literature in other membrane-related areas. The main reason is probably due to the fact that module technology has been developed commercially in the form of patents which are treated as proprietary knowledge by industry.

This paper starts by summarizing the basic types of membrane module used for aqueous separations with a focus on hollow fiber modules and related mass transfer models. Then we discuss passive process enhancement techniques involving module/fiber configuration designs and active process enhancement techniques involving shear enhanced aids (vibrations/oscillation, bubbles and ultrasound etc) [10-12]. The focus is given to the latest developments in hollow fiber module design concepts and principles of mass transfer enhancement, because hollow fiber membrane technology is an attractive platform for many engineering processes. Moreover, by analyzing the working principle of each enhancement mode for practical applications, their benefits, limitations and technical requirements are addressed in terms of economic considerations (fabrication cost and complexity, energy demand) and processing engineering (scale-up potential and niche applications). It is hoped that this review can provide insights and inspire novel module design to enhance system performance of membrane-based processes for liquid separations.

2. Development of membrane modules for liquid separation

Industrial membrane separation requires large areas of membrane surface to be economically and effectively packaged. These packages are called membrane modules. Effective module design is one of the critical achievements that has led to the commercialization of membrane-based separation units [2].

Generally, there are four basic types of module: plate-and-frame, spiral wound, tubular and hollow fiber modules. The earliest module designs were based on simple filters and consisted of flat sheets of membranes confined in a filter press called “plate-and-frame” modules. Due to its simplicity, these plate-and-frame modules have been widely used in lab-scale and industrial applications. Although each type of membrane module configurations has its own pros and cons, hollow fiber modules have received the most attention because of their unique characteristics of self-support, high membrane packing density and high contact surface to volume ratio. The surface to volume ratio (m^2/m^3) is typically 350–500 for plate-and-frame modules, and 650–800 for spiral wound modules. In contrast, hollow fiber membrane modules may have the ratio as high as 7000–13000. In addition to this, hollow fibers have the greatest potential to be arrayed in different forms for various applications [13]. The most common form is the conventional axially-parallel fiber arrangement, as shown in Fig. 1.

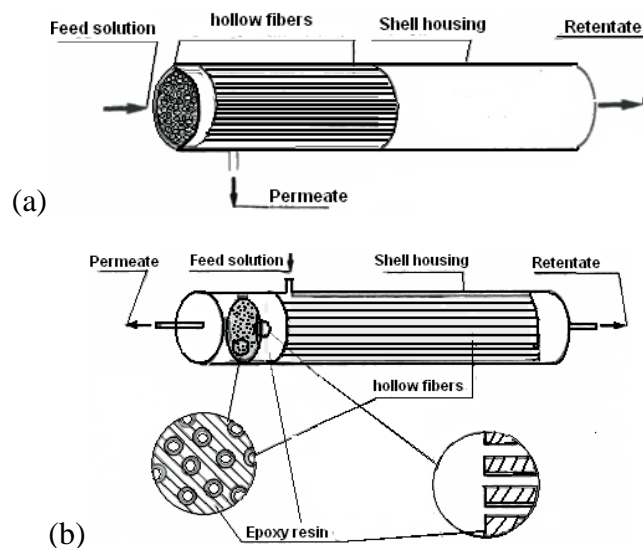


Fig. 1. A conventional parallel flow hollow fiber module

(a) tube-feed; (b) shell-feed with dead end (redrawn from [13])

Hollow fiber modules typically operate using one of two flow patterns: tube-side (or lumen-side) feeding or shell-side feeding. The former is commonly used in biotechnology applications and the latter for water applications. In some cases, such as membrane contactors, both tube and shell sides require controlled flows. Hydrodynamic challenges with the shell-side flow pattern of hollow fiber membrane modules include: bypassing, channeling and dead zones, which result in a loss in separation efficiency. While channeling may not be apparent in small scale bench tests, it becomes a serious concern in full scale applications [13]. This concern has led to major efforts in improving hydrodynamic conditions to overcome this problem, which is discussed in detail in Section 4.1 of this paper.

3. Mass transfer analysis for hollow fiber module design

Fundamentally, in all membrane separation processes, a molecule or particle is transported across a membrane due to a force acting on it, when this driving force is kept constant, a constant

flow will occur through the membrane after the establishment of a steady state. For a general liquid-liquid membrane separation process, the overall flux J_s of the solute to be removed or retained can be expressed by a proportionality relationship [14]:

$$J_s = k\Delta F \quad (1)$$

where ΔF is the overall driving force of the process, and the proportionality factor k is the overall mass transfer coefficient, which determines how fast the component is transported through the membrane, or in other words, k is a measure of the resistance exerted by the whole transport process.

In a hollow fiber module, the transport of this component will follow three basic steps: from bulk feed to the membrane surface, across membrane and from the other surface of the membrane to the bulk permeate. By assuming the feed is flowing through the shell-side, k can be expressed based on the resistance-in-series model [15]:

$$\frac{1}{k} = \frac{1}{k_m} + \frac{1}{k_{tube}} + \frac{1}{k_{shell}} \left(\frac{d_{t\ in}}{d_{t\ out}} \right) \quad (2)$$

where k_m is the membrane mass transfer coefficient; k_{shell} , the mass transfer coefficient through the boundary layer on the feed side (most commonly used correlations are shown in Table 1) and k_{tube} , the mass transfer coefficient through the boundary layer in the permeate side; $d_{t\ in}$ and $d_{t\ out}$ are the inner and outer diameters of the fiber, respectively. All mass transfer coefficients here are calculated based on the inner membrane surface of the fiber. It should be noted that these theories which are involved in solute transport are not applicable to some special processes (e.g. membrane distillation (MD)) whose component of interest is the solvent itself (water).

With rapid advancement of membrane science, currently available membranes used in many applications are so effective that the separation process is limited mainly by the mass transfer rate to the bulk-membrane interface rather than through the membrane itself [7]. To further interpret the mass transfer occurring in fluids, it is conventional to correlate parameters in a dimensionless form, such as the Graetz number (Gz), Schmidt number (Sc) and Sherwood number (Sh). Gz is a dimensionless duct length, which can be expressed as the product of three dimensionless groups, as shown in Eq. (3). [16]

$$Gz = Re \, Sc \, \frac{d_h}{L} \quad (3)$$

where Re is the Reynolds number, d_h is the hydraulic diameter of the flowing channels in the shell-side, L is the effective length of the module. Sh is the most common term by which mass transfer is described. It is defined as the ratio of convection to diffusion and is dependent on the shape of the duct and its dimension, as indicated by Eq. (4)

$$Sh = \frac{k \, d_h}{D} = f(Gz) \quad (4)$$

where D is the diffusion coefficient of the solute in the feed side solution, Generally the mass transfer correlations can be expressed as [17]:

$$Sh = a Re^\alpha Sc^\beta \left(\frac{d_h}{L}\right)^\psi \quad (5)$$

where a is a function of module geometry, α , β and ψ are constants determined experimentally. Sh can be viewed as the ratio of the characteristic dimension of the flow path to the boundary layer thickness on the membrane surface. In laminar flow, some applicable correlations contain an additional factor involving the characteristic dimension divided by the length of the flow path (d_h/L) [16].

Among various membrane processes involving liquid separation using membranes, for pressure-driven systems (e.g. reverse osmosis (RO), microfiltration (MF) & ultrafiltration (UF)), only the feed side (shell-side feeding pattern is assumed) may be subject to concentration polarization, which describes the phenomena of concentration build-up within the boundary layer near the membrane surface, due to the poor hydrodynamics and hence the low mass transfer coefficient [7]; while for most concentration-driven systems (e.g. membrane contactors), both tube and shell-side flows may have great impact on the overall module performance (e.g. artificial kidney, blood oxygenator, membrane distillation processes, etc) [18]. In some cases, even the membranes may play an important role in the overall mass transfer resistance [19]. Therefore, to design a well-performed hollow fiber module requires not only a better understanding of fluid dynamics on the shell-side, but also the flow on the tube-side and the mass transfer resistance across the membrane. A comprehensive summary of prior development of mass transfer models are provided in the following section.

3.1 Mass transfer in shell-side

Although many studies have focused on either empirical or fundamental approaches to describe the shell-side mass transfer coefficient in conventional cross-flow hollow fiber modules [20-24], none of them are presented in a general form which can be applied to all membrane processes involved liquid phases. Most of the studies on mass transfer inside hollow fiber modules are based on membrane contactors, and the blood oxygenator and CO₂ contactor are the most adopted processes to study the shell-side fluid behavior due to their simplicity. However, more and more rigorous engineering approaches have been developed by many researchers in recent years, for example, a generalized correlation using the analogy between heat and mass

transfer was proposed by Lipnizki and Field [21]. This approach covers a wide range of packing densities, the effect of flow mal-distribution, both laminar and turbulent flow, the entrance effects, and the development of both the hydraulic and concentration profiles. It can be interpreted as:

1. Laminar flow ($Re < 2300$)

(i) Both hydrodynamic and concentration profiles are fully developed,

$$Sh_1 = 3.66 + 1.2\phi^{-0.4} \quad (6)$$

where ϕ is the packing density.

(ii) A developing concentration profile with full hydrodynamic development, and the entrance effect is taken into account:

$$Sh_2 = 1.615(1 + 0.14\phi^{-0.25}) \left(\frac{Re Sc d_h}{L} \right)^{0.33} \quad (7)$$

(iii) Both profiles are developing, the entrance effects are considered:

$$Sh_3 = \left(\frac{2}{1 + 22Sc} \right)^{1/6} \left(\frac{Re Sc d_h}{L} \right)^{0.5} \quad (8)$$

if there is a need to include entrance effects even when the fluid leaves the module with fully developed profiles. Sh_1 and Sh_2 can be combined to predict the overall average mass transfer coefficient:

$$Sh = (Sh_1^3 + Sh_2^3)^{1/3} \quad (9)$$

If the fluid leaves the module with fully developed hydrodynamic profile and developing concentration profile, and entrance regions should be included, the overall average Sh can be expressed as:

$$Sh = (Sh_2^3 + Sh_3^3)^{1/3} \quad (10)$$

Similarly, if the complete range of profile developments is considered, Sh for the whole range can be calculated by:

$$Sh = (Sh_1^3 + Sh_2^3 + Sh_3^3)^{1/3} \quad (11)$$

2. Turbulent flow ($2300 < Re \leq 10^6$)

The mass transfer correlation can be derived based on a heat transfer analogy for flow through an annulus by Stephan [25], with $Sc \gg 0.0454$:

$$Sh = 0.021 \phi^{-0.225} Re^{0.8} Sc^{0.33} \quad (12)$$

As mentioned above, most of the empirical correlations developed by different researchers are based on specific studies of various systems and operating conditions, and most importantly, some influential factors such as the entrance effects, the development state of hydrodynamic and concentration profiles, the impact of packing density and mal-distribution phenomenon, and fiber polydispersity are neglected. To make the model more comprehensive, Lipnizki and Field [21] incorporated the effect of packing density and mal-distribution into the hydraulic diameter, divided the hollow fiber module into segments and proposed the prediction of average Sh via a sum of the local Sh_k :

$$Sh_{av} = \frac{1}{A} \sum_{k=1}^n A_k Sh_k(Re_k, \phi_k) \quad (13)$$

where k refers to the segment. However, the fiber polydispersity is not considered in this model. Hence, there are several new correlations developed by other authors that include random packing density [23, 26, 27].

As an important variation developed based on the conventional cross-flow modules, transverse-flow hollow fiber module has been intensively reported to have larger mass transfer coefficient, minimal flow channeling and better scale-up characteristics. For more precise performance prediction, several correlations have been proposed to describe its shell-side mass transfer [20]. To give an example, one of the shell-side mass transfer correlations has been developed based on the free surface model [28], which agrees well with the experimental results of the best-known Liqui-Cel[®]Extra-Flow module:

$$Sh = 2.15Re^{0.42}Sc^{0.33} \quad (14)$$

Here Re varies from 0.8 to 20. The detail of Liqui-Cel[®]Extra-Flow module is discussed in Section 4.1.2.

An overview of the historical development of mass transfer correlations, is summarized in Table 1, which contains some popular models developed by various researchers in recently years. Although there is already a comprehensive review on hollow fiber membrane contactors by Gabelman and Hwang in 1999 [20], this paper focuses mainly on the developments since 2000. In addition, regardless of the increasingly comprehensive models that have been developed, it should be noted that there is still no universal form which can be applied due to the complexity of coupling factors. However, a relatively rigorous approach is still feasible to analyze the hollow fiber module performance and hence help to identify the bottlenecks of module design in terms of process engineering.

3.2 Mass transfer in the tube-side

For some membrane processes dealing with liquid phases, both shell-side and tube-side flows have major contributions to the overall mass transfer, such as membrane contactors. In fact, the

flow is usually laminar instead of turbulent in the hollow fibers because of the small fiber diameter and comparatively long length. Any turbulent flow will eventually be reduced to laminar flow after passing a certain length, due to friction with the membrane wall [29].

Therefore the fluid flowing in the tube-side is generally treated as laminar flow, and the individual mass transfer coefficient k_{tube} is dependent on the flow velocity. Though there are several correlations available for the tube-side flow calculation [6, 30], the L  v  que solution ($Gz > 4$) [31] has been widely accepted in the literature to predict k_{tube} with a reasonable degree of accuracy:

$$Sh = \frac{k_{tube} d_{t\ in}}{D_t} = 1.62 Re^{0.33} Sc^{0.33} \left(\frac{d_{t\ in}}{L} \right)^{0.33} \quad (15)$$

$$k_{tube} = 1.62 \left(\frac{D_t^2 v}{L d_{t\ in}} \right)^{0.33} \quad (16)$$

where D_t is the diffusion coefficient of the solute in the tube side solution. However, Eq (15) always overestimates the tube-side mass transfer coefficients when $Gz < 4$. To develop a more rigorous correlation for hollow fiber systems, Wickramasinghe et al [6] incorporated the polydispersity of hollow fiber diameters to calculate the average. Their commonly used correlations for the tube-side mass transfer are also summarized in Table 1.

3.3 Mass transfer across the membrane

As mentioned previously, sometimes the membrane itself may present as the major resistance in the overall mass transfer, especially in some membrane contactor processes. Here, the local mass transfer coefficient k_m can be defined as [32]:

$$k_m = \frac{D\varepsilon}{\tau\delta_m} \quad (17)$$

where D is the diffusion coefficient of the solute through the membrane, which can be calculated by applying the Wilke and Chang method [33]; ε is the membrane porosity, δ_m is the thickness of membrane wall and τ is tortuosity. Thus, k_m is merely depending upon the solute diffusivity and the membrane structure regardless of the operating parameters (It is noted that this solute transport mechanism across the membrane is not applicable in the MD process because it involves only water vapor transport).

Table 1. Correlations for shell/tube-side mass transfer in hollow fiber modules

| Correlation | Eq no. | Operating Conditions | Packing density (%) | Remarks | Ref |
|--|--------|----------------------|---------------------|---|----------|
| Shell-side axial flow | | | | | |
| $Sh = 0.048Re^{0.6}Sc^{0.33}\left(\frac{d_h}{d_{t\ out}}\right)$ | (18) | -- | -- | From Toyobo's RO module, its flow condition is not clear | [24, 34] |
| $Sh = \beta(1-\phi)Re^{0.6}Sc^{0.33}\left(\frac{d_h}{L}\right)$ | (19) | $Re > 500$ | 0.4-40 | β is 5.8 for hydrophobic and 6.1 for hydrophilic membranes | [35] |
| $Sh = (0.53 - 0.58\phi)Re^{0.53}Sc^{0.33}$ | (20) | $Re = 20-350$ | 32-76 | Remixing and splitting of fluid is considered, fresh fluid constantly presents on the membrane surface is assumed | [36] |
| $Sh = 1.25Re^{0.93}Sc^{0.33}\left(\frac{d_h}{L}\right)^{0.93}$ | (21) | $Re < 1000$ | 2.5, 26 | -- | [30] |
| $Sh = 0.019ReSc\frac{d_h}{L}$ | (22) | $Gz < 60$ | | Closely packed modules of various geometries | [6] |
| $Sh = 8.8\left(Re\frac{d_h}{L}\right)Sc^{0.33}$ | (23) | Laminar | 15 | Channeling needed to be incorporated | [37] |
| $Sh = 8.71Re^{0.74}Sc^{0.33}\left(\frac{d_h}{L}\right)$ | (24) | $Re = 0.16-7.30$ | 30 | -- | [38] |
| $Sh = 0.09(1-\phi)Re^{(0.8-0.16\phi)}Sc^{0.33}$ | (25) | $Re \leq 10$ | 35-97 | For regularly packed fibers cases. | [39] |

| | | | | | |
|---|------|--|-----------|---|------|
| $Sh = (0.31\phi^2 - 0.34\phi + 0.10)Re^{0.9}Sc^{0.33}$ | (26) | $Re = 32-1287$ | 8-70 | Flow mal-distribution is taken into account. | [40] |
| $Sh_k = [4.212Gz_k(1 + 0.14\phi_k^{-0.25})^3 + 0.302Gz_k^{1.5}Sc_k^{-0.5}]^{1/3}$ | (27) | $Re < 2300$ | 10-75 | Entrance effects, packing density and flow mal-distribution are taken into consideration a hollow fiber module with fully .developed hydrodynamic and developing concentration boundary layer profiles. where Sh_k and Sh_{ov} are the local and overall average Sherwood number, respectively. | [21] |
| $Sh_{av} = \frac{1}{A} \sum_{k=1}^n A_k Sh_k(Re_k, \phi_k)$ | (13) | $Sc \gg 1$ | | | |
| $Sh = 0.021 \phi^{-0.225} Re^{0.8} Sc^{0.33}$ | (12) | $2300 < Re \leq 10^6$ $Sc \gg 1/22$ | 10-75 | Derived based on a heat transfer for flow through an annulus by Stephan | [25] |
| $Sh = (-0.4575\phi^2 + 0.3993\phi - 0.0475) \times Re^{(4.0108\phi^2 - 4.4296\phi + 1.5585)} Sc^{0.33}$ | (28) | Laminar | 30.6-61.2 | Based on osmotic distillation systems, Re is a function of packing density | [22] |
| $\langle Sh \rangle_r = \beta \frac{1}{(0.86 - 0.3\phi_0)\phi_0} (ReSc \frac{d_h}{L})^{(0.3\phi_0 + 0.14)}$ | (29) | $Re = 68-1194$ $Gz = 70-5039$ | 20-50 | The overall average mass transfer coefficient $\langle Sh \rangle_r$ incorporated the randomcity of fiber/flow distribution, β is a dimensionless group presenting the deviation of randomly packed module from uniformly packed one. | [26] |

| | | | | | |
|---|------|---------------|-------|---|------|
| $Sh = \frac{d_h}{D} \frac{\int_0^\infty \int_0^\infty \phi v f(\phi) g(r) d\phi dr}{2\pi l \int_0^\infty r g(r) dr}$ $\times \ln \frac{\int_0^\infty \int_0^\infty \phi v f(\phi) g(r) d\phi dr}{\int_0^\infty \int_0^\infty e^{-2k\pi r l / (\phi v)} \phi v f(\phi) g(r) d\phi dr}$ | (30) | -- | 25-75 | For the first time both randomness of flow distribution and polydispersity of fiber diameter on shell-side mass transfer are considered together. | [23] |
| Shell-side transverse flow | | | | | |
| $Sh = 0.575 Re^{0.556} Sc^{0.33}$ | (31) | $Re < 1000$ | -- | The analogy of a well-established heat transfer correlation for flow across or transverse to a “staggered bank” of tubes. | [41] |
| $Sh = 0.15 Re^{0.8} Sc^{0.33}$ | (32) | $Re > 2.5$ | -- | Developed by alternative module geometries, such as cylindrical/helically wound bundles and rectangular-bed configuration. | [6] |
| $Sh = 0.12 Re Sc^{0.33}$ | (33) | $Re < 2.5$ | -- | Obtained by the similar configurations with Eq (32) under conditions which may induce uneven flow channels among fibers. | [6] |
| $Sh = 1.38 Re^{0.34} Sc^{0.33}$ | (34) | $1 < Re < 25$ | 70 | Developed from tightly packed module for O ₂ or CO ₂ removal, it was based on heat transfer correlations of single tubes. | [30] |
| $Sh = 0.9 Re^{0.4} Sc^{0.33}$ | (35) | $1 < Re < 25$ | 7 | Similar to Eq (34), for loosely packed modules. | [30] |

| | | | | | |
|---|------|-------------------------------------|-----|---|----------|
| $Sh = 0.61Re^{0.363}Sc^{0.33}$ | (36) | $0.6 < Re < 49$ | 0.3 | For extremely low packing density cases. | [42] |
| $Sh = 1.45Re^{0.32}Sc^{0.33}$ | (37) | -- | -- | Obtained from bubble-free aeration of water using transverse flow fiber arrangement. | [43] |
| $Sh = 0.24(\frac{d_h}{L}Re)^{0.59}Sc^{0.33}$ | (38) | -- | -- | Similar to Eq (37), but used a sealed fiber bundle unconfined in a jet stream instead. | [44] |
| $Sh = 2.15Re^{0.42}Sc^{0.33}$ | (14) | $0.8 < Re < 20$ | -- | Developed based on free surface model, which agrees well with the experimental results of the best-known Liqui-Cel®Extra-Flow module | [28] |
| Tube-side mass transfer | | | | | |
| $Sh = 1.62Re^{0.33}Sc^{0.33}(\frac{d_{t\ in}}{L})^{0.33}$ | (15) | $Gz > 4$ | | Reasonably accuracy is obtained for mass transfer coefficients estimation when $Gz > 4$ cases, but overestimates when $Gz < 4$. | [31] |
| $\langle Sh \rangle = Sh[1 - (18Sh/Gz + 7)x_0 + \dots]$ | (39) | -- | | Polydispersity of hollow fiber diameters is incorporated into calculating the average $\langle Sh \rangle$, Sh is for a uniform distribution of fiber radii, x_0 represents the deviation divided by the mean. | [6] |
| $Sh = 0.023Re^{0.8}Sc^{0.33}$ | (40) | $Re > 2000$ | | Based on Chilton-Colburn and Deissler analogies. | [45, 46] |
| $Sh^* \propto \left(\frac{f'}{f}\right)^m Re^{(1-0.25m)} Sc^{0.33} \left(\frac{Sc}{Sc_w}\right)^{0.11}$ | (41) | $10^4 < Re < 10^5$, $Sc > 1000$ | -- | Sh^* , the corrected Sherwood number under conditions of porosity and variable properties, Sc_w is the corrected Schmidt number on the membrane wall; f is the friction factor and f' | [17] |

is the corrected friction factor, $m=0.5$ or 1.0
depends on smooth or porous/rough surface.
Applicable for Newtonian flow

Note:

1. This table contains most of the correlations developed after 1999; some earlier models were reviewed by Gabelman and Hwang [20].
2. Only applications for liquid separation are presented, i.e. gas separation such as adsorption is not included.
3. No chemical reaction is involved in these cases. Some special transverse flow correlations derived from hollow fiber fabric modules are not presented in this table, they will be given below in the case study.

3.4 Basic principles for mass transfer enhancement

The above discussions clearly indicate that the mass transfer in a hollow fiber module is closely linked to the fluid hydrodynamics and membrane module geometry. Using the membrane contactor as an example, while the mass transfer through the membrane (k_m) is independent of the flow conditions, the mass transfer on shell and tube sides (k_{shell} and k_{tube}) are functions of the flow conditions and fiber/module geometries. The semi-empirical mass transfer correlations shed some light on strategies to improve the mass transfer by varying flow conditions and flow channel design.

On the tube side, Eq (15) is widely used to predict the mass transfer coefficients, where Re represents the hydrodynamic conditions. However, the predictions by this model slightly overestimate the experimental data when the flow velocity is very low [20], which may be due to the non-uniform flow distribution inside the tube. It was found that it is not only related to the flow velocity (via Re), but may also relate to the effect of fiber length and fiber dimensions. As a certain degree of uniformity is reached, the mass transfer coefficient k_{tube} can be predicted reliably. It increases with increasing Re and the diffusivity of the solute of interest [20], but decreases with increasing inner diameter and fiber length. Under given conditions, Re seems to be the dominant factor affecting k_{tube} .

On the other hand, the prediction of the shell-side mass transfer coefficient Sh_{shell} is more challenging, since the shell-side geometry and hydrodynamics are more complicated to correlate. Though there are numerous studies that focus on the shell-side, none are universally applicable due to the various parameters incorporated in the different models. However, the basic principle of mass transfer enhancement shown in these correlations is similar. According to the

increasingly complex form of the model development, it can be concluded that the mass transfer depends on many factors and their combinations, such as the flow velocity (Re), states of hydrodynamic/concentration profiles, hydraulic shell diameter and effective length of the module, entrance effects, fiber polydispersity, packing density, and flow mal-distribution. Furthermore, it may also be influenced by the interaction between the surface properties of the membrane (i.e. hydrophobic/hydrophilic character) and the diffusivity of the solute of interest, which is playing a role in calculating k value [20]. For example, hydrophilic membranes may facilitate the transport of inorganic solutes, while hydrophobic membranes may transport the organic solutes preferentially [19, 20].

Clearly, the main objective of improved membrane module design is to enhance the overall mass transfer. The basic strategies include enhancing the module's capabilities to create more eddies or turbulence between fibers, reduce the boundary layer thickness and provide better mixing. To achieve these goals, various methods and devices have been employed to enhance the mass transfer inside the module (e.g. the passive enhancement techniques, and active enhancement techniques). These strategies are reviewed in the following sections (refer to Section 4.1 and Section 4.2).

4. Process enhancement techniques

4.1 Passive enhancement techniques

The majority of laboratory or industrial scale modules are designed for use with flat sheet membranes, because the membrane structure is simple and the membrane replacement is easy. From a commercial standpoint, however, hollow fiber modules are more productive as they have

much larger surface area per volume. Despite the relatively high fabrication cost, hollow fiber modules can play an important role and gain better performance to minimize the cost per unit product volume [47-49].

Most hollow fiber modules are designed for pressure-driven filtration processes rather than concentration-driven or thermally-driven contactor processes. However, from the process enhancement point of view, their applications may be potentially extended to suit and improve other separation processes.

4.1.1 Fabric hollow fiber modules

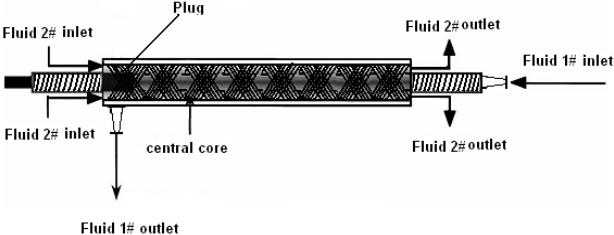
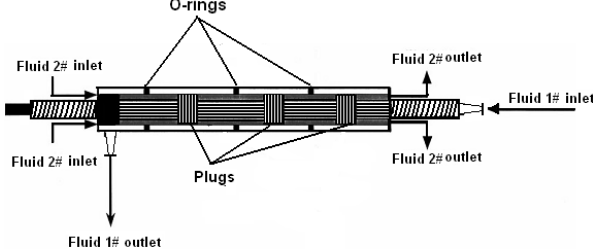
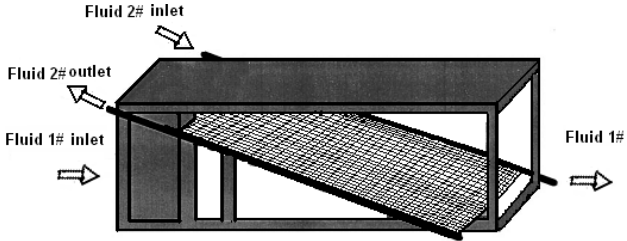
In the early days, due to limited materials and fabrication methods, membranes themselves tended to be the controlling resistance in membrane-based separations. With the advancement of membrane fabrication techniques, it has been possible to produce thinner membranes with higher permeability. As a result, improving mass transfer of the process has shifted to alternative geometries that are able to offer better performance than the conventional parallel flows.

It is widely reported that flow mal-distribution in the membrane modules may lead to decrease module performance and hence a reduction of the average mass transfer coefficient [50]. To overcome the problems of non-uniform fiber spacing in hollow fiber modules, which often results in a flow mal-distribution, several researchers have introduced fiber-woven fabric into hollow fiber modules to gain more uniform spacing and baffles to create better mixing [47-49]. The results showed that the shell-side mass transfer coefficient was significantly higher than that of the commercial parallel modules. In order to make a comparison, they designed and tested

various configurations (Figs.2-6). Their detailed features can be found in an earlier review by Gabelman and Hwang [20]. In this paper, only a brief summary is given in Table 2.

To provide some perspectives on hollow fiber module design, some researchers [48, 49] correlated Re (flow velocity) and Sh (mass transfer coefficient) with single-fiber modules under different flow regimes and developed a set of analogous shell-side mass transfer correlations which showed good agreement with the experimental data (some of these correlations are listed in Table 1). According to their observations, counter-current flow patterns had much better mass transfer. Surprisingly, the baffled rectangular module performed more poorly than non-baffled modules and a cylindrical module with fewer baffles was comparable to a fully-baffled one. Therefore, it can be seen that baffles can constrain the hydrodynamic conditions in some cases. These previous studies suggest that turbulence promoters do not always enhance module efficiency. The effectiveness depends on how the promoters are arranged and how the flow channels are actually distributed. However, most of the mentioned configurations improve the fluid distribution and mixing to achieve much higher mass transfer coefficients in both gas and liquid separations [48, 49].

Table 2. Summary of fabric hollow module configurations

| Module type | Key characteristics & performance | Figures |
|-----------------------------------|--|--|
| Axial helically wound module [49] | <ul style="list-style-type: none"> • All fibers wound helically around a central core; • The Feed was forced radially outward by a plug for perpendicular flow distribution. |  <p>Fig. 2 Axial helically wound module</p> |
| Fabric woven module [49] | <ul style="list-style-type: none"> • Woven hollow fiber fabric wound helically around a central core, with mounted plugs and O-rings; • Mass transfer coefficients were 10 times higher than commercial parallel-module at very low flow rates; • The mounted plugs and O-rings created multiple shell-side passages. |  <p>Fig. 3 Fabric woven module</p> |
| Vane module [49] | <ul style="list-style-type: none"> • A vane of hollow fiber fabric mounted diagonally inside an open ended box; • It had the highest mass transfer coefficient compared to the first two configurations. However, the fabrication is the most complicated. |  <p>Fig. 4 Vane module</p> |

Partially-baffled
rectangular module
[48]

- Woven fibers with 2 baffles;
- Both countercurrent contact and perpendicular flow were supplied to the well spaced fibers;
- Performed more poorly than non-baffled modules, due to the stagnation of liquid caused by the baffles.

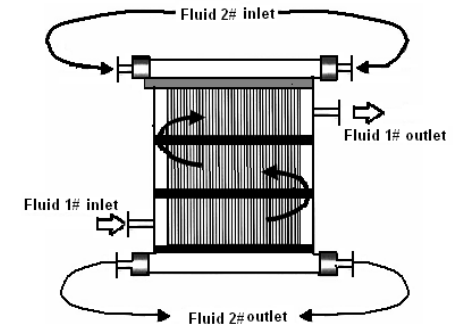


Fig. 5 Partially-baffled rectangular module

Fully-baffled
cylindrical module
[48]

- Woven fibers with 5 baffles;
- A better fluid mixing since the fluids tended to flow backwards and forwards between the spacers;
- Achieved the highest mass transfer coefficients.

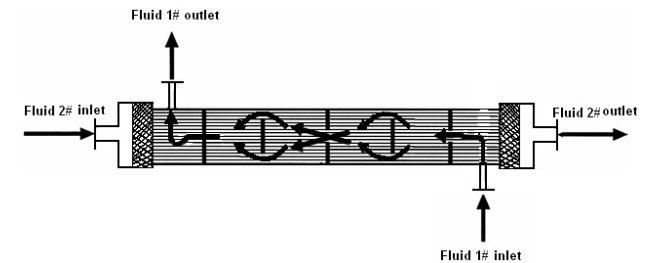


Fig. 6 Fully-baffled cylindrical module

4.1.2 Hollow fiber modules with transverse flow

Another geometry-based membrane module improvement technique is known as “transverse flow” or “radial cross-flow” (baffled modules have some of this feature). With this technique, the membrane module has a central tube for shell-side feed distribution. The flow pattern in the module is radial cross-flow. The function of the central tube is to eliminate the concentration polarization and enhance the process in the upstream when scaling-up to a larger diameter (e.g. 0.3m). This configuration can also be achieved by forming a membrane bundle with knitted hollow fiber fabrics instead of individual hollow fibers, similarly to the modules discussed in section 4.1.1.

One of the best-known commercial modules with a central tube is the Liqui-Cel[®]Extra-Flow module (as shown in Fig. 7), which was patented by CELGARD LLC [51]. This module contains Celgard[®] microporous polypropylene fibers that are woven into a fabric and wrapped around a central tube feeder that supplies the shell-side fluid. The woven fabric allows a more uniform fiber spacing, which leads to better flow distribution and higher mass transfer coefficients than those obtained with individual fibers. The fibers are potted into a solvent-resistant epoxy or polyethylene tube-sheet (Fig. 7).

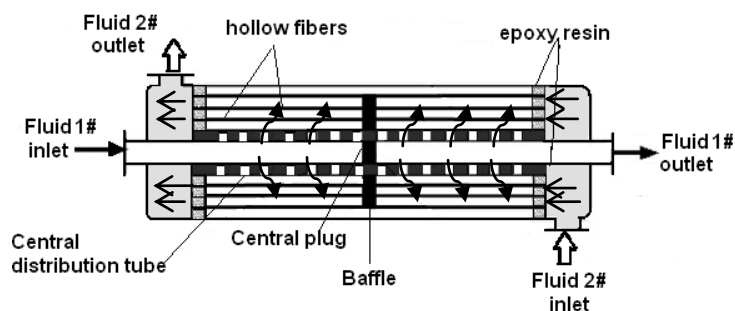


Fig 7. The design features of Liqui-Cel[®]Extra-Flow module (redrawn from [51])

The Extra-Flow module has a central shell-side baffle which improves the module efficiency by minimizing shell-side bypassing and provides a radial cross-flow to achieve a higher mass transfer coefficient than that of conventional parallel flows (see Section 4.1.1). The largest module can handle liquid flow rates of thousands of liters per minute [52].

Another transversal flow membrane module for liquid separation contains a number of hollow fibers which are arranged perpendicularly to the longitudinal axis of the module [53]. This type of module comprises many transverse-current flow segments formed by the seals between the main body and the shell. The channels in the fibers are connected to a space presented around the body which is further surrounded by a shell.

The concept of transversal flow in hollow fiber modules arose from the fabrication of similar flat sheet modules [54, 55]. It has recently been widely applied to gas-liquid absorption such as CO₂ removal from natural gas, pervaporation of ethanol from water, concentration of organic substances from aqueous solutions and dialysis in the artificial kidney (as shown in Fig. 8). Similarly, a rectangular cross-flow module, which introduces transverse flow with staggered fiber arrangement and mounted face plate, was designed by Sirkar et al for MD process recently [56]. Compared to the conventional contactors such as mixing towers or columns, the membrane contactors can avoid the constraints of flooding, loading, entrainment and foaming. In addition, this membrane module featured with a special fiber layout provides better mixing, higher recovery and lower energy consumption than the hollow fiber module with a parallel layout. In spite of the complication in module assembly, one of the most competitive advantages of the transversally-arrayed hollow fiber module in liquid separation is the reduction of channeling and polarization phenomena. It may also help to avoid the membrane wetting since the whole system employs a relatively low velocity due to the smaller hydrostatic pressure drop along the fiber [57]. Hence, such configurations can be potentially applied to new processes like membrane-based extraction and membrane distillation which are subject to pore wetting.

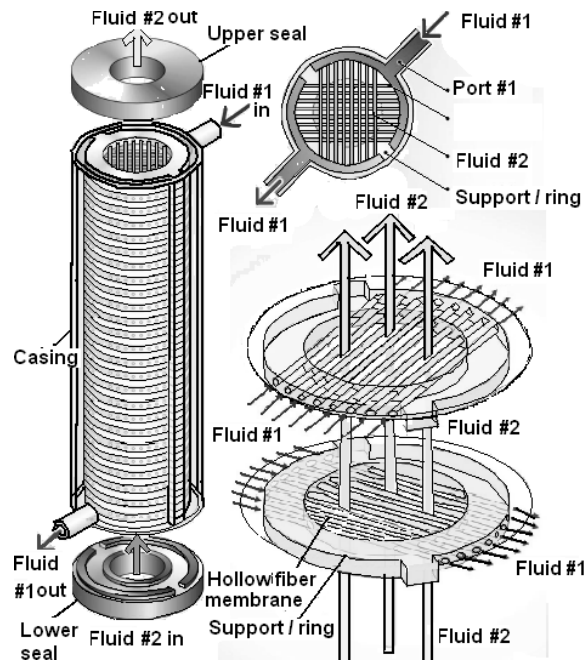


Fig. 8. Transversal membrane contactor (redrawn from [53])

4.1.3 Dual hollow fiber modules/ U-shape modules

A dual hollow fiber module for CO₂ removal is shown in Fig 9. This integrates the absorption and desorption processes in one module with different fluids flowing in the two bundles, respectively [57]. Similar designs can be found from previous studies [58, 59]. Compared to conventional linear modules, this type of module comprises one or more hollow fiber bundles which not only increases the contact area between fluids, but also crimp the flow channels to create better hydrodynamics. It was reported that this may favor both the gas and liquid separation due to the improved permeate flow characteristics and improved space/volume characteristics. The fiber bundles are very flexible and can be of any shape rather than being subject to mechanical stretching by the sealing epoxy. The possible configurations are shown in Fig. 10. To avoid or minimize liquid film transfer resistance, the liquid within the module needs to be agitated by circulating or other means (e.g. stirrer).

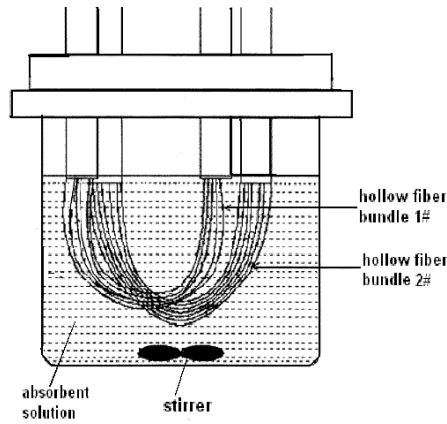


Fig. 9. Dual hollow fiber module (redrawn from [57])

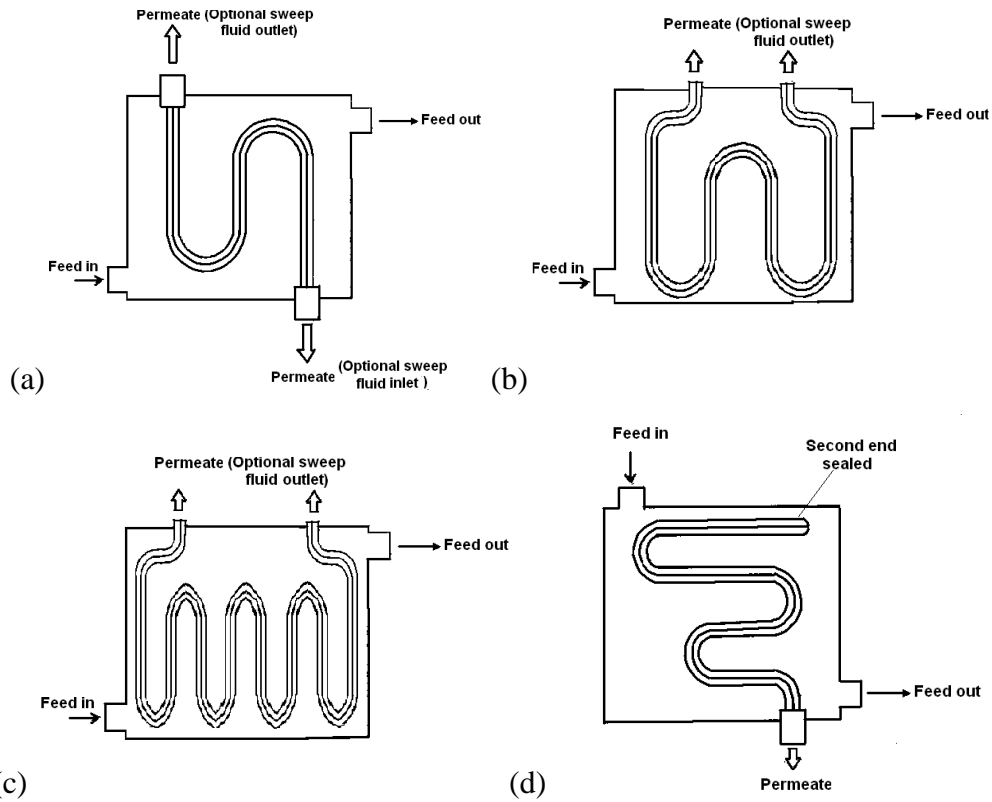


Fig. 10. Flexible U-shape hollow fiber modules: (a) coiled; (b) French horn; (c) spiral; (d) one-ended U shape (redrawn from [59])

4.1.4 Hollow fiber module with modified fiber geometries

Most researchers focus on introducing channeled designs to enhance the flow passage, presenting various fiber layouts to even the flow distribution effectively, inserting turbulence promoters such as spacers, screens or baffles. Limited work [8, 60-63], however, has been done

to investigate the enhancement effect of hollow fiber configurations with wavy geometries such as crimped, braided and twisted fiber geometries, shown in Fig 11.

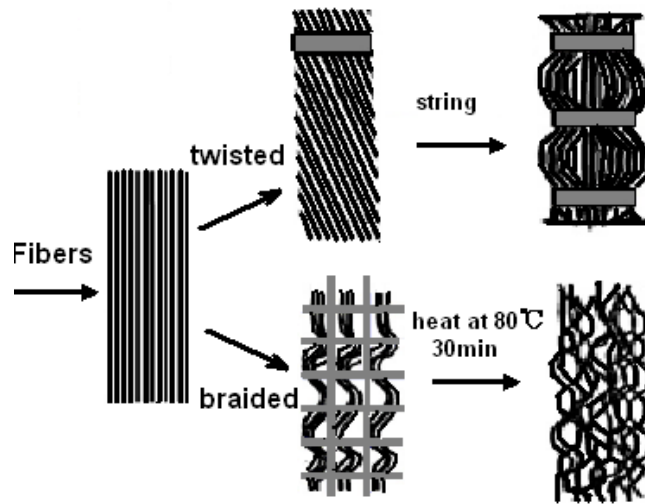


Fig. 11. Hollow fiber configurations with wavy geometries (redrawn from [8])

As reported by Teoh et al [8], the application of different hollow fiber configurations with wavy geometries in the MD process led to flux enhancement by as much as 36% compared to that of a conventional straight-fiber module without inserting any external turbulence promoter. This seems to be more efficient than window or helical baffles assisted systems which correspond to 20~28% enhancements, respectively. Ghogomu et al [61] studied MD using a hollow fiber module with coiled fibers. It was found that all the curved geometries, such as those that are helically coiled, twisted sinusoidal or meander-shaped, can induce dean vortices (secondary flows) which can significantly enhance the process as compared to conventional straight fibers. Li et al [62] also reported the use of a commercial hollow fiber module Monsanto's Prism® with crimped fibers which was made by Monsato Company. Although it has been successfully used in H₂ recovery, it may have the potential to be applied in liquid separation processes in the future.

Curved fibers as a geometry improvement can efficiently and easily be applied to increase the fluid-membrane contact area per unit volume, create better hydrodynamic conditions and

enhance membrane flux. Moreover, this type of configuration can find its place in a broad range of industrial applications.

4.1.5 Other hollow fiber modules

Some other passive, geometry-based membrane module improvements are described here. One hollow fiber module with a plurality of membrane units was designed to perform an attempted separation of components from a multi-component feed (Fig. 12), each unit contains a number of elongated hollow fibers which are connected to the collecting manifolds. The axial movement of fibers is allowable due to unrestrained manifolds. This design solves a classical problem in hollow fiber modules that fibers should have a longer length than the shell in case of axial shrinking, and it alleviates the differential expansion between the membrane tubes and shell since the novel design allows the membrane tubes to expand independently. In order to meet different requirements of different separation processes, an integral two-stage (in parallel or series) module with two embodiments is also applicable [62] (Fig. 13).

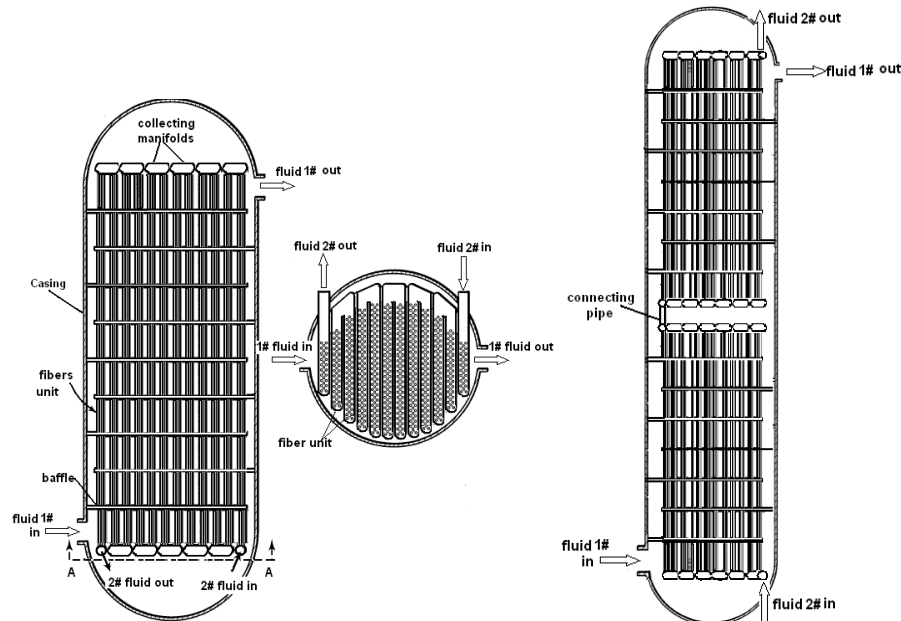


Fig 12. Novel hollow fiber modules for fluid separation (redrawn from [64])

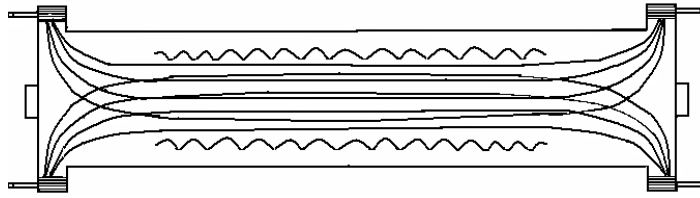


Fig. 13. Internal staged permeator for fluid separation (redrawn from [65])

Submerged membrane modules are more versatile in aqueous separation processes; they are widely used in membrane bioreactor processes [66] (Fig. 14). In most cases, due to the severe fouling in biological wastewater treatment, the submerged module is operated with air sparging which can enhance the process effectively. This approach will be discussed in more detail in Section 4.2.2.

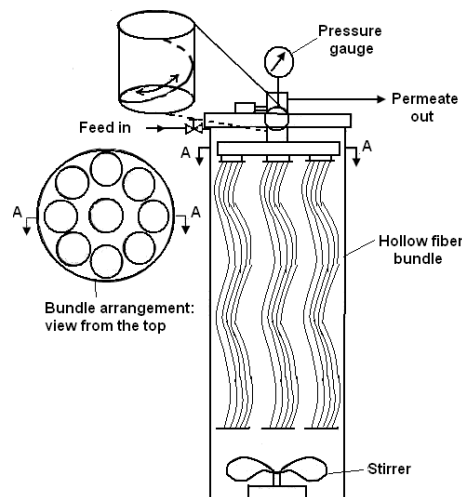


Fig. 14. Advanced submerged hollow fiber module (redrawn from [66])

Some of the membrane separation processes (such as membrane distillation or osmotic distillation) require extra cooling or heating devices for post treatment or to increase process driving force. Multi-functional modules, which serve separation as well as heat exchange purposes, have been developed [67, 68]. If the heat exchange operation is sufficient in a single module, then subsequent connected heat exchangers may be rendered unnecessary. For example, Memstill[®] technology developed by TNO institute and Keppel Seghers Company is now operated at the pilot scale in Singapore [67]. It combines a continuum of evaporation stages in countercurrent flow pattern which makes the heat recovery process simultaneously. A similar concept can be found from a European patent (Fig. 15).

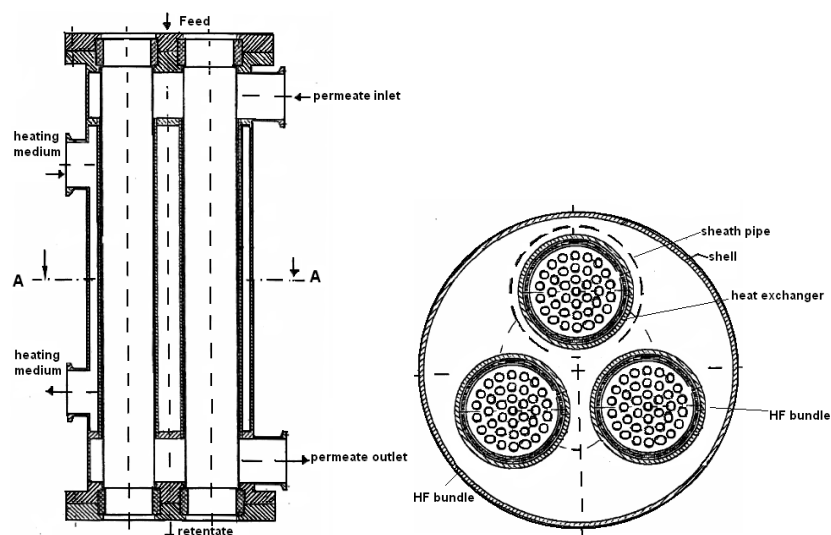


Fig. 15. Filter membrane module comprising an integrated heat exchanger (redrawn from [68])

Most of these passive enhancement modules have been applied in gas/liquid contactors. However, they have the potential to be used for concentration-driven liquid/liquid mass transfer and temperature-driven MD processes, where their simplicity (and hence ease in manufacturing) would be an advantage. Furthermore, these configurations offer substantially higher mass transfer rates. Some of the commercially available filtration modules can also be applied to concentration-driven or thermally-driven processes such as MD, as they are able to provide good mixing conditions to enhance heat and mass transfer in both the bulk solution and the solution-membrane interface. It should be noted that most of these modules have only been studied in laboratory scale, except the Liqui-Cel[®] Extra-Flow module. As reviewed previously by Gabelman and Hwang [20], there are some other commercial examples which have not been categorized due to insufficient information, such as the DISSO3LVETM module (W.L. Gore & Associates) which was primarily applied for the ozonation of semiconductor wastewater treatment; the SeparexTM EFM-530 module (Pall Corporation) which was used in ultrapure water applications as a bubble-free gas/liquid membrane contactor; and modules designed for oxygenation processes in bioremediation and aeration (Membrane Corporation) were also used as bubble-free gas/liquid membrane contactors in wastewater treatment. In addition, in the late 1980s, Enka AG developed a commercial hollow fiber module for the MD process [69].

4.2 Active enhancement techniques

Advancements in membrane materials and membrane fabrication techniques and the resulting increase in broad applications of membrane-based processes have facilitated development of technologies for membrane modules. There have been several generations of membrane modules to meet the demands of various applications. While the passive enhancement techniques described above can enhance membrane performance significantly by utilizing and distributing the energy of the fluid flow itself, there are still limitations that allow these techniques to only offer a moderate enhancement in mass transfer, which is limited by concentrated or viscous feed solutions.

In contrast, active enhancement techniques allow the introduction of various forms of external energy to improve membrane processes. Fane and Chang [9] have briefly summarized various active enhancing strategies and reviewed their development up to early 2005. Those strategies include pulsed flow, high shear devices (rotating and vibratory systems), two-phase flow systems, electro-filtration, ultrasound-enhanced filtration, etc. This paper will focus on the mass transfer enhancing mechanisms and progress in the past five years on the most commonly proposed techniques, such as bubbling, vibration waves and ultrasound. Additionally, the benefits and drawbacks of these active enhancement techniques will be highlighted in this section, and they are then further compared in Section 5 with respect to fabrication cost, energy demand, scale-up potential, etc.

4.2.1 Bubbling system

The most widely used active approach to avoid membrane fouling in membrane-based processes, especially in membrane bioreactors, is air bubbles [11, 70-73] to induce liquid movement and promote surface shear and reduce membrane fouling. Especially in a membrane bioreactor process, air sparging serves the double purpose of providing aeration and causing two-phase flow to control fouling. As reviewed by Cui et al [74], the mechanisms of process enhancement and fouling control using bubbling systems (gas flow applied either inside or outside of the fiber) includes:

- (1) Bubble induced secondary flow;
- (2) Displacement of the concentration polarization layer;

- (3) Passing bubble induced pressure pulsing;
- (4) Increase of superficial cross-flow velocity;
- (5) Movement of the fibers (external bubbling and loose fibers).

To correlate the bubble size/characteristics (effects of air flow rate, orifice size, fluid properties, submergence. etc) and bubble induced fiber movement into the module performance, it is essential to characterize the uniqueness of the bubbling system and distinguish the contribution from bubbles of different sizes. Many researchers [11, 75-78] have investigated the effect of bubble size on module performance in the submerged MBR systems. For example, to observe the relationship between bubbling and module performance via critical flux, trans-membrane pressure (TMP) and membrane fouling formation, Wicaksana et al [11] studied the interaction between bubbling and fiber movement in submerged hollow fiber membranes. It was found that a lower fouling rate could be achieved by more fiber movement under certain conditions such as fiber looseness, smaller bubbles, higher air flow rate, lower feed viscosity and lower solid concentration. The authors also stated that the fiber movement was enhanced by using thinner and longer fibers, but it was insensitive to nozzle sizes (bubble sizes) used in the system. To study the fouling mechanism in submerged hollow fiber membrane modules with bubbling, Yeo et al [76, 79] used particle image velocimetry (PIV) to examine the bubble-induced phenomena by varying and correlating different operating parameters, they also stated that many small bubbles are better than few large bubbles.

Although Fane & Chang [80] and Cui et al [74], have extensively documented the development of membrane processes associated with bubbling and demonstrated the benefits of bubbling systems that have caused an upsurge of interest in the use of air bubbles to enhance membrane process (e.g. submerged membrane bioreactors), there are some limitations in the applications of this coupled system. For instance, in most bio-separation processes using UF/MF hollow fiber modules [74], the fragmentation of protein or micro-organisms [81-83] could occur and aggregation could easily happen due to the high shear rate when bubbles burst. Therefore, bubble-flow induced bio-separation process can only perform well under relatively low air sparging rates. In high pressure membrane processes (NF and RO), the air can be dissolved into

the feed at the high pressure side and released into the permeate side which may lead to back pressure build-up and lower the efficiency of the separation process. Additionally, a certain volume of gas must be injected into the modules at the operating pressure to achieve a higher critical flux. This could be energy intensive for high pressure applications.

Although the concept of gas sparging to enhance transport and reduce fouling formation can be very effectively applied to various membrane processes, a comprehensive study is deficient on the characteristic flow patterns (bubble flow, slug flow, churn flow and annular flow) in this gas-liquid two phase system, the dominant role of the slug flow regime, parameters contributing to pressure losses and fouling rate controlling factors. In hollow fiber modules, it is important to determine that the bubbles should be introduced through tube or shell sides, and to overcome the difficulty in ensuring even air distribution in a confined hollow fiber module.

4.2.2 Vibrating membranes

The original concept of dynamic filtration to improve membrane performance by applying vibration was initiated by Armando et al [84] from New Logic International Inc. The system is known as vibratory shear enhanced processing (VSEP), and contains a stack of membrane disks mounted in a circular casing connected to a torsion spring and a motor. The motor generates a vibrating force on the membrane elements. The vibrations can help to disrupt the concentration and/or temperature polarization and fouling layer formation, which as described above are the major challenges in membrane-based processes. This concept has also been commercialized by Pall Filtration, US [85], their product was named as PALL-Sep Vibrating Membrane Filter.

Compared with the conventional cross-flow system, a vibrating membrane offers several advantages. The conventional cross-flow system has a relatively low shear rate (less than 10,000~15,000/s), which limits its application for high-concentration and high-viscosity feed solutions. Moreover, in spite of the high flow rate introduced into the system, membrane fouling and flux decline still easily occur due to an insufficient shear rate that cannot prevent the accumulation of retained particles on the membrane surface. In comparison, the vibrating system of the VSEP unit induces a much higher shear rate (100,000~150,000/s) that increases turbulence

at the membrane surface and promotes the back diffusion of particles to the bulk solution effectively. Comparison of the working principles and wall velocity distributions in a conventional cross-flow and a VSEP system are shown in Fig. 16 and Fig. 17, respectively. It is clearly illustrated that in a VSEP system, the maximum flow velocity occurs near the membrane wall which will break down the boundary layer and keep particles suspended above the membrane surface [86]; while in a conventional cross-flow system, the flow adjacent to the membrane wall is stagnant.

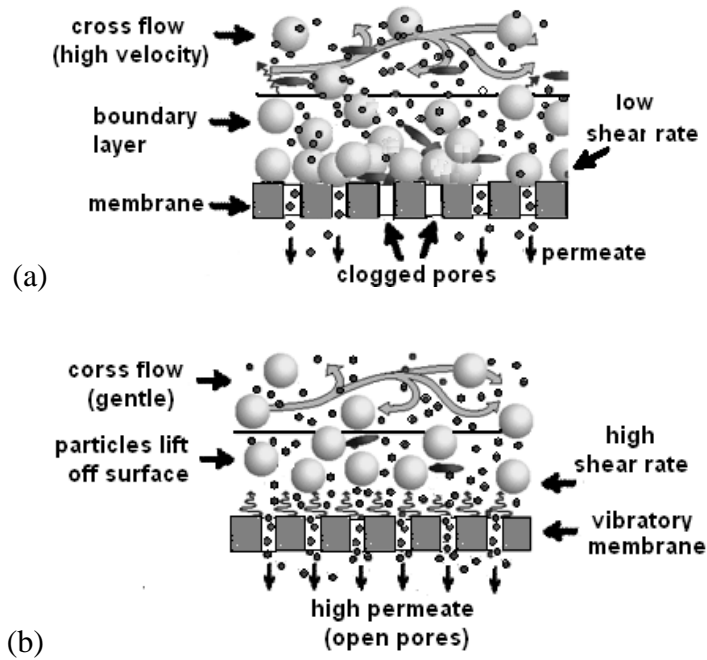


Fig.16. The mechanism of particles removal due to shear in (a) conventional cross flow system and (b) VSEP unit (redrawn from [87])

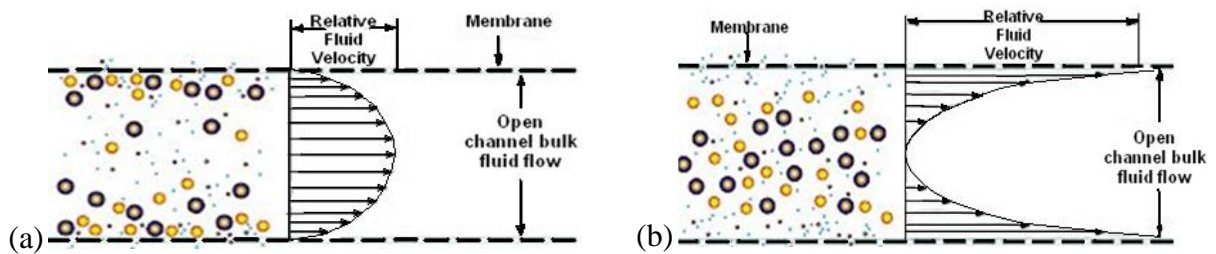


Fig. 17. Comparison of velocity distribution profiles in:
(a) cross-flow system; (b) VSEP system

Due to its benefits, the commercial VSEP module has been successfully used in treating concentrated feeds such as landfill leachate and high-salinity seawater (mainly reverse osmosis) in industry [86]. Recently, several researchers have tried to broaden its applications in the food industry [88, 89] or pervaporation process [90], and extend this concept to hollow fiber modules such as submerged membrane bioreactors [91-94]. Although the hollow fiber modules have higher potential for practical applications, there are only limited studies involving vibrating assisted hollow fiber modules [10, 91, 93, 94] (a vibrating submerged hollow fiber module is shown in Fig. 18).

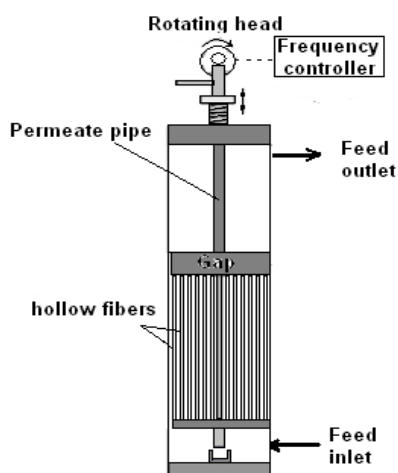


Fig. 18. Hollow fiber vibrating membrane bio-reactor (VMBR) associated with vibrating device (redrawn from [93])

To summarize the vibrating membrane techniques, vibrating the membrane itself, as opposed to vibrating the flow, can advantageously achieve the most relative motion on the membrane surface. This motion between bulk solution and membrane can greatly reduce the liquid boundary layers and the membrane fouling, polarization effects on both sides of the membrane. As a result, vibratory systems might have the potential to be coupled with other processes which suffer from low permeability or severe polarization, such as MD or membrane distillation bioreactors (MDBR).

Thus, it is hoped that the vibrating concept can be implemented for various applications because it offers economically competitive advantages in treating high-salinity water and has the potential to greatly advance the use of membranes in desalination. However, there are also some limitations in this area, such as the potentially high demand of external energy input (detailed in Section 5) and the complexity of rotating devices, which lead to the relatively high operation maintenance and equipment cost in the system.

4.2.3 Ultrasonic systems

Ultrasonic waves, as one of the active enhancement techniques in membrane separation, refers to acoustic waves of frequency between 20 kHz and 10 MHz accompanied by some concomitant physical effects, such as those to do with mechanics, thermotics and cavitation. The propagation of ultrasonic waves in various media is beneficial to many physical and chemical processes.

By introducing ultrasonic vibration, micro-streaming induced rapid fluid movement, acoustic heating and cavitations, ultrasonic technique owns the practical capability to enhance filtration and membrane separation by mitigating membrane fouling, reducing concentration/temperature polarization effects and removing fine particles from the surface. It has been successfully applied to several membrane processes [95-105], such as MF, UF and dialysis which suffer from concentration polarization and subsequent fouling. It has also been reported that acoustic vibration and induced heating could enhance thermally driven processes such as MD [12, 106], and improve their permeability, greatly reduce temperature polarization and membrane fouling. The mechanism of an ultrasonic irradiation system is illustrated in Fig. 19.

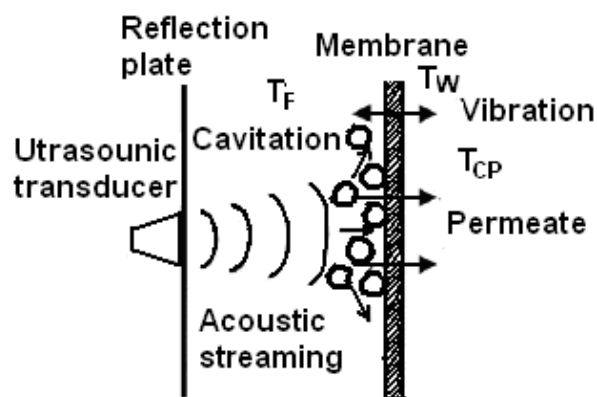


Fig. 19. Mechanism of ultrasonic irradiation system (redrawn from [106])

This concept was initiated by Madsen [107] who investigated the influence of ultrasound on hyper-ultra-filtration membrane separations. Later, Kobayashi et al [98, 108] observed for a cross-flow UF of dextran that the high frequency vibration of the membrane surface resulted in the reduction of concentration polarization, thus increasing permeate rate as compared to the classical stirred system. The authors also stated that ultrasonically assisted dead-end UF system can be equally effective. It is also reported that the ultrasonic wave can induce convective currents and cavitation which are able to mitigate the concentration polarization [109]. Other authors found that ultrasound is a promising technique to recover trans-membrane flux [110, 111]. For example, 70~80% recovery was achieved for a Cu–Polyethylenimine (Cu–PEI) solution. A detailed review on flux recovery with ultrasound can also be found in other literature [9].

By combining the experimental data and mathematical modeling, researchers found that process improvement increased with increasing intensity, and decreased with increasing acoustic frequency, solution temperature or even active membrane area. In an ultrasonic-assisted air gap MD system [106], the predicted enhancement was up to 200% with 0 to 5 Wcm⁻² intensity of the ultrasonic irradiation.

In previous studies, the ultrasonic enhancement technique was mainly applied to filtration process using flat sheet membranes, and only a few studies have been reported on other membrane processes involving hollow fiber modules [99, 104, 105, 112]. This may be due to the

difficulty in identifying the appropriate position to place the reflection plate (refer to Fig. 19) and the transducer on the module; and the process enhancement factor can be affected. Thus, the enhancement potential of ultrasonic-assisted hollow fiber modules (membrane contactors) is yet to be exploited.

Notwithstanding the positive enhancement to several membrane processes, some authors have reported that ultrasonic radiation at inappropriate frequencies and intensities may damage the membrane. For instance, it has been observed via field emission scanning electron microscopy (FESEM) that some polymeric materials can be restructured by ultrasonic irradiation [110, 113, 114], such as polyethersulphone (PES), cellulose nitrate with cellulose acetate (CN–CA) or nylon6 (N6); on the other hand Poly Vinylidene Fluoride (PVDF) and Poly Acrylonitrile (PAN) showed no observable damage with long term exposure. Though some work has been done to examine the mechanism of membrane damage by ultrasound [115], caution must be taken to choose proper membrane materials, ultrasonic intensity and irradiation duration to avoid membrane damage.

4.2.4 Miscellaneous techniques

Beside vibration, bubbling and ultrasonic techniques which have been intensively reported, there are other techniques, such as magnetic stirring, ozonation [116] the use of electric fields [117] and even the introduction of bi-disperse suspensions for higher critical flux in RO systems [118], that can create enhanced hydrodynamic conditions in membrane separation systems to enhance the permeation and reduce membrane fouling.

5 Qualitative comparison of enhancement techniques

Both passive and active enhancement techniques described above have demonstrated the feasibility of enhancing membrane performance. However, the main objectives of module design should not only focus on maximization of the system performance through optimizing the flow geometries and external assistant devices, but also include minimization of the cost per unit of

mass transferred (energy consumption and module production cost), the potential of scale-up and niche applications. Considering these factors, these two enhancement modes have their own advantages and limitations.

As the fiber arrangement and module configuration will directly affect the shell side mass transfer, which plays an important role in membrane contactors, the passive methods offer reasonable flux enhancement by effectively distributing the energy of the flow itself and broaden the potential applications of hollow fiber modules. However, the flow channels have to be carefully designed. For example, the fabric woven module (Fig.3) gained 10 times higher mass transfer coefficient than the conventional parallel module at very low flow rates, due to the uniform fiber spacing and the multiple flow passages created by the mounted plugs and O-rings, while the baffled-rectangular module (Fig.5) performed more poorly than baffled ones due to the stagnation of liquid caused by the baffles. This suggests that passive turbulence promoters do not always enhance the module performance. From the engineering point of view, the complex fabrication procedures of these high-performing hand-built modules may outweigh the benefit of enhancement and restrict their commercialization [20]. Additionally, complex module geometries tend to form closely packed configurations or patterns? which may not be applicable to the treatment of concentrated or viscous solutions.

In contrast, the active enhancement methods show advantages in treating concentrated or viscous feed solutions, such as municipal waste-water treatment with MBRs incorporating bubbling [92] or vibratory devices [93], and the treatment of landfill leachate and RO brines by vibratory modules [86, 87]. Typically, the active methods offer 3-15 times enhancement with the same membrane area by creating the shear-induced liquid movement and hence suppressing the polarization and fouling rate. However, energy consumption and capital cost are of major concern in the active enhancement systems, as these systems involve potentially high demand for external energy input (e.g. ultrasound) or greater design complexity (vibratory or rotating systems).

Fortunately, many energy-reduction strategies have been proposed by different researchers. For example, in submerged MBR systems, the main contribution of energy consumption is aeration and effective implementation of intermittent air-sparging and appropriate nozzle sizes can minimize the cost and suppress the fouling rate effectively [74, 122, 123]. In high flux vibratory systems, the major energy consumption is rotation or vibration. For some applications, the VSEP system uses the resonant frequency to maximize the vibrating amplitude and presents a low specific energy consumption (*SEC*) in industrial modules. The *SEC* is estimated as 4.1 kWh m⁻³ product in MF and 2.5 kWh m⁻³ product in RO for waste water treatment with a 150 m² VSEP module and would be 22% less for surface water treatment [87]. Genkin et al [92] applied vibrations to a submerged membrane system to achieve considerably higher critical fluxes of 130 L h⁻¹ m⁻² (normally around 20 L h⁻¹ m⁻²) by applying a combined axial-transversal oscillation at a low frequency of 10 Hz. Furthermore, with the addition of a reasonable amount of coagulant (34 mgL⁻¹) the critical flux reached a maximum value of 86 L h⁻¹ m⁻² at low frequency (1.7 Hz) instead of at high frequency (10 Hz) when floc break up tended to occur. (This low frequency operation required a *SEC* of only 0.29 kWh m⁻³). With the application of ultrasound, researchers [12, 103] also found performance enhancement at certain frequency (lowest 20 kHz) and irradiation intensity, but the *SEC* was relatively higher (e.g. 352.9 kWh m⁻³, with flux of 16 L h⁻¹ m⁻² in a lab-scale air gap membrane distillation system using PTFE flat sheet membrane [12]) than other systems because of the high frequency imposed. Thus, compared to the other techniques, the ultrasonic system may be the least economic method.

Finally in terms of the potential for scale-up, the applications of bubbling and vibratory systems on MBRs and concentrated feed treatments have received attention both in laboratory and industrial scales. Although external energy and complex assistant devices are required, they can be easily scaled up [119, 120]. However, some active methods such as ultrasound may not be suitable for larger scale applications, because of the need to provide even distribution of ultrasonic radiation, the attachment of reflection plate and also the streaming-induced heat which

may cause additional problems[104, 121], though it is found to benefit some thermal processes such as MD or MDBR [106].

To qualitatively evaluate various enhancement methods, a brief comparison based on the analysis of potential process enhancement, fabrication cost and complexity, energy demand, scale-up potential and niche applications is give in Table 3.

Table 3. Qualitative comparison of various enhancement methods

| Enhancement technique | Passive enhancement (modified configurations) | Active enhancement (Dynamic shear-induced systems) | | |
|---|---|--|---|--|
| | <i>Fabric woven modules</i> | <i>Bubbling system</i> | <i>Vibratory system</i> | <i>Ultrasonic system</i> |
| Potential enhancement | Moderate | Moderate | High | High |
| Fabrication cost (equipment cost/complexity) | High | Low to moderate | High | High |
| Energy demand | Low | Low to moderate | Moderate to high | High |
| Scale-up potential | Moderate to high | High | High | Low |
| Niche applications | Membrane contactors (gas-liquid or liquid-liquid separations) | Submerged MBRs (municipal/industrial waste- water treatment) | Concentrated & viscous feed solutions (e.g. Landfill leachate, RO brine), or MBR systems | UF systems, Seawater desalination (e.g. MD, MDBR) |
| Reference | [20] | [120] | [119, 124] | [12, 109] |

Above all, the approaches for energy reduction in membrane processes are focused on minimizing the fouling rate while maintaining an optimum performance, which is associated with reliable and economic operation to produce high quality products. This is the key factor that drives membrane technology to be more competitive than the conventional separation methods.

6 Conclusions

Membrane-based separation processes have found numerous applications in industries in recent decades. However, concentration and temperature polarization (in MD) and membrane fouling-induced high energy consumption, low productivity and short membrane lifespan continue to present severe technical challenges to the commercialization of most membrane processes for liquid separation. Novel membrane module technology is one of the key technologies to tackle the challenges.

Attempts to improve membrane module design should begin with a better understanding of the mass transfer, therefore this review provides a summary of prior studies on the mass transfer models related to both the shell-side and tube-side fluid dynamics. Based on the mass transfer analysis, two types of membrane performance enhancements have been discussed. The primary approach (referred to as “passive enhancement techniques”) is to design and fabricate effective modules with optimized flow geometry to suppress the undesirable concentration polarization, temperature polarization and fouling. The other method (referred to as “active enhancement techniques”) is to utilize external energy to induce a high shear so as to facilitate the mixing and reduce the thickness of the concentration/temperature boundary layer over the membrane surface.

Generally, the passive enhancement techniques offer moderate increases in mass transfer but cannot provide a convenient means to control the degree of process enhancement. Active techniques, on the other hand, have been shown to provide 3~15 times enhancement on the permeation flux. However, both enhancement modes have their advantages and disadvantages. Regardless of which enhancement mode is chosen, there is still much to be done in achieving optimum operating conditions.

As most of the concepts mentioned here have not been industrialized, and some commercial applications have not yet reached their full potential, attempts to develop novel modules should begin with better understanding of the mass transfer in the membrane module. Fundamentally, for industrial applications, the design objectives should minimize the cost per amount of mass transferred and optimize other features such as fiber characteristics (diameter, thickness, porosity, tortuosity and length), packing density, operating flow rate, flow direction and fluid properties, etc. Constrains of module fabrication cost, scale-up potential, operating period, fouling control and membrane replacement should also be considered. It is hoped that this review can provide inspiration for novel module development.

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Nomenclature

| | |
|---------------------|---|
| A | effective membrane area, m^2 |
| a | constant of Eq. (5) |
| d_h | hydraulic diameter, m |
| $d_{t \text{ in}}$ | inner diameter of the hollow fiber, m |
| $d_{t \text{ out}}$ | outer diameter of the hollow fiber, m |
| D | diffusion coefficient of the solute through the membrane, $\text{m}^2 \cdot \text{s}^{-1}$ |
| D_s | diffusion coefficient of the solute in shell-side, $\text{m}^2 \cdot \text{s}^{-1}$ |
| D_t | diffusion coefficient of the solute in tube-side, $\text{m}^2 \cdot \text{s}^{-1}$ |
| ΔF | overall driving force of the mass transfer |
| Gz | Graetz number, $Re \cdot Sc \cdot \frac{d_h}{L}$ |
| J | overall flux J_s of the solute to be removed or retained, $\text{kg} \cdot \text{m}^{-2} \cdot \text{s}^{-1}$ |
| k | overall mass transfer coefficient, $\text{m} \cdot \text{s}^{-1}$ |
| k_{shell} | local mass transfer coefficient in shell side, $\text{m} \cdot \text{s}^{-1}$ |
| k_{tube} | local mass transfer coefficient in tube side, $\text{m} \cdot \text{s}^{-1}$ |
| k_m | local mass transfer coefficient in membrane, $\text{m} \cdot \text{s}^{-1}$ |
| L | effective fiber length, mm |
| Re | Reynolds number, $\frac{d_h v \rho}{\mu}$ |
| Sc | Schmidt number, $\frac{\mu}{\rho D}$ |
| Sh | Sherwood number, $\frac{k d_h}{D}$ |
| v | fluid velocity, $\text{m} \cdot \text{s}^{-1}$ |

Greek letters

| | |
|---------------|--|
| α | constant in Eq. (5) |
| β | constant in Eq. (5) |
| ψ | constant in Eq. (5) |
| ϕ | packing density |
| δ_m | membrane thickness, μm |
| η | pump efficiency, % |
| μ | viscosity of the fluids, $\text{Pa}\cdot\text{s}^{-1}$ |
| τ | membrane tortuosity |
| ε | Porosity |

Suffix

| | |
|-----|---------------|
| m | Membrane |
| p | Permeate |
| s | Solute |
| w | membrane wall |

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