

# Hydrophobic Hollow Fiber Membranes for Treating MTBE-Contaminated Water

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Soluble contaminants with low Henry's constant, such as methyl *tert*-butyl ether (MTBE), require innovative solutions for water treatment. Given the increased frequency at which MTBE is detected at contaminated sites, the development of new technologies is of considerable relevance. Hydrophobic hollow fiber membranes (HFM), used in industrial and medical applications, have interesting physicochemical properties that make them particularly suitable to deal with these contaminants. The hydrophobicity of the fiber maintains adequate separation between aqueous and gaseous phases, permitting an efficient separation of volatile and semivolatile compounds from water to gas. The hollow nature of the fiber and its high porosity permit high rates of mass transfer across the membrane. The mass transfer process can be accelerated using pervaporation and by increasing the solution's temperature to increase the Henry's constant and the overall mass transfer coefficient. In these studies, we evaluate the removal efficiency of MTBE from water using a commercial HFM module and develop the corresponding dimensionless mass transfer correlations necessary for the design of industrial-scale systems. We found that the Lévêque correlation for the tube-side mass transfer coefficient is in general applicable for MTBE pervaporation through a hydrophobic HFM. MTBE removal is a strong function of membrane length, water flowrate, and solution temperature but is almost independent of gas-phase parameters.

## Introduction

The use of membranes for water treatment has gained increasing importance for special applications where the properties of the pollutant(s) require innovative thinking. In general, membranes have not been considered for the separation of volatile organic compounds (VOC) from water, since it is generally perceived that these solutes would be too small for an efficient separation through a membrane. In addition, for many VOCs, air stripping or GAC is sufficiently cost-effective to warrant their continued use. However, for very soluble VOCs, such as methyl *tert*-butyl ether (MTBE), air stripping or GAC have been shown to be rather ineffective (1). In this case, the physicochemical properties of the solutes warrant the use of new approaches.

MTBE is a gasoline additive used throughout the United States to reduce emissions to the atmosphere by improving

the combustion efficiency and reducing the volatility of the mixture and increase gasoline's octane rating. In California and other air quality nonattainment areas, MTBE makes up to 11% (v/v) of the gasoline. Given its high water solubility, it transfers rapidly from gasoline leaks or spills to the surrounding water phase. MTBE's low sorption in soils and very low rates of natural biodegradation under most conditions have resulted in significant contamination throughout the United States and elsewhere where MTBE is used (2-5). Odor and taste detection levels are as low as 5 µg/L, making its efficient removal very important.

A membrane is a semipermeable thin layer of material separating two fluids based on their physicochemical properties. There are several types of membranes, including microfiltration (MF), reverse osmosis (RO), and hollow fiber membranes (HFM). MF membranes can remove particulates, RO membranes retain solutes as water permeates the membrane, and HFM can be used to degas liquids, taking advantage of the membrane's hydrophobic nature (6). Interest in membrane technology has driven research in various fields, such as environmental engineering, water and wastewater treatment, biomedical applications, and the food industry (7).

Microporous HFM have been successfully used to strip various compounds from water such as VOCs, hydrogen sulfide, sulfur dioxide, oxygen, carbon dioxide, bromine, and ammonia using a sweep gas to remove the gases and vapors from the membrane surface (8-13). Pervaporation uses a vacuum sweep to remove gases and vapors from the membrane surface. The activity difference driving this process is maintained with a partial vacuum on the permeate side (7). A large concentration gradient is established between the aqueous phase concentration and the concentration in the gas phase. Uses of pervaporation include the dehydration of alcoholic azeotropes and removal of VOC from water (13). Common polymers for hydrophobic membranes include poly(tetrafluoroethylene) (PTFE), polyvinylidene fluoride (PVDF), polyethylene, polycarbonate, or isotactic polypropylene (7).

HFM improves the mass transfer rate of dissolved VOCs from water to air. Contaminated water is pumped through the lumen side of bundled microporous hydrophobic hollow fibers, while a vacuum is drawn counter-currently on the outside of the fibers. The hydrophobic membrane allows for efficient transfer of volatile compounds from aqueous to gas phase. While the water is pumped through the hollow fibers, volatile components volatilize and diffuse through the gas-filled pores of the hollow fiber, due to the large concentration gradient. Volatile compounds can then be pulled through by a vacuum and/or swept away by a sweep gas. Pollutants can be effectively transferred through the gas-filled pores to the gas phase, while water does not permeate due to the polymer's hydrophobic nature. Hollow fibers have a large surface area per unit volume, allowing greater contact between the phases. This provides relatively fast removal of the contaminant. Studies in refs 8 and 14 showed that mass transfer of VOCs could be an order of magnitude greater than achievable by packed tower aeration when using HFM with a sweep gas.

There are several advantages of hollow fiber membranes over air-stripping: (1) lower air flowrates can be used, requiring small vapor-phase treatment units, (2) water and air flowrates can be controlled independently to maximize mass transfer because they are separated by the membrane, (3) there is no aqueous flow channeling due to the hollow fiber configuration, and (4) the air stream will contain little

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water due to the hydrophobic nature of the membrane (14). This allows more efficient treatment of the contaminant in the gas phase.

This study presents the evaluation of a polypropylene hollow fiber membrane system to remove MTBE. Since MTBE has a low Henry's constant and typically does not sorb well, conditions that lead to high removal efficiency using HFM need to be determined. We present the results of our evaluation of the controlling parameters and a generalization of the relationship between the mass transfer coefficients and these parameters.

## Theory

Gas transfer from the lumen of the hollow fiber membrane through the micropores is driven by a concentration gradient. At equilibrium, the concentration in the water ( $C_w$ ) is related to the concentration in the air ( $C_g$ ) by the dimensionless Henry's law constant ( $H = C_g/C_w$ ). Henry's law constant increases with temperature, making diffusion into the gas phase more favorable at higher temperatures.

The constant airflow past the membrane pores effectively keeps the air phase concentration of the contaminant near zero, allowing constant diffusion from the water to the membrane surface and into the air-filled pores. The contaminant's rate of diffusion is limited by three resistances: (1) diffusion through the water to the membrane surface ( $1/k_w$ ), (2) diffusion through the air-filled pores in the membrane ( $1/k_m$ ), and (3) diffusion from the exterior membrane surface into the bulk air ( $1/k_g$ ). The transfer coefficients through the air-filled pores and the bulk air are expressed in terms of water-phase transport by multiplying by  $H$ . The overall mass transfer coefficient,  $K_{oL}$  ( $m\ s^{-1}$ ) for the contaminant is the reciprocal of the sum of the individual resistances (15, 16):

$$\frac{1}{K_{oL}} = \frac{1}{k_L} + \frac{1}{Hk_M} + \frac{1}{Hk_G} \quad (1)$$

The aqueous and gas phase mass transfer coefficients depend on operating conditions, especially water ( $u_L$ ) and gas ( $u_G$ ) velocities in  $m\ s^{-1}$ , the characteristic length ( $d_e$ , inner diameter of the fiber,  $m$ ), the kinematic viscosity of the fluid,  $\nu$ , in  $m^2\ s^{-1}$ , and the diffusivity of the pollutant in either phase,  $D$ , in  $m^2\ s^{-1}$ . The diffusivity of most volatile and semivolatile compounds is approximately 4 orders of magnitude smaller in water than air (17, 18). Therefore, the resistance due to diffusion through the water phase generally controls the overall mass transfer rate, while the membrane has negligible resistance, especially for the thin hollow fibers used in this module (around 100  $\mu m$  in thickness).

Aptel and Semmens (19) modeled the removal of VOCs from water assuming a plug flow reactor with a constant mass transfer coefficient along the length of the fiber

$$-u_L \frac{dC_w}{dx} = K_{oL} A (C_w - C_g^*/H) \quad (2)$$

where  $x$  = linear dimension along the length of HFM unit ( $m$ ),  $A$  = interfacial area of membrane per unit volume in module ( $m^2\ m^{-3}$ ), and  $C_g^*$  = equilibrium concentration of VOC in the gas phase ( $mol\ L^{-1}$ ).

If the vacuum pressure is sufficiently large, it can be assumed that  $C_g^*$  is negligible. Equation 2 can be solved to find the concentration at the outlet,  $C_{w,L}$

$$C_{w,L} = C_{w,o} \exp[-K_{oL} a(L/u_L)] \quad (3)$$

where  $L$  is the actual length of the fibers.

The relationship between the mass transfer coefficients and the operating parameters can be determined from a

dimensional analysis. A dimensionless correlation between the Sherwood ( $Sh$ ), Reynolds ( $Re$ ), and Schmidt ( $Sc$ ) numbers has been proposed (12)

$$Sh_p = q_p Re_p^{r_p} Sc_p^{t_p} \quad (4)$$

where

$$Sh_p = \frac{k_p d_e}{D_p}, \quad Re = \frac{u_p d_e}{\nu_p}, \quad Sc_p = \frac{\nu_p}{D_p}$$

and  $q_p$ ,  $r_p$ , and  $t_p$  are empirical coefficients obtained experimentally for different hollow fibers. The subscript  $p$  refers to either the gas or aqueous phase. Once these empirical coefficients are determined, it is possible to design membranes or membrane systems for the desired MTBE removal efficiency or for any other VOC. Since the membranes are commercialized in modules of fixed length, it may be necessary to link two or more membranes in series to achieve the desired removal efficiency.

The gas-phase diffusivity of MTBE was estimated using the method of Fuller et al. (20), as described in ref 17

$$D_{g,MTBE} = 10^{-7} \frac{T^{1.75} [(1/m_{air}) + (1/m_{MTBE})]^{1/2}}{P [\bar{V}_{air}^{1/3} + \bar{V}_{MTBE}^{1/3}]^2} \quad (m^2\ s^{-1}) \quad (5)$$

where  $T$  = temperature (K),  $m_{air}$  = molecular weight of air (28.97  $g\ mol^{-1}$ ),  $m_{MTBE}$  = molecular weight of MTBE (88.14  $g\ mol^{-1}$ ),  $P$  = gas-phase pressure (atm),  $\bar{V}_{air}$  = average molar volume of the gases in air ( $\sim 20.1\ cm^3\ mol^{-1}$ ), and  $\bar{V}_{MTBE}$  = molar volume of MTBE ( $\sim 119\ cm^3\ mol^{-1}$ ).

The aqueous diffusivity of MTBE was estimated using the method proposed by Othmer and Thakar (21), as presented in ref 17

$$D_{w,MTBE} = \frac{13.26 \times 10^{-9}}{\mu_w^{1.14} (\bar{V}_{MTBE})^{0.589}} \quad (m^2\ s^{-1}) \quad (6)$$

where  $\mu_w$  is the dynamic viscosity ( $kg\ m^{-1}\ s^{-1}$ ) of the aqueous solution, assumed to be equal to the viscosity of water. Both of the methods for calculating diffusivities are expected to produce results within 10% of the experimental values. The diffusivity is a function of temperature, since the viscosity of water varies from  $1.519 \times 10^{-3}\ kg\ m^{-1}\ s^{-1}$  at 0 °C to  $0.653 \times 10^{-3}\ kg\ m^{-1}\ s^{-1}$  at 40 °C; we have fitted the following temperature dependence of  $\mu_w$  based on published data (22):

$$\mu_w = 10^{-3} \exp(-1.1319 - 1,443.9T^{-1} + 521,275T^{-2}) \quad (kg\ m^{-1}\ s^{-1}) \quad (7)$$

Published values of MTBE's Henry's constant vary over an order of magnitude. Based on our recent study (23), we have developed the following correlation for MTBE:

$$H_{MTBE} = \exp\left(9.8077 - \frac{3,346}{T}\right) \quad (-) \quad (8)$$

## Experimental Method

A hydrophobic hollow fiber membrane module (Liqui-Cel 2.5"  $\times$  8", with Celgard X-40 fibers, from Celgard, Inc.) was used in these studies. The hollow fibers have an inside diameter of  $\sim 240\ \mu m$ , a wall thickness of  $\sim 30\ \mu m$ , with pores of  $\sim 0.03\ \mu m$ . The fibers are made of polypropylene, specially extruded to develop a well-characterized porosity ( $\sim 25\%$ ), which allows the transport of organic molecules from the inside of the fiber (aqueous phase) to the outside of the fiber

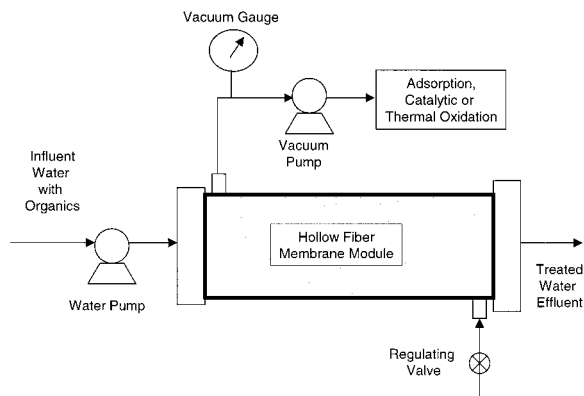


FIGURE 1. Process diagram for pervaporation setup.

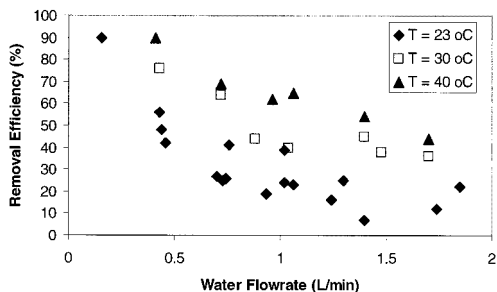


FIGURE 2. Removal efficiency as a function of temperature and flowrate under high vacuum pressure.

(gas phase). The membrane module has inlet and outlet ports for the aqueous and gaseous phases (Figure 2). The module has an overall mass transfer area of  $1.4 \text{ m}^2$ , an effective area to volume of  $292 \text{ m}^2 \text{ m}^{-3}$ , and a fiber length of  $0.203 \text{ m}$ . The packing density in the Liqui-Cel module is very near the jamming limit (24), at around  $435 \text{ fibers/cm}^2$ . The fibers are potted on polyethylene tubesheet, in a polypropylene casing. The flow characteristics of these membrane modules are described in ref 24.

Figure 1 presents the general flow diagram for pervaporation. Water is pumped into the lumen-side of the fibers. Vacuum is applied to the shell-side of the module, and air is allowed to bleed into the module at a controlled rate, as a sweep gas. Due to the hydrophobic nature of the fiber, only trace amounts of water pass through the membrane to the gas phase (25).

Twenty grams of MTBE (> 99% purity, from Fisher Scientific) was added to approximately 200 L of tap water in a closed container and recirculated for 2 h to achieve complete mixing, producing a constant source at about  $100 \text{ mg/L}$  ( $\sim 1.1 \text{ mM}$ ). This concentration is common at underground storage tank remediation sites (1); we expect the membrane system to perform similarly at lower concentrations. Contaminated water was then pumped through the membrane at various flowrates and sampled directly before and after the hollow fiber unit. The treated stream was collected in a waste tank for subsequent treatment if it did not comply with discharge requirements.

The experiments were run at 23, 30, and  $40 \text{ }^\circ\text{C}$ . The spiked water was heated by passing it through copper tubing in a Fisher Scientific Isotemp 220 water bath. Water flowrates ranged from  $0.2$  to  $1.8 \text{ L min}^{-1}$  ( $0.05$ – $0.5$  gallons per minute). Two gas flowrates were evaluated,  $11.3 \text{ L min}^{-1}$  ( $0.4 \text{ scfm}$ ) and  $39.7 \text{ L min}^{-1}$  ( $1.4 \text{ scfm}$ ), which corresponded to vacuum pressures of  $8.1 \times 10^{-4} \text{ Pa}$  ( $0.80 \text{ atm}$ ) and  $5.1 \times 10^{-4} \text{ Pa}$  ( $0.50 \text{ atm}$ ). As more air is allowed to bleed in, the vacuum pressure decreases.

Water samples were collected before and after the HFM unit using 40-mL vials, sealed with a septum-lined cap.

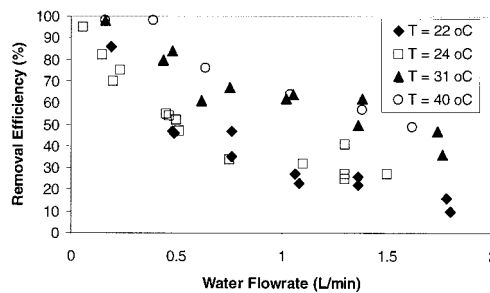


FIGURE 3. Removal efficiency as a function of temperature and flowrate under moderate vacuum pressure.

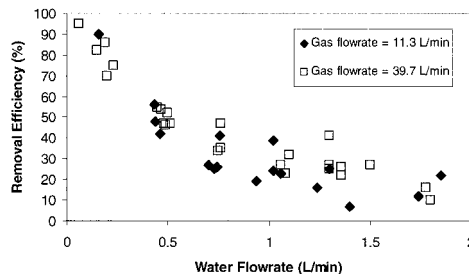


FIGURE 4. Removal efficiency as a function of flowrate at  $23 \text{ }^\circ\text{C}$ .

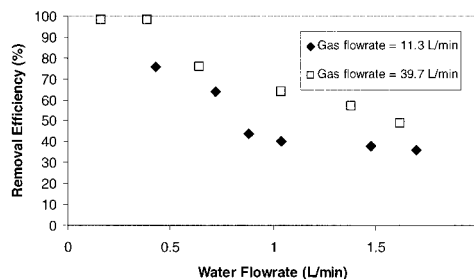


FIGURE 5. Removal efficiency as a function of flowrate at  $30 \text{ }^\circ\text{C}$ .

Samples were extracted almost immediately using a  $100\text{-}\mu\text{m}$  polydimethyl siloxane (PDMS) solid-phase microextraction (SPME) fiber and analyzed in a HP 5890 gas chromatograph with a Supelco VOCOL capillary column coupled to a HP 5970 mass spectrometer. Removal efficiency, defined as  $1 - C_{w,L}/C_{w,0}$ , was calculated for each experimental condition.

## Results

Mass transfer is a strong function of liquid phase flowrate (Figures 2 and 3). Removal efficiency decreases almost linearly with increasing liquid flowrate. Increasing temperature has a positive effect on MTBE's Henry's constant as well as the diffusivities, increasing the removal efficiency. The effect is more significant at the higher flowrates, where a  $10 \text{ }^\circ\text{C}$  increase in temperature can increase the removal efficiency by 10–20%. However, the temperature response is nonlinear, since there appears to be less increase in removal efficiency as the temperature increases from  $30$  to  $40 \text{ }^\circ\text{C}$  than for a  $20$ – $30 \text{ }^\circ\text{C}$  increase.

One of our hypotheses was that vacuum pressure, and the corresponding gas flowrate, would increase mass transfer, as gases are swept more rapidly at the membrane surface, thus increasing the driving force. However, it appears that the effect is small (Figures 4–6) or almost negligible for most conditions. It is probable that the removal of MTBE molecules from the outside surface of the hollow fibers is already fast enough and that the increase in the gradient is small.

The results of the combinations of operating parameters were analyzed using eqs 3–8. The empirical coefficients  $q_p$ ,  $r_p$ , and  $t_p$  were fitted using least-squares analysis. Figure 7 presents a comparison between experimental and theoretical

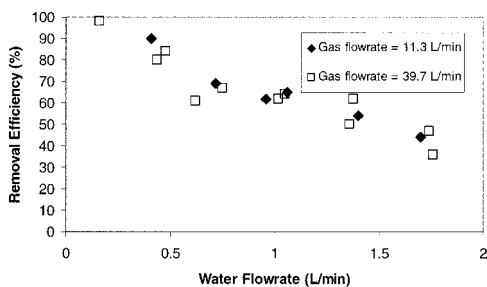


FIGURE 6. Removal efficiency as a function of flowrate at 40 °C.

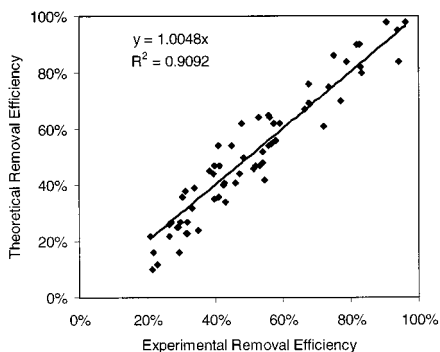


FIGURE 7. Comparison between theoretical and experimental removal efficiency using the Reynolds number correlation (eqs 9 and 10).

removal efficiency using the following empirical correlations for the mass transfer coefficients:

$$Sh_w = 180Re_w^{0.8} Sc_w^0 \quad (9)$$

and

$$Sh_g = 0.001Re_g^0 Sc_g^0 \quad (10)$$

Given eq 6, it is not surprising that  $Sh_w$  is independent of  $Sc_w$ , since neither the diffusivity nor the kinematic viscosity varies significantly over this range of temperatures. The comparison between experimental and theoretical removal efficiency indicates that eq 9 does a good job at the higher removal efficiencies (> 35%) but tends to underestimate removal at low removal efficiencies. This result has been observed for the tube-side (water in lumen) mass transfer coefficient by several other studies, where it is the Lévêque correlation (24) that has been successfully applied to predict the tube-side mass transfer coefficient:

$$Sh_w = 162(d_e^2 u_L / LD_w)^{1/3} \quad (11)$$

Our analysis using eqs 10 and 11 to estimate the overall mass transfer coefficient indicates that eq 11 does not provide a better prediction of the mass transfer coefficient (Figure 8), but that eq 9 is not significantly better. A better fit is obtained by modifying the coefficients in the Lévêque correlation to

$$Sh_w = 79(d_e^2 u_L / LD_w)^{0.79} \quad (12)$$

although eq 12 still does not provide a better fit to our experimental results than eq 9.

The gas-phase resistance of MTBE is minimal under pervaporation conditions, and thus  $Sh_g$  is independent of gas flowrate or gas-phase properties, as seen in eq 10. This might not be the case for less soluble or more volatile VOCs (i.e. with larger  $H$ ). Previous work (8, 14) indicated some dependence on the gas-phase resistance for VOCs with larger

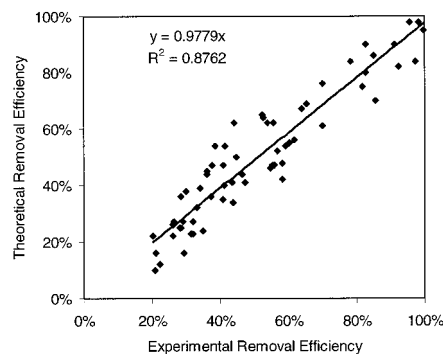


FIGURE 8. Comparison between theoretical and experimental removal efficiency using Lévêque's equation for the tube-side (water) mass transfer coefficient.

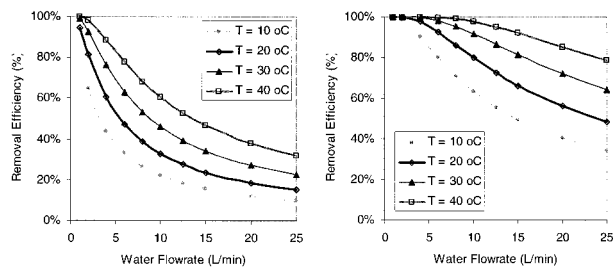


FIGURE 9. Predicted performance for commercial membrane: (a) one unit and (b) four units in series.

$H$  and for the case where a sweep gas is used in the shell side rather than applying a vacuum.

Based on these results, we have estimated the removal efficiency of a larger HFM module available from Liqui-Cel. The 4" × 28" module has an effective surface area of 19 m<sup>2</sup> or over an order of magnitude greater than the 2.5" × 8" module. Since the type of hollow fibers is the same in both units, eqs 9 and 10 are applicable to the larger unit. Given the tight packing of the hollow fibers in these units, the flow characteristics are expected to be quite similar. Figure 9 presents the expected removal efficiency of the larger unit as a function of flowrate and temperature, for one unit (Figure 9a) and for four units in series (Figure 9b). Considering that the total footprint of the four-membrane system, including pumps, membranes, and control panel is approximately 1 m<sup>2</sup> and a height of 1.8 m, this setup would certainly have advantages over a comparable air stripping unit. In addition, a comparable air-stripping unit operating at 19 L min<sup>-1</sup> (~5 gallons per minute) and with similar removal efficiency for MTBE would require at least 1900 L min<sup>-1</sup>, generating a significant amount of contaminated air, which might require treatment; the tower height would be > 5 m. The modularity of the membrane system allows for scaling to the required flowrate and removal efficiency.

Hollow fiber membrane technology has made significant advances in certain fields but has not generally been incorporated into the environmental field, due to economic reasons as well as a general lack of knowledge of their applicability. The treatment of compounds such as MTBE requires novel approaches, and HFM may be a cost-effective solution for addressing this challenge. Our results indicate that the removal efficiency of MTBE using HFM is significant.

Knowledge of the effects of operating parameters is important. We have evaluated a significant range of operating conditions to determine the relationship between the mass transfer coefficients and the controlling parameters. While liquid flowrate and temperature play a significant role, gas flowrate and the related vacuum pressure play a very minor role under these conditions. This also reflects the fact that

for MTBE and similar compounds, the liquid-phase resistance controls mass transfer through the membrane to the gas phase.

These results can be used to scale-up to field-scale systems. Since hollow fiber characteristics will have some effect on the mass transfer coefficient, it is important to determine experimentally the relationship between the fiber characteristics and the mass transfer coefficient for each type of fiber. Results from the same fiber can be scaled to different membrane module sizes.

One of the limitations of membrane technologies in general, and HFM in specific, is that capital costs do not scale well; the capital costs increase almost linearly with flowrate. Thus, HFM is most effective for lower liquid flowrates (<300 L min<sup>-1</sup>). There is also an issue with scaling, when water with considerable hardness is treated, since the carbonates and iron oxides will form precipitates. This can be controlled by metering a low-volatility acid solution before the inlet to the HFM modules.

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