Reduction of VOC emissions by a membrane-based gas absorption process

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Abstract

A membrane-based gas absorption (MGA) process was evaluated for the removal of volatile organic compounds (VOCs) based on C_6H_6/N_2 mixture. The absorption of C_6H_6 from a C_6H_6/N_2 mixture was investigated using a hydrophobic polypropylene hollow fiber membrane contactor and the aqueous solution of N-formyl morpholine (NFM) as absorbent. The effects of various factors on the overall mass transfer coefficient was investigated. The experimental results showed that the removal efficiency of C_6H_6 could reach 99.5% in present studied system. A mathematical model based on resistance-in-series concept was presented to predict the value of overall mass transfer coefficient. The average error between the predicted and experimental values is 7.9%. In addition, conventional packed columns for VOCs removal was also evaluated for comparison.

Key words: VOCs removal; membrane contactor; membrane-based gas absorption; mass transfer model

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Introduction

Volatile organic compounds (VOCs) released from the chemical and industrial processes can cause major hazards to the environmental and public health. Thus, the tighter regulations were established in many countries for the removal of VOCs from the emissions prior to discharging into air (Sohn *et al.*, 2000; Liu *et al.*, 2006). Several techniques are available for VOCs removal including adsorption, absorption, advanced oxidation, incineration and biological treatment (Guizard *et al.*, 2001; Kim and Kim, 2005). However, most of those techniques not only require high capital and operational costs and the limitation of the feed condition in actual application, but also produce a problem of post-treatment (Kim *et al.*, 2002). During last several years, vapor permeation (VP) technique is well established for VOCs removal from exhaust gas. This process is based on silicone-rubber membrane which is highly selective for VOCs over inert gases. VP is likely favored over other processes for smaller feed gas flow rates and not-too-low VOCs concentration in air (Obuskovic *et al.*, 2003). Such VP processes are, however, not very efficient in bringing down the VOCs concentration in the treated gas to very low levels (Poddar and Sirkar, 1997). This greatly limits the potential applications of VP technique in VOCs treatment.

Zhang and Cussler (1985a, 1985b) firstly introduced membrane-based gas absorption (MGA) process in the application of CO_2 removal. Since then, the MGA process has been widely developed in the application of acid gas removal (Bhaumik *et al.*, 1998; Dindore *et al.*, 2004; Wang *et al.*, 2006). The MGA process is based on a gas-liquid contact across hydrophobic microporous membrane. The membrane acts as a physical barrier between the liquid and the gas phases, and the fibers used in the contactor are generally not selective, the absorbent offers the selectivity instead (Li and Chen, 2005). Poddar *et al.* (1996a, 1996b) adopted MGA process for the removal of VOCs from gas steams using paratherm and silicone oil as absorbents. It was found that VOCs concentration could be reduced to remarkably low level at the medium or low VOC concentrations. Moreover, MGA process can avoid the problems, such as flooding, loading, entraining and channeling which often encountered in the conventional absorption devices, and these membrane contactors are operational flexibility, have high mass transfer rates and compactness, and can be easily scaled up (Al-Marzouqi *et al.*, 2008; Zhang *et al.*, 2008).

In present study, a hydrophobic polypropylene hollow fiber membrane contactor was accessed for C_6H_6 removal from N_2 using the aqueous solution of N-formyl morpholine (NFM) as absorbent. The effects of various factors, such as flow rate of feed gas and absorbent, concentration of C_6H_6 in feed gas and absorbent, on the C_6H_6 concentration at outlet and the overall mass transfer coefficient k_{ov} were investigated. A mathematical model was developed using the resistance-in-series concept combining with process conditions, membrane properties and module geometric characteristics. The removal efficiency and the height of a transfer unit (HTU) of conventional packed contactor were also investigated for comparison.

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1 Materials and methods

1.1 Theory

For the non-wetted mode of MGA process, mass transfer was determined by the consecutive steps in three phases (Fig. 1), including the diffusion of gaseous component i from bulk gas to membrane wall, the diffusion through the pores of the membrane to the membrane-liquid interface, and the dissolution into liquid absorbent, followed by liquid phase diffusion (Mavroudi *et al.*, 2006).

For gas-liquid membrane contacting process, the resistance-in-series model was used to describe the mass transfer mechanism (Lu *et al.*, 2008). The overall mass transfer resistance, $1/k_{ov}$, can be expressed as Eq. (1).

$$1/k_{ov} = H/k_g + H/k_m + 1/k_l \qquad (1)$$

where, k_g, k_m, and k_l are individual mass transfer coefficients of gas phase, membrane and liquid phase, respectively. H is Henry law constant.

Many correlations have been proposed to determine the mass transfer coefficient of shell side and tube side. In general, it can be expressed as Eq. (2):

$$Sh = aRe^b Sc^c \qquad (2)$$

where, Sh, Re, Sc are Sherwood, Reynolds, Schmidt numbers, respectively, a, b and c are constants. In this study, the mass transfer coefficients k_g and k_l are calculated as the Eqs. (3) and (4).

$$Sh_g = \frac{k_g d_i}{D_{i,g}} = 1.62(\frac{d}{L} Re_g Sc_g)^{0.33} \qquad (3)$$

$$Sh_l = \frac{k_l d_e}{D_{i,l}} = 5.85 (1 - \phi) (\frac{d_e}{L}) Re_l^{0.6} Sc_l^{0.33} \qquad (4)$$

where, d_i is the inside diameter of fiber, d_e is the equivalent diameter of the membrane contactor, $D_{i,g}$ and $D_{i,l}$ are diffusion coefficient in gas side and liquid side, and determined by the following Eqs. (5) and (6), respectively.

$$D_{i,g} = \left(1 \times 10^{-7} T^{1.75} \left(\frac{1}{M_{C_6H_6}} + \frac{1}{M_{N_2}}\right)^{\frac{1}{2}}\right) \bigg/ \qquad (5)$$
$$P\left(\left(\sum \upsilon_{C_6H_6}\right)^{\frac{1}{3}} + \left(\sum \upsilon_{N_2}\right)^{\frac{1}{3}}\right)^2$$

$$D_{i,l} = (7.4 \times 10^{-8}(fM)^{0.5}T) \big/ (\mu_1 V_{bA}^{0.6}) \qquad (6)$$

where, the values of correction factor f, viscosity μ_1, molar volume V_{bA}, critical volume $\sum \upsilon_{C_6H_6}$ and $\sum \upsilon_{N_2}$ are obtained from literature (Poling *et al.*, 2006).

For non-wetted microporous membrane, the pores are gas filled, the membrane coefficient k_m can be independently evaluated with their property of pore structures, which include porosity ε, thickness δ and tortuosity τ.

$$k_m = D_m \frac{\varepsilon}{\delta \tau} \qquad (7)$$

where, D_m is the coefficient of effective membrane diffusion; it is a combination of bulk coefficient $D_{i,g}$ and Knudsen diffusion coefficient D_k.

$$1/D_m = 1/D_{i,g} + 1/D_k \qquad (8)$$

For gas-filled pores, the diffusion through membrane can be either bulk diffusion or Knudsen diffusion depending on pore diameter (d_p) (Li and Chen, 2005). If $d_p < 1 \times 10^{-7}$ m, Knudsen diffusion was dominant; if $d_p > 1 \times 10^{-5}$ m, bulk diffusion governs; if 1×10^{-7} m $< d_p < 1 \times 10^{-5}$ m, both types of diffusion exist.

The bulk diffusion coefficient can be calculated by Eq. (6), and the Knudsen diffusion coefficient can be determined by Eq. (9).

$$D_k = \frac{4\varepsilon d_p}{3\tau} \sqrt{\frac{8RT}{\pi M}} \qquad (9)$$

Based on the experimental results, k_{ov} can be expressed by Eq. (10).

$$k_{ov} = \frac{V_l(C_{l,out} - C_{l,in})}{A_T \Delta C_m} \qquad (10)$$
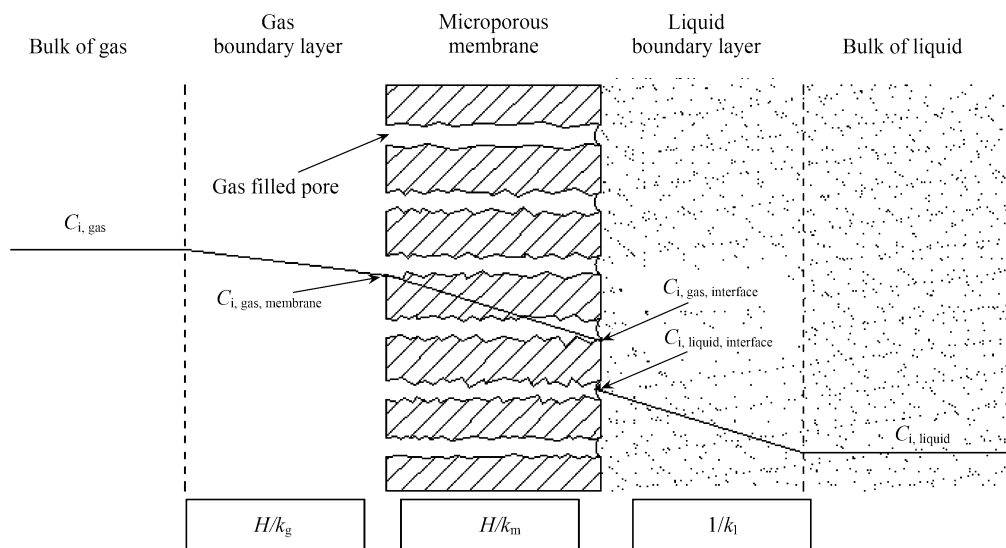

Fig. 1 Mass transfer regions and resistance-in-series in non-wetted membrane contactor.

where, V_1 is the volumetric flow rate of liquid phase, and A_T the mass transfer area based on the surface of gas-liquid contact. $C_{l,in}$ and $C_{l,out}$ are C_6H_6 concentrations at liquid inlet and outlet, respectively. The logarithmic mean concentration difference based on liquid phase concentrations can be calculated as Eq. (11).

$$\Delta C_m = \frac{(HC_{g,in} - C_{l,out}) - (HC_{g,out} - C_{l,in})}{\ln(HC_{g,in} - C_{l,out})/(HC_{g,in} - C_{l,out})} \tag{11}$$

where, $c_{g,in}$ and $c_{g,out}$ are C_6H_6 concentrations at gas inlet and outlet, respectively.

1.2 Materials and experimental

The absorption experiments were carried out with polypropylene microporous hollow fiber membrane contactor (Hangzhou Kaihong Membrane Technology Co., Ltd., Zhejiang, China). Specifications of the hollow fiber membrane modules used in this study are as follows: fiber diameter inside/outside 200 μm/400 μm, average pore diameter 0.2 μm, average length 30 cm, shell ID 3 cm, the number of fibers 2000, and porosity 40%. N_2 is commercially cylinder gas and the purity is more than 99.99% (Nanjing Real Special-gas Co., China). The purity of NFM was 99.9% (Huai'An Huatai Chemical Co., Ltd., Jiangsu, China).

Figure 2 illustrates the schematic diagram of experimental setup for VOC absorption experiments in membrane contactor. N_2-containing benzene (feed gas) was prepared in a gas-prepared system to a given concentration in advance and passed through the lumen side of the membrane contractor at a predetermined controlled rate. The gas flow rate was controlled by mass flow controllers (Beijing Seven Star Electronics Co., Ltd., China). Fresh absorbent was bumped into the shell side of the contactor in which mass transfer from gas phase to liquid phase occurred. The absorbent exiting on absorb-membrane was connected to a heater, and hot absorbent passed through the lumen

side of the regeneration membrane. A vacuum pump was connected with the regeneration membrane module via a condenser. Regenerated absorbent from the regeneration membrane module was recycled to the absorbent vessel for reuse. Both inlet and outlet gas samples were analyzed on-line using an Agilent 6820 gas chromatograph. The concentration of benzene in the absorbent was determined using headspace technique.

In present experiments, gas flow rates were varied from 40 to 300 mL/min, and liquid flow rates were in the range 20–100 mL/min. All data were obtained after 30–60 min operation when system was stabilized. Steady state was indicated by a constant C_6H_6 concentration in the exiting gas stream.

2 Results and discussion

2.1 Effect of operating conditions

2.1.1 Liquid flow rate

The dependence of concentration of C_6H_6 in gas outlet, expressed in a dimensionless form $C_{g,out}/C_{g,in}$, on the liquid flow rates is shown in Fig. 3a. It was observed that the amount of C_6H_6 in gas outlet decreased with an increase of liquid flow rate (V_1). An increase of the liquid flow rate resulted in lower liquid phase mass transfer resistance and, hence, in more efficient gas removal. This effect was more pronounced at higher gas flow rate. For higher gas flow rate, equivalent removal efficiency could be achieved if a higher volume module is used providing more gas-liquid contact area. The MGA process shows an excellent performance in terms of mass transfer for the VOCs removal from gas mixture (Mavroudi *et al.*, 2003).

Figure 3b illustrates the effect of liquid flow rate in shell side on the overall mass transfer coefficient. As the liquid flow rate (V_1) increasing, the overall mass transfer coefficient (k_{ov}) increased as well. The increased of liquid
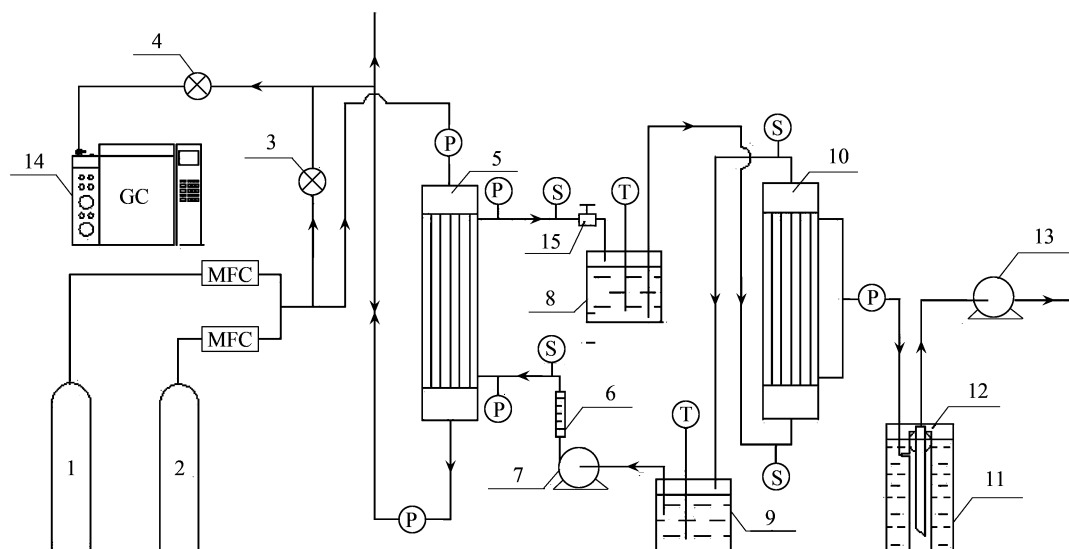

Fig. 2 Schematic diagram of experimental setup for VOC absorption experiments in membrane contactor. (1) mixed-gas cylinder; (2) N_2 cylinder; (3) and (4) electromagnetic valve; (5) absorption membrane contactor; (6) liquid flow meter; (7) liquid pump; (8) and (9) absorbent tanks with heater; (10) absorbent regeneration membrane contactor; (11) condensator; (12) gas-liquid separator; (13) vacuum pump; (14) gas chromatography; (15) back pressure value. S: sample point of liquid; P: pressure transducer; T: temperature transducer.
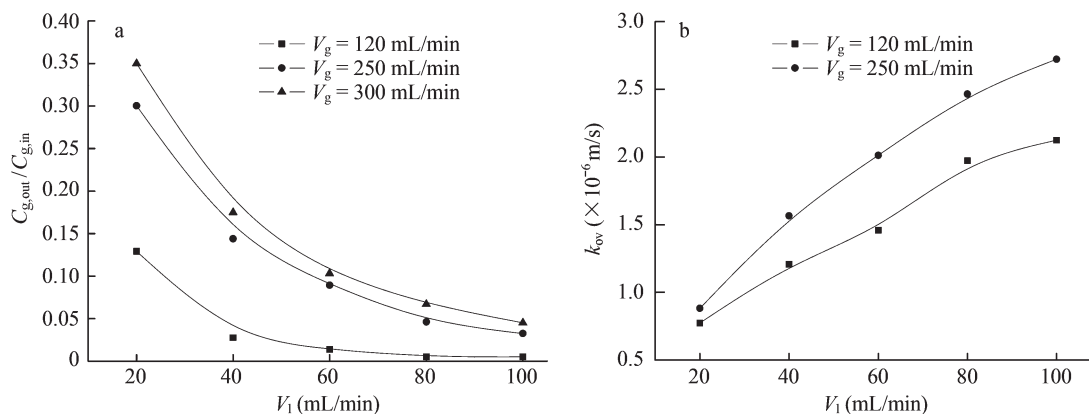
Fig. 3   Effects of liquid flow rates (V_1) on the outlet C_6H_6 concentration (a) and the overall mass transfer coefficient (k_{ov}) (b).

flow rate can lead to the increase of C_6H_6 concentration gradient, and thus improve the diffusion of C_6H_6 from gas to liquid phase. Known from film theory, mass transfer resistance of liquid phase dominated the process of mass transfer, thus, the thickness of liquid-film decreased with increasing liquid flow rate, which contributed to the decrease of mass transfer resistance on liquid phase and the enhancement of mass transfer of C_6H_6.

2.1.2 Gas flow rate

The concentration of C_6H_6 in gas outlet increased with increasing gas flow rate (Fig. 4a). This was obviously owing to smaller residence time of gas at higher gas flow rates, which reduced the diffusion rate of C_6H_6 through the membrane (Park *et al.*, 2008). Removal efficiency (($C_{g,in} - C_{g,out})/C_{g,in}$) was one of the separation properties of MGA process. In this study, the removal efficiency of C_6H_6 was as high as 99.48% at volume ratio of NFM 40%, flow rate of absorbent 100 mL/min, and flow rate of gas 40 mL/min. Figure 4b shows that the overall mass transfer coefficient was slightly increased with the increase of gas flow rate. Known from film theory for physical absorption, the mass transfer resistance is dominated in liquid film and the resistance from gas film is neglectable. Because the liquid flow rate was a constant in these experiments, the thickness of liquid film was almost constant, and thus the

overall mass transfer coefficient was almost unchanged. The experiment results were in better agreement with theoretical analysis.

2.1.3 Absorbent concentration

NFM is an excellent industrial solvent for aromatic substance extraction and have high dissolvability as well as thermal stability. Moreover, the hydrophobic membrane fibers are hard to be wetted by NFM, which can effectively decrease the membrane resistance in the mass transfer process. As shown in Fig. 5, the overall mass transfer coefficient increased with increasing absorbent concentration. According to film theory, for mass transfer across a membrane interface, the absorption of gas occurs in a zone locate at some points in the liquid-film, so the higher equilibrium concentration in the gas-liquid interface based on Henry's Law is obvious at higher concentrations of the absorbent, which can obviously improve the absorption of C_6H_6.

2.1.4 C_6H_6 concentration in feed gas

A series of experiments were carried out to examine the effect of inlet C_6H_6 concentration on the overall mass transfer coefficient. The overall mass transfer coefficient slightly increased with increasing C_6H_6 concentration in feed gas $C_{g,in}$ (Fig. 6). In the range of 0.79–10.85 mg/L,
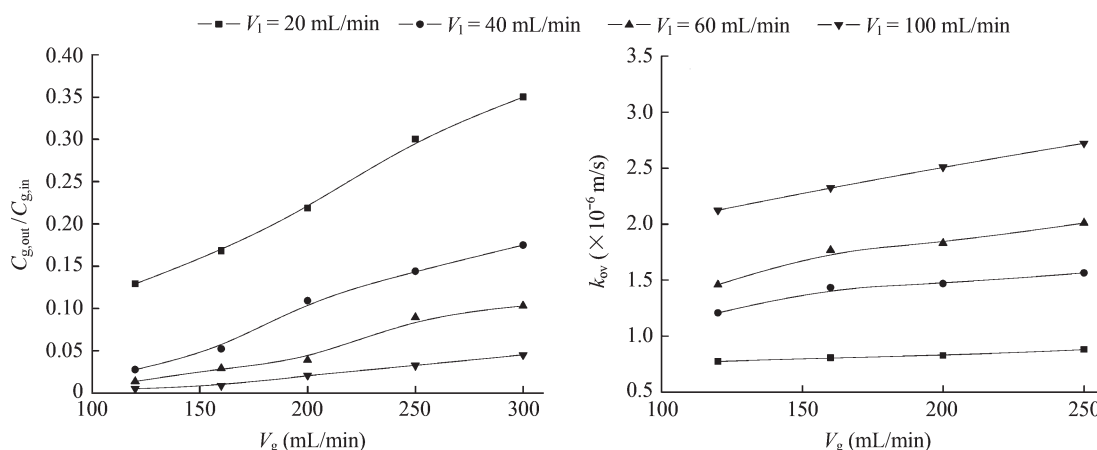

Fig. 4   Effect of gas flow rates (V_g) on the outlet C_6H_6 concentration (a) and the overall mass transfer coefficient (k_{ov}) (b).
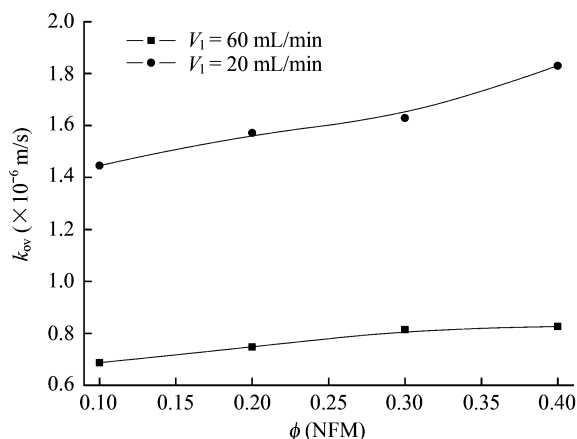
Fig. 5 Effect of absorbent concentration (ϕ) on the overall mass transfer coefficient (k_{ov}).
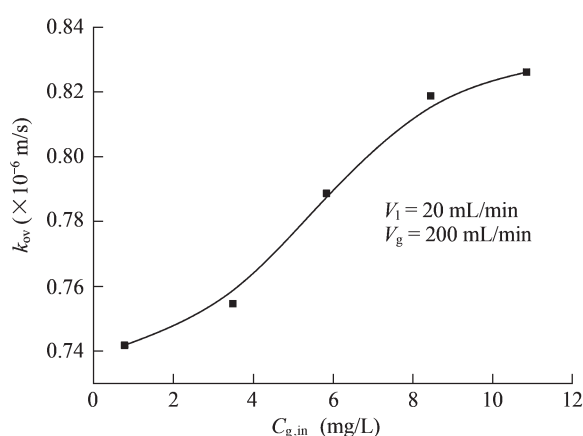

Fig. 6 Effect of inlet C_6H_6 concentration ($C_{g,in}$) on the overall mass transfer coefficient (k_{ov}).

the value of the overall mass transfer coefficient increased average near 11.36%. It was mainly because that the increase of $C_{g,in}$, i.e., the increase of C_6H_6 partial pressure of gas phase, could improve the physical solubility of C_6H_6 in the solution, and the accumulation of C_6H_6 concentration increased in liquid interface as well. Because the C_6H_6 concentration in the liquid interface was much lower than C_6H_6 saturated concentration in the absorbent for low $C_{g,in}$, the increase of C_6H_6 concentration in liquid interface could improve the diffusion of C_6H_6 from liquid film to liquid bulk, which lead to an increase of the overall mass transfer coefficient. When the C_6H_6 concentration in the liquid interface was achieved saturation, however, the liquid mass transfer resistance might not be neglected. Analyzing the data from Fig. 6, when $C_{g,in}$ reached 10.85 mg/L, the increase gradient of the curves was smaller than those of low $C_{g,in}$, which can be explained that C_6H_6 concentration in the liquid interface was in approximate saturation. It can be expected that the value of the overall mass transfer coefficient will decrease with the further increase of $C_{g,in}$.

2.2 Comparison of the overall mass transfer coefficient obtained from model and experiment

The values of the overall mass transfer coefficient obtained from model are plotted with those of experiment. In this study, model prediction value was in better agreement
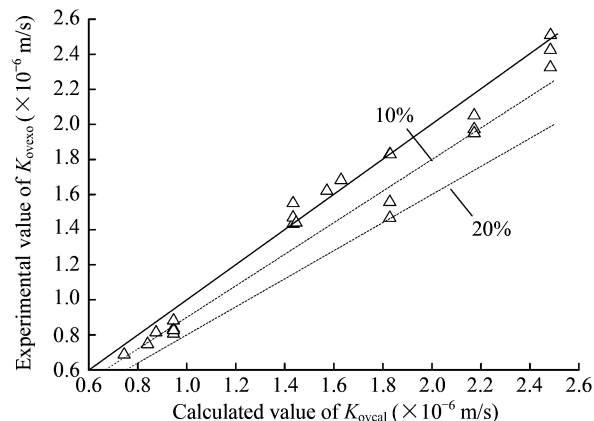

Fig. 7 Comparison of model and experimental k_{ov} values.

with experimental value with the average error 7.9%. As can be seen in Fig. 7, most of model values (k_{ovcal}) are higher than those experimentally measured (k_{ovexo}), especially for higher gas and liquid flow rates. An additional mass transfer resistance may be considered as a probable reason of this deviation (Mavroudi *et al.*, 2003). The trace impurities and long term operation of membrane are presumed to modify the surface hydrophobicity, and thus allow some penetration of liquid into the pores, resulting in higher mass transfer resistance (Rangwala, 1996). Previous studies showed that even 13% pores were penetrated by liquid, the resistance of the liquid filled pores accounted for over 98% of the membrane resistance and for 20%–50% of the total resistance to absorption (Mavroudi *et al.*, 2006). Because the model was conducted in unwetted condition, the difference between model and experimental data will be caused.

2.3 Comparison of η and HTU in membrane process and packed contactor

In order to evaluate the efficiency of membrane-based gas absorption process, the absorption performance of MGA process was compared with that of a conventional packed contactor. The classical measure of absorption technology efficiency, i.e., the height of a transfer unit (HTU), can be calculated as Eq. (12) (Yang *et al.*, 2006):

$$\text{HTU} = \frac{V}{k_G a} \tag{12}$$

where, V is gas velocity, $k_G a$ is overall volumetric mass transfer coefficient based on gas phase.

The required HTU for physical absorption of C_6H_6 is plotted as a function of liquid flow rate for the MGA process and packed contactor with Raschig rings of diameter d 10 mm (Fig. 8a). Under the same operating conditions, HTU values for conventional packed contactor (12.19–33.90 cm) were significantly higher than that for MGA process (7.16–13.85 cm). A low HTU_G value indicated high efficiency for absorption. Moreover, the residence time in the membrane contactor varied from 0.063 to 0.47 min. The residence time in membrane contactor is about 12 times smaller than that in conventional packed contactor, e.g., 0.71–5.30 min. In case of membrane contactor, due to high surface area per volume maintained at any flow rate of
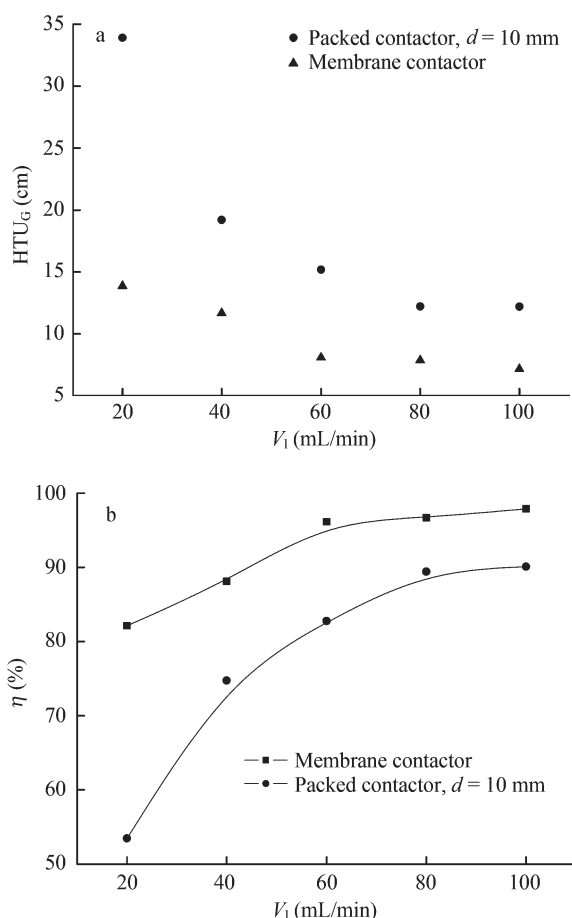
Fig. 8 Comparison of HTU (a) and removal efficiency (b) of MGA and packed contactor for the C_6H_6 absorption in N-formyl morpholine (NFM).

the gas and liquid phases, the rate of mass transfer is relatively high comparing with conventional packed contactor (Ghosh *et al.*, 1994). Thus, even with shorter residence time, gas is absorbed more efficiently and need smaller reactor to achieve the comparable efficiency in packed contactor. As shown in Fig. 8b, the experimental values of removal efficiency were in the range of 82.7%–97.9% for the membrane contactor and 53.5%–90.1% for the packed contactor. It is expected that MGA process would be useful for the separation of VOCs from industrial gas streams because of lower specific area requirements, lower investment cost and higher removal efficiency comparing with packed contactor.

3 Conclusions

Membrane-based gas absorption using hydrophobic hollow fiber membrane contactor can be used to reduce VOC gas emissions. Present experimental results show that high levels of C_6H_6 removal (up to 99.5%) can be achieved with the aqueous solution of NFM. To simulate the performance of MGA process, a mathematical model based on resistance-in-series concept is presented. The model takes into account mass transfer at gas, liquid and membrane interfaces. Comparison of model estimations with experimental results shows that the value of overall mass transfer

coefficient based on experimentally measured are much lower than those theoretically calculated for a completely non-wetted pore, it is mainly because that the model predictions are not taking into account membrane mass transfer resistance caused by some possible penetration of liquid into the membrane pores. Compare to conventional techniques, MGA process offer high specific surface area and, therefore, the calculated HTU values under the same operating conditions are significantly smaller. It is proposed as an alternative to more conventional techniques for removal of VOCs from exhaust gas.

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