

Separation and Purification Technology 22-23 (2001) 377-382



www.elsevier.com/locate/seppur

Clean technology for acetone absorption and recovery

E. Márki *, B. Lenti, Gy. Vatai, E. Békássy-Molnár

Department of Food Engineering, Szent István University, Ménesi út. 44, H-1118 Budapest, Hungary

Abstract

This paper concerns the removal of acetone from air by absorption in water in a packed column and recovery of acetone from water by pervaporation from its dilute aqueous solution. The measurements were carried out in a laboratory scale absorption column and a laboratory scale pervaporation unit. The absorption with Sulzer-packing was a suitable process for the removal of acetone vapours: a satisfactory removal efficiency was achieved. Using a CMG OM-10 pervaporation membrane from Celfa high fluxes and separation factors were obtained for the removal of acetone from water. Models were formulated to determine the most effective operating parameters which minimize the costs. According to the experiments and calculations the proposed absorption—pervaporation system can be technically feasible and economically attractive method for vapour absorption and recovery. © 2001 Elsevier Science B.V. All rights reserved.

Keywords: Clean technology; Acetone; Absorption-pervaporation system

1. Introduction

1.1. Removal of organic vapours from air by absorption

The industrial procedures involve scrubbing of waste gases with reagents that enhance the absorption. The application of recovery of vapours of volatile organic compounds from air is generally targeted at pollution control, solvent recovery, and organic concentration for special processes. The widely used absorption apparatus are packed columns [1]. Structured and non-structured packings could be used for mass transfer processes.

E-mail address: emarki@omega.kee.hu (E. Márki).

The mass transfer coefficient characterizes the transport rate of the certain component. The Sherwood equation [1] is used to describe the mass transfer in case of absorption:

$$Sh_{G} = \frac{k_{G}d_{eq}}{D_{G}} = aRe_{G}^{b}Sc_{G}^{c}$$
 (1)

where Sh_G is the Sherwood number of the gas phase, k_G is the gas side mass transfer coefficient, d_{eq} is the equivalent diameter of the packing, D_G is the diffusional coefficient, Re_G is the Reynolds number of the gas phase, Sc_G is the Schmidt number of the gas phase and a, b, and c are constants. The values b and c depend only on flow regime and not on fluid properties; generally b = 0.8 and c = 0.333 for packed towers for turbulent flow regime [1].

^{*} Corresponding author. Tel.: +36-1-3726232; fax: +36-1-3726323.

1.2. Recovery of organics from water

Aqueous solutions of organics may be difficult to separate because of azeotropic points, the relatively high diffusivity and solubility of the solvents, and their associated health risks. There are many possible alternative methods to solve this problem, which include air stripping, carbon adsorption, biological treatment, stream stripping and incineration. Pervaporation is economically viable alternative over a wide range of concentration and type of organic material [2].

Pervaporation is a membrane process which can separate a mixture of two or more miscible liquids into more concentrated products of the constituents. The analogous conventional non-membrane based process is distillation, although the mechanism of separation is quite different.

The process of pervaporation can quite selectively recover the organic species [3–5]. In the pervaporation process, a membrane is used to selectively separate solvents by reducing the vapour pressure on the permeate side of the membrane. For the separation of dilute organics from water, a resistance-in-series model is used to describe the transfer of water and organic from the bulk liquid to the permeated vapour [6]. Because organics are extracted preferentially from dilute aqueous solutions, the concentration polarization at the liquid-membrane interface can control the mass transfer rate of the organics.

The choice of membrane for a specific separation is a critical factor in the overall efficiency of the process. The separation performed by a pervaporation can be caracterized by the separation factor, defined as:

$$\alpha_{ij} = \frac{C_i^{\text{perm}}/C_j^{\text{perm}}}{C_i^{\text{feed}}/C_i^{\text{feed}}} \tag{2}$$

where C_i^{feed} and C_j^{feed} (% of weight) are the concentrations of the components i and j in the feed solution, and C_i^{perm} and C_j^{perm} (% of weight) mean the concentrations of the components in the condensed permeate stream.

Another important parameter to characterize the membrane is the flux of permeate:

$$J_{\rm P} = \frac{m_{\rm P}}{F\tau} \tag{3}$$

where $J_{\rm P}$ is the flux of permeate (kg/(m² h)), $m_{\rm P}$ is the mass of the permeate (kg), F means the surface of the membrane, and t is a period of time (h).

For the removal of low concentration organics from water by vacuum-pervaporation, organophylic nonporous membrane have to be applied, which exhibits high flux for organic compound and relatively low flux of water.

2. Experimental

2.1. Absorption in a packed column

The absorber used is shown schematically in Fig. 1. A glass column of 0.1 m diameter and 1 m height, packed with Sulzer Mellapak 250.Y structured packing was used. The storage tank of the water as absorbent was connected with a thermostat to adjust liquid temperature, and a peristaltic pump was installed. The air was transported by ventilator and entered into the bottom of the column. The vapours of thermostated acetone were stripped into the air and mixed by static mixer before entering. The concentration of the inlet air was adjusted by changing the stream of stripping air and the

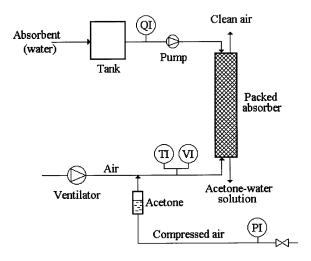


Fig. 1. Schematic diagram of the absorption apparatus.

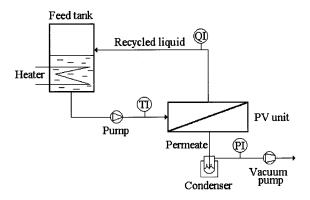


Fig. 2. Flow diagram of the laboratory pervaporation unit.

temperature of acetone $(T_{\rm F})$. The value of the superficial gas velocity $(v_{\rm G})$ was between 0.2 and 1 m/s, the liquid flow-rate was L=2000 to 3050 $1/({\rm m}^2~{\rm h})$. These experimental conditions were below the flooding point of the column [7]. The concentration of the inlet air $(C_{\rm G,in})$ was approximately 20000 ppm (by volume).

The mass balance was checked by analysing the gas and liquid output. In some cases repeated measurements were carried out to check the reproducibility of the measurement.

The concentration of the inlet $(C_{G,in})$ and outlet air $(C_{G,out})$ were determined by hydrogen flame ionized detector produced by HITEKA Ltd., Hungary. The acetone content of the outlet water (C_{L}) were determined by density measurements using Gibertini apparatus (standard error \pm 0.05% acetone).

The removal efficiency was defined as follows:

$$E = \frac{C_{G,in} - C_{G,out}}{C_{G,in}} \times 100 \%$$
 (4)

where E is the removal efficiency (%), $C_{G,in}$ is the concentration of the inlet air (% of weight), and $C_{G,out}$ is the concentration of the outlet air (% of weight).

The removal efficiency was investigated in function of superficial gas velocity (v_G) and liquid flow rate (L).

2.2. Laboratory scale pervaporation system

The pervaporation system used is illustrated in

Fig. 2. A peristaltic pump was used to recirculate the solution with required temperature from the feed tank through the module. The liquid which had $\sim 1-5\%$ of weight of acetone entered the cell perpendicular to the surface of the membrane and then run parallel with the membrane. The feed flow was assumed to be laminar at $L_{\rm R} = 50-250$ l/h. The temperature of the feed was changed from 25 to 65°C.

In this application, flat sheet organofil membrane, CMG OM-10 type from Celfa was used. The membrane surface area was 180 cm². The pressure of the permeate side was 10 mbar. Liquid nitrogen was used to condense vapours.

Effects of feed temperature $(T_{\rm F})$ and the liquid flow rate $(L_{\rm R})$ were examined. An experimental design illustrated in Table 1 was followed.

3. Results and discussion

3.1. Results of the experiments with absorption column

The concentration of the outlet air was between 5000 and 10000 ppm. The acetone content of the outlet liquid was 4–4.2% of weight. The effect of superficial gas velocity on the removal efficiency is illustrated in Fig. 3. The value of removal efficiency was in the range of 32–83%.

Table 1 Experimental design of operating parameters in pervaporation of acetone—water system by Celfa CMG OM-10 membrane

Operational parameters		Operational parameters (standardized factors)	
$T_{\rm F}$ (°C)	$L_{\rm R}~({ m L/h})$	$T_{\text{F(stand)}}$	$L_{ m R(stand)}$
25	50	-1	-1
25	250	-1	+1
45	150	0	0
65	50	+1	-1
65	250	+1	+1

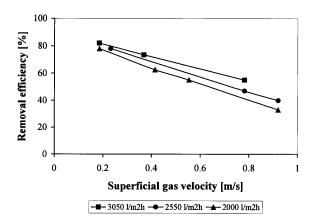


Fig. 3. Influence of temperature on removal efficiency of the absorption tower packed with Sulzer Mellapak 250.Y.

On the basis of experiments a simple correlation was created for removal efficiency:

$$E = 12.17v_{G}^{-0.41}L^{0.16} \tag{5}$$

This means the removal efficiency increases when superficial gas velocity decreases, thus residence time increases, and increases when liquid flow-rate increases. The validity of the Eq. (5) is: $v_{\rm G} = 0.2-1$ m/s and L = 2000 to 3050 l/(m² h).

The error of mass balance was no more than 20% which is satisfactory in case of mass transfer procedures.

The optimal operating parameters for absorption were: 0.4 m/s superficial gas velocity and 950 l/(m² h) (600 l/h) of water flow.

3.2. Results of the experiments of pervaporation

On the base of batch experimental results it can be concluded that the permeation rate reduces strongly exponentially with time. Because of this change the fluxes were determined after 2 h.

Change of total permeate flux (J_P) in function of liquid flow rate and temperature is illustrated in Fig. 4. The permeate flux reached 1.03 kg/(m² h) while the acetone content of the permeate was between 50 and 58% of weight.

An increase in feed temperature (T_F) increased the flux (J_p) exponentially (following the Arrhenius equation). As the liquid flow rate (L_R) was increased, the flux increased slightly.

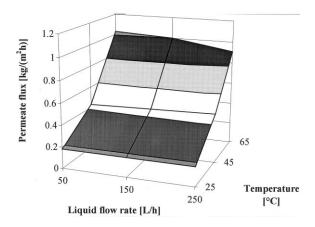


Fig. 4. Influence of temperature and liquid flow rate on total flux of Celfa CMG OM-10 membrane.

The separation factor (α) were calculated using Eq. (2). The effect of temperature and liquid flow rate on the α was illustrated in Fig. 5. An increase in feed temperature decreased the separation factor. As the liquid flow-rate was increased, α increased also.

Experimental models were created to establish the relationships between the operating parameters (such as feed temperature and liquid flow rate), and the permeate flow rate and the separation factor. Regression equations in case of Celfa CMG OM-10 membranes are as follows:

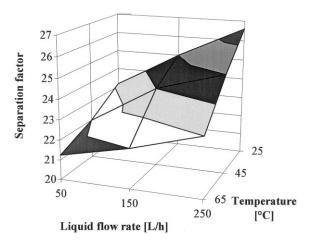


Fig. 5. Influence of temperature and permeate-side pressure on separation factor of Celfa CMG OM-10 membrane.

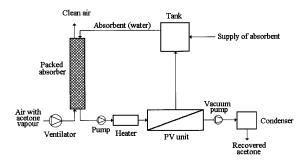


Fig. 6. Schematic diagram of the absorption-pervaporation system.

Regression equation for total flux of permeate with unstandardized coefficients:

$$J_{\rm P} = -0.344 + 2 \times 10^{-2} T_{\rm F} - 1.5 \times 10^{-4} L_{\rm R}$$
 (6)

and with standardized coefficients:

$$J_{\rm P} = 0.541 + 0.415T_{\rm F (stand)} - 0.048L_{\rm R (stand)}$$
 (7)

Regression equation for separation factor with unstandardized coefficients:

$$\alpha = 20.80 - 6.88 \times 10^{-2} T_F + 4.37 \times 10^{-2} L_R \qquad (8)$$
 and with standardized coefficients:

$$\alpha = 24.11 - 0.556T_{\text{F (stand)}} + 2.985L_{\text{R (stand)}}$$
 (9)

The validity of the Eqs. (6)–(9) is in the ranges of $T_F = 25-65$ °C and $L_R = 50-250$ l/(m² h).

On the basis of our experiments the optimal operating parameters that represented the highest separation with acceptable flux are as follows: $T_{\rm F}=65^{\circ}{\rm C}$ and $L_{\rm R}=250$ l/h.

3.3. Cost analysis

A cost analysis using recent economic data [8,9] were performed. Investment and operational cost of an absorption-pervaporation system were studied (Fig. 6) to treat 1000 m³/h feed (air with 2% of weight of acetone). In the absorption column the air containing acetone is counter-currently contacted with the absorbent. The outlet water with dissolved acetone is led to the pervaporation unit after preheating (if necessary). The vaccum pump insures the driving force on the permeate side of the membrane, and the passed vapours are condensed after the pump. The retentate can be reused for acetone-absorption.

The needed diameter of absorption tower packed with Sulzer Mellapak 250.Y packing is 0.8 m. The concentration of outlet gas is 0.02% of weight of acetone, and of the outlet liquid is 4% of weight.

A total of 70 m² membrane area of Celfa CMG OM-10 membrane is needed in case of 10 mbar permeate-side pressure.

The optimal parameters of the absorption-pervaporation system were determined by multicriterial optimization method. That means the operation parameters in case of minimal costs. The optimum of the whole system often differes from the parameters that represent the highest separation [10]. The optimal parameters of the system: 0.4 m/s superficial gas velocity and 950 l/(m² h) (600 l/h) of absorbent flow, 40°C pervaporation temperature and 250 l/h (500 l/(m²h)) recycled liquid flow. The weight factors were 0.2, 0.2, 0.4 and 0.2 consequently.

The main investment and operating expenses in case of optimal operating parameters are expressed in percents of the total cost and illustrated in Table 2.

As Table 2 represents the most significant expense is the investment cost of membrane, the sum of operation costs is less than 20% of the total cost.

The calculated total cost was 77 200 US dollar/year and the cost of recovered and reusable ace-

Table 2 Main investment and operating costs at optimal operating parameters (Celfa CMG OM-10 pervaporation membrane, area: 70 m^2 , P=10 mbar)

Costs	US dollars/year	% of Total Cost
Investment costs Absorption tower Pervaporation unit	4500 59 100	5.8 76.6
Sum of investment costs	63 600	82.4
Sum of operation costs	13 600	17.6
Total cost	77 200	100

tone was 113 000 US dollar/year, therefore the process could be economic.

4. Conclusions

The absorption with water is a suitable process for the removal of acetone vapours, a satisfactory removal efficiency was achieved.

Pervaporation is a technically feasible and economically attractive method for removing acetone from water, high fluxes and separation factors were obtained for the removal of acetone from water using pervaporation membrane.

The proposed absorption-pervaporation process is compact, closed and may be made continuous.

Valuable and harmful organic solvents can be economically recovered, purified, and reused by this type of clean technology.

Acknowledgements

The authors express their gratitude to the Sulzer and Celfa companies for the packing and membranes.

References

- [1] R. Billet, Packed Towers, VCH, Weinheim, 1995.
- [2] M. Mulder, Basic Principles of Membrane Technology, Kluwer Academic Publishers, Dordrecht, 1996.
- [3] H. Eustache, G. Histi, Separation of aqueous organic mixtures by pervaporation and analysis by mass spectrometry or a coupled gas chromatograph-mass spectrometer, J. Membr. Sci. 8 (1981) 105–114.
- [4] J.G. Wijmans, J. Kaschemekat, J.E. Davidson, R.W. Baker, Treatment of organic-contaminated wastewater streams by pervaporation, Environ. Progr. 9 (4) (1990) 262–268.
- [5] E. Márki, A. Mali, E. Békássy-Molnár, Gy. Vatai, Separation of chlorinated organic solvents from water by pervaporation and membrane distillation, 13th International Congress of Chemical and Process Engineering, CHISA'98, vol. 4, Prague, 23–28 August 1998, p. 166.
- [6] C. Lipski, P. Côté, The use of pervaporation for the removal of organic contaminants from water, Environ. Prog. 9 (4) (1990) 254–261.
- [7] H.L. Schulman, C.F. Ullrich, N. Wells, Performance of packed columns I, II, III, AIChE J. 1 (2) (1955) 247– 264.
- [8] Austrolab, Private communication, 1999.
- [9] Celfa, Private communication, 1999.
- [10] E. Békássy-Molnár, E. Márki, I. Bágyi, Gy. Vatai, Absorption-desorption procedure for regenerable sulphur dioxide recovery. Distillation and Absorption '97, Proceedings, vol. 1, Maastricht, The Netherlands, 8–10 September, 1997, pp. 467–476.