Brazilian Journal
of Chemical
Engineering

ISSN 0104-6632
Printed in Brazil
www.abeq.org.br/bjche

Vol. 26, No. 01, pp. 219 - 226, January - March, 2009

EXPERIMENTAL STUDY OF A ROTATING PACKED BED DISTILLATION COLUMN

J. V. S. Nascimento, T. M. K. Ravagnani* and J. A. F. R. Pereira

School of Chemical Engineering, University of Campinas,
PO Box. 6066, 13083-970, Campinas - SP, Brazil.
E-mail: kakuta@feq.unicamp.br

(Submitted : April 1, 2007 ; Revised: March 23, 2008 ; Accepted: September 12, 2008)

Abstract - The purpose of this work was to study the mass transfer performance of rotating packed beds applying the "Higee" process. The operations were carried out with the n-hexane/n-heptane distilling system at atmospheric pressure and under total reflux conditions. The rotating speed could be varied between 300 and 2500 rpm, which provided centrifugal forces from 5 to 316 times the Earth's gravity. The effects of concentration, vapor velocity, rotating speed and packing type (two different Raschig ring sizes and structured wire mesh packing) on mass transfer behavior were analyzed. Experimental results showed that the mass transfer coefficient depends on the liquid flow rates and rotating speed. The equipment had high separation efficiency in a reduced bed volume.

Keywords: Distillation; Mass transfer coefficient; Rotating packed bed.

INTRODUCTION

Packed columns are important for large-scale vapor-liquid operations, such as distillation, absorption and stripping. The capacity of these columns for countercurrent mass transfer is limited by flooding. As a consequence, columns with a large diameter are usually required to obtain high separation degrees. To overcome this limitation the "Higee" technology (" high g" – high gravity) utilizes a rotating packed bed (RPB), first developed by Ramshaw and Mallinson in 1981 (Ramshaw, 1983), which can induce centrifugal forces over 1000 times that of the Earth's gravity. The increased driving force allows for increased throughput in the unit and improved mass transfer efficiency.

In addition, packings with larger specific area and higher void fraction can be used. Under high centrifugal acceleration, thinner films and smaller droplets may be formed. According to several authors (Ramshaw, 1983; Kelleher and Fair, 1996; Lin et al., 2002), this column type is highly efficient and allows to use smaller, achieving the goal of a reduction in volume of two to three orders of magnitude over conventional packed beds. A critical view of the developments in understanding the transport processes in rotating packed beds and the directions taken to better achieve the goal are outlined by Rao et al. (2004).

According to Kelleher and Fair (1996), a lack of understanding of the contact process plus a relatively high cost have limited its industrial use. However in special situations, the smaller size and lower weight of the RPB compared with the traditional contact columns made the Higee more competitive than larger and bulkier columns.

The centrifugal column offers many other advantages, such as lower liquid hold-up; higher vapor and liquid flows; quicker achievement of steady state; the ability to process dangerous

---

*To whom correspondence should be addressed operations; and the opportunity to process lower inventories of corrosive, toxic or inflammable materials, reducing the risks of fire or explosion. Also, heat-sensitive materials can be processed effectively, so RPBs have been applied to processes such as ozone oxidation and synthesis of drug and inorganic nanoparticles (Chandra et al., 2005; Chen et al., 2005a,b; Chen et al., 2006; Yang et al., 2006).

The gas-side and liquid-side mass transfer and hydrodynamic behavior in high-gravity contactors have been studied. However, the Higee distillation process is still poorly understood. Kelleher and Fair (1996) first reported hydrodynamic data and mass transfer for distillation in rotating packed beds using the cyclohexane/n-heptane system at operating pressures of 166 and 414 kPa and under conditions of total reflux as well as design correlations. Lin et al. (2002) presented rotating packed bed distillation experiments using the methanol/ethanol system at atmospheric pressure and total reflux conditions with stainless wire mesh packing. An empirical correlation for pressure drop and HETP (height equivalent of a theoretical plate) is proposed in this paper.

The present work was undertaken to study the behavior of mass transfer in a rotating packed column distillation for different kinds of packing. Experimental data were obtained to determine the volumetric overall vapor-side mass transfer coefficients, $K_y a_e$, for the n-hexane/n-heptane binary system and to analyze its dependence on rotating speed and mass flow.

EXPERIMENTAL SETUP AND PROCEDURE

Figure 1 shows a diagram of the experimental distillation unit and the main structure of the rotating packed bed (rotor) used in this study. An external cylindrical case, made of glass, houses the centrifugal equipment. The rotor is shaped like a big doughnut, within which the packing is arranged. The rotating torus of packing has an outer radius of 8.0 cm, an inner radius of 2.2 cm and an axial length of 4.0 cm. Three different packings (r01 and r02 random Raschig ring packing and e01 structured wire mesh packing) were used in this rotor for each experiment. The wire mesh packing was composed of 35 layers of wire mesh sheet with a standard 1.0 mm sieve opening. The characteristics of the packings are shown in Table 1. These packings were chosen mainly due to the fluid flow path inside the column; while in the conventional packing column an axial fluid flow is found, in the RPB column the main fluid flow path is a radial flow. In addition, the feasibility of adapting them to a small size doughnut-shaped rotor and availability were very important factors that affected the choice.

The vapor was introduced into the housing and flowed radially in countercurrent to the liquid from the outer edge to the central gap of the rotor called the “rotor eye.” A 3 cm tall weir was provided to avoid liquid drainage at the vapor inlet.

The liquid from the condenser was sprayed by a distributor system from the rotor eye to as far as the outer edge because of the centrifugal force. This distributor at the “rotor eye” was formed of a group of fins (4.0 cm long and 1.7 cm wide) arranged helicoidally. Figure 1 shows a detail of the distributor.

A motor was connected to the bed through a pulley system to produce the bed rotation and its rotating speed was varied within the range from 300 to 2500 rpm, which resulted in a centrifugal force equivalent to 5 g to 316 g.

In this work, all experiments were carried out under total reflux conditions at atmospheric pressure. The liquid loads ranged from about 5.7 cm^3/s to 29.0 cm^3/s for the r01 and r02 packings and from about 2.0 cm^3/s to 18.5 cm^3/s for the e01 packing. For these, the power of six electrical resistances ranged between 1500 W and 7500 W. For total immersion of these electrical resistances, 5 liters of liquid holdup were provided in the reboiler.

The composition of n-hexane in the reboiler ($x_{A,F}$) was maintained at 3 mole% during the conduction of the experiments. Liquid samples at rotor outlet ($x_{A,0}$) and liquid reflux of the column ($x_{A,1}$) were taken and analyzed after the steady state was obtained. The average time to attain the steady state was about 20 minutes after establishing liquid reflux. All compositions were analyzed through refractive index measurement at 20°C. The physical properties of the pure components were calculated using experimental data provided by Vargaftik (1975) and Reid et al. (1987).

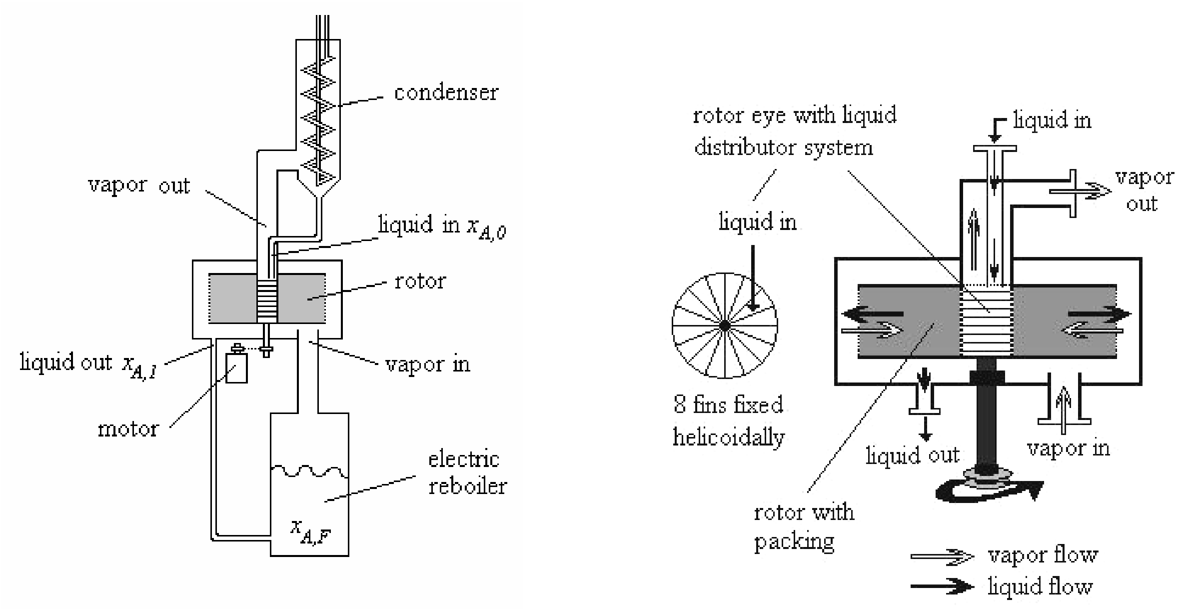


Figure 1: Sketch of the distillation column and the rotating packed bed

Table 1: Rotor and packing characteristics

Packing	r01	r02	e01
size 10^2 (m)	0.90	0.70	-
thickness 10^2 (m)	0.20	0.15	-
a (m^2/m^3)	627	765	2100
void fraction	0.62	0.55	0.74
type	ring	ring	wire mesh
material	ceramic	ceramic	stainless steel

RESULTS AND DISCUSSION

Volumetric Overall Mass Transfer Coefficient

The $K_y a_e$ obtained for the centrifugal bed could be determined using mass balances on a differential volume of the Higee rotor and transfer unit concepts, as shown in Figure 2. The equilibrium vapor composition and temperature at the average local atmospheric operating pressure of 709 mmHg were predicted using the Wilson equation. The binary interaction coefficients were obtained from Gmehling and Onken (1977).

Liu et al. (1996) presented an equation for the volumetric overall vapor-side mass transfer coefficient for the process depicted in Figure 2, assuming constant liquid and vapor flow rates and equimolar counterdiffusion mass transfer, and it can be applied to systems where the equilibrium curve is predicted by Henry's law ($y^*= m x$). Since the liquid composition of n-hexane in the reboiler ($x_{A,F}$) was kept at 3 mole% and the rotor outlet ($x_{A,1}$) and inlet ($x_{A0)}$) liquid composition depended on efficiency (i.e., on the operating conditions), Henry's law not always could be applied. Thus, a similar equation was deduced for a more general equilibrium curve ($y^*= m x+b$):

$$K_y a_e = \frac{V_{mol}}{h_a \pi (r_e^2 - r_i^2)} \frac{1}{S-1} \ln\left[(1-S)\frac{y_{A,1} - m\ x_{A,1} - b}{y_{A,2} - m\ x_{A,1} - b} + S\right] \quad (1)$$

where S is a distillation factor defined by

$$S = \frac{mV_{mol}}{L_{mol}} \quad (2)$$

The values of m and b were determined for each run fitting the best straight line between $x_{A,0}$ and $x_{A,1}$ at the VLE curve.

Figures 3 and 4 show the effect of rotating speed on the volumetric overall vapor-side mass transfer coefficient for various liquid flow rates at the same concentration for the Raschig ring and structured packings, respectively. According to the results shown in these figures, the effect of rotating speed on $K_y a_e$ at high liquid flow rates is more considerable than at low liquid flow rates for both packings.

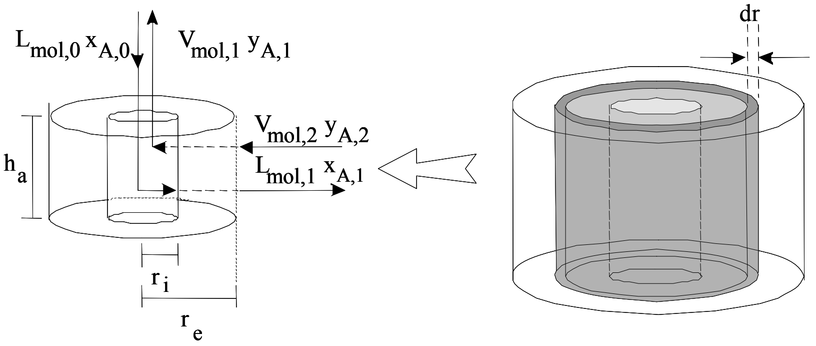
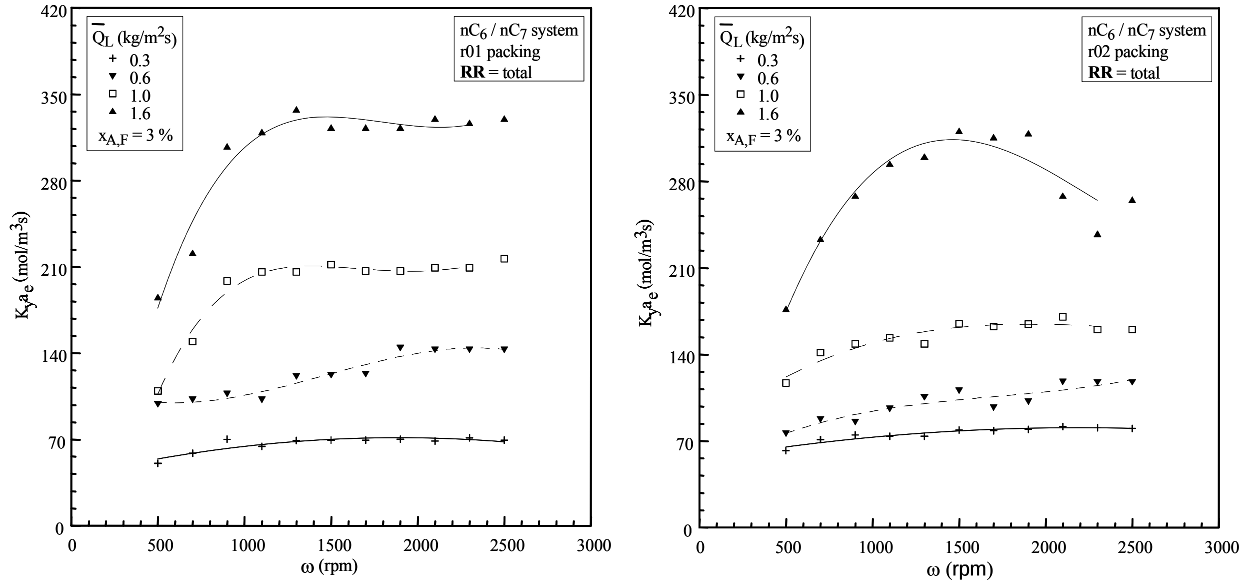


Figure 2: Differential volume of the Higee rotor



(a) r01 packing (b) r02 packing

Figure 3: Effect of rotating speed on K_ya_e for different size of Raschig rings

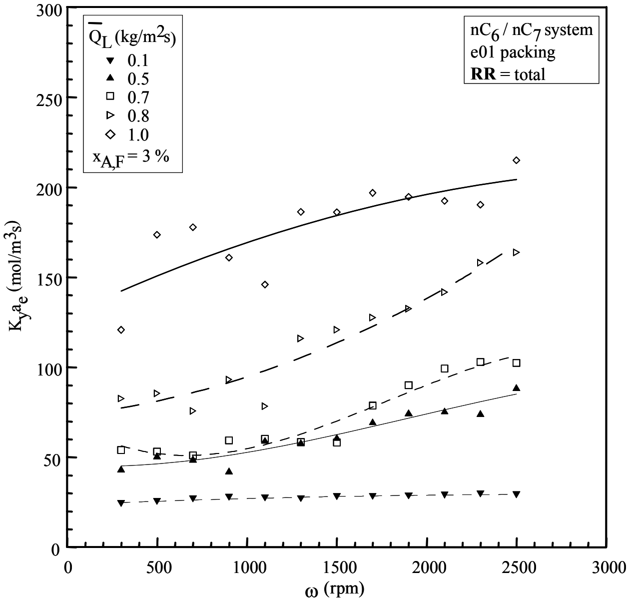


Figure 4: Effect of rotating speed on K_ya_e for the e01 packing

As shown in Figure 3a, for higher liquid flow rates such as 1.0 kg/m^{2}s and 1.6 kg/m^{2}s, there were considerable increases in K_ya_e at rotating speeds up to 1100 rpm, showing the effect of the centrifugal force. Above this value, K_ya_e was practically constant, probably due to the small residence time of the liquid and vapor currents. This behavior was similar to those shown in Figure 3b, at the high liquid flow rate that provided a maximum K_ya_e at around 1500 rpm. However, Figures 3a and 3b show that at low liquid flow rates, such as 0.3 kg/m^{2}s and 0.6 kg/m^{2}s, K_ya_e increased with liquid flow rate and slightly increased with increasing rotating speed, showing a similar trend. This suggests that at lower liquid flow rates centrifugal force had practically no effect on K_ya_e. Comparison of Figures 3a and 3b, shows that the r01 packing (higher void fraction) provided higher K_ya_e values than the r02 packing (higher specific area). This result led us to conduct a new set of experiments using the e01 packing, which provided the highest void fraction.

Figure 4 shows the effect of rotating speed on K_ya_e for the e01 packing under the same conditions as those presented for the r01 and r02 packings. The same trend can be seen for the e01 packing, i.e., the effect of rotating speed on K_ya_e at high liquid flow rates was greater than at low liquid flow rates. However the considerable increase in K_ya_e at rotating speeds up to 1100 rpm did not occur. Also, the results show that, at low liquid flow rates (such as 0.6 kg/m^{2}s) the e01 packing had the lowest K_ya_e value, but at high liquid flow rates (such as 1.0 kg/m^{2}s) this packing had the highest K_ya_e value, allowing us to conclude that the effect of liquid flow rate was stronger for the e01 than for the Raschig ring packing. It is very important to observe that the operation with liquid flow rate near 1.6 kg/m^{2}s was not possible due to excessive liquid holdup inside the condenser, probably caused by the increasing pressure drop.

Area of Transfer Unit – ATU

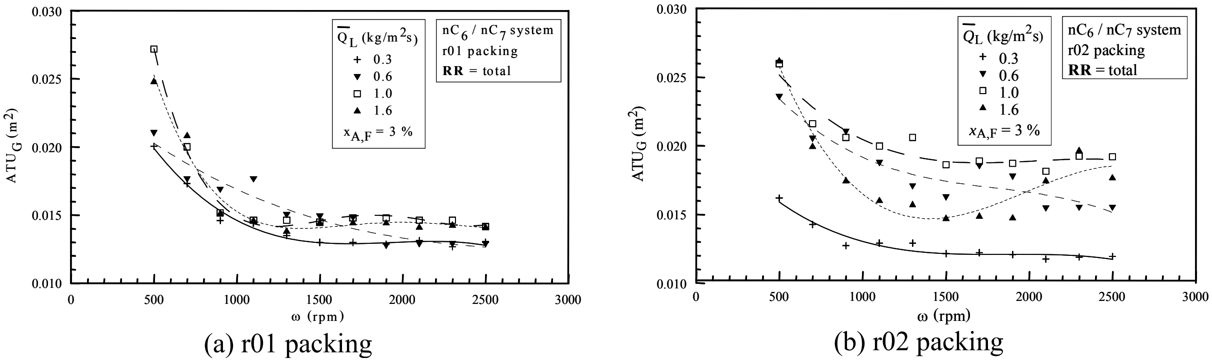
According to Equation 1, the factor V_{mol} / (h_a K_ya_e) is defined as ATU_G (area of transfer unit). The ATU concept is in a form similar to that of HTU_G (height of a transfer unit), used in the conventional column, and it denotes a measure of column efficiency. According to the equipment configuration, the Higee is a polar rather than a Cartesian coordinate system, i.e., the ATU_G definition is a convenient way to accomplish radial change.

HETP (height equivalent of a theoretical plate) is the other way to express equipment efficiency. Lin et al. (2002) presented HETP data. The rotating size utilized in our work was small, resulting in a very low number of theoretical plates and so HETP data can not be distinguished properly.

Changes in K_ya_e with rotating speed are directly reflected in the value of separation efficiency. The effect of rotating speed on ATU_G for various liquid flow rates, using the r01 and the r02 packings at a fixed concentration, is shown in Figure 5. ATU_G decreased with increasing rotating speed, except at the highest liquid mass flow rate for the r02 packing, which provided minimum ATU_G at around 1500 rpm, as shown in Figure 5b. Figure 5a shows that in the range of 300 to 1300 rpm ATU_G was highly dependent on rotating speed for all liquid flow rates. This suggests that the centrifugal column achieved greater efficiency at higher rotating speeds and at lower liquid flow rates.

Those figures also show that the efficiency of the r01 packing was only slightly affected by liquid flow rate, while for the r02 packing, the effect of $\overline{Q}_L$ on ATU_G was stronger. Efficiency decreased with increasing $\overline{Q}_L$, reaching a minimum value (maximum ATU_G) at 1.1 kg/m^{2}s. After that, it began to increase. This behavior occurred because this condition was found near the operational limit of the column (2.0 kg/m^{2}s).

In Figure 6 the ATU_G results for the e01 packing are presented. As expected, the centrifugal column achieved the highest efficiency at higher rotating speeds and at lower liquid flow rates. The effect of $\overline{Q}_L$ on ATU_G was very strong, showing the same behavior as seen for the r02 packing. The lowest efficiency was achieved at 0.7 kg/m^{2}s.



(a) r01 packing

(b) r02 packing

Figure 5: Effect of rotating speed on ATU_G

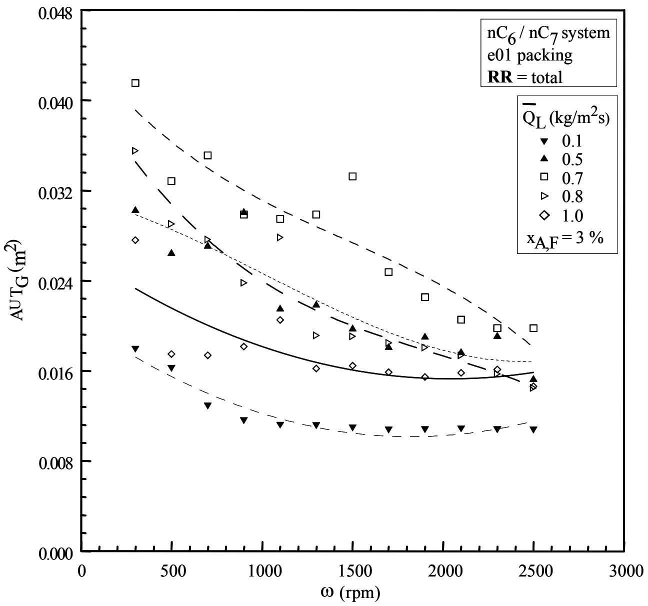


Figure 6: Effect of rotating speed on ATU_G

Comparative Analysis of Conventional and RPB Columns

The conventional and centrifugal columns were compared for the Raschig packing.

Figure 7 contains the results on the centrifugal K_ya_e obtained experimentally with Equation 1 for RPB and K_ya_e calculated using the correlation proposed by Onda et al. (1968) for the conventional packing column under the same liquid mass flow that those obtained for the RPB.

The centrifugal K_ya_e was higher than the conventional K_ya_e (up to 15.1 times), as shown in Figure 7. Furthermore, the centrifugal K_ya_e was highly dependent for all liquid flow rates at the same rotating speed.

The volume of the conventional equipment was also calculated. For the same degree of separation (equal NTU_G), a conventional bed whose volume of r01 Raschig ring packing was 8 to 15 times greater was required, as shown in Table 2.

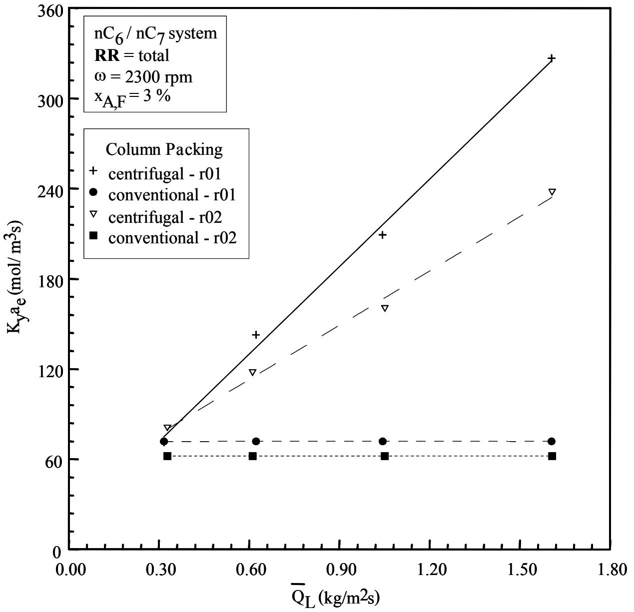


Figure 7: Comparison of the centrifugal and conventional K_ya_e

Table 2: Comparison of the volume of conventional and centrifugal beds

$\overline{Q_L}$ = 1.6 kg/m^2 s, $\overline{x}_{A,F}$ = 0.03, Raschig ring – r01			
Centrifugal column: ν_{cent} = 7.4 $1\tilde{0}^{4}$ m^3, $h_{a,cent}$ = 0.04 m			
ω (rpm)	**Conventional column**		**ν_{conv} /ν_{centr}**
	$h_{a,conv}$ (m)	**ν_{conv} .10^3 (m^3)**	
500	0.0634	1.896	2.6
900	0.1046	3.150	4.3
1300	0.1145	3.455	4.7
1900	0.1097	3.305	4.5
2500	0.1109	3.343	4.5

CONCLUSIONS

This study investigated the behavior of a centrifugal packed bed distillation column using the n-heptane/n-hexane system. The volumetric overall vapor-side mass transfer coefficient was determined.

The results of the experimental analyses indicate that the effect of rotating speed on $K_y a_e$ at high liquid flow rates was greater than at low liquid flow rates for all packing types. At lower liquid flow rates, $K_y a_e$ slightly increased with increasing rotating speed. The Raschig packing with a higher void fraction (r01) provided higher $K_y a_e$ values than the packing with a higher specific area (r02).

Lower liquid flow rates were obtained for the structured packing. The same trend of the effect of rotating speed on $K_y a_e$ as that observed for the r01 and r02 packings was observed for the e01 packing. However the effect of liquid flow rate was stronger for the e01 than the random packing.

The separation efficiency of a rotating packed bed represented by ATU_G increased with rotating speed. The same trend in rotating speed was seen for all liquid flow rates with rotating speeds up to 1300 rpm, except at the highest liquid mass flow rate for the r02 and e01 packings. Therefore, the centrifugal column was more efficient at higher rotating speeds and at lower liquid flow rates.

For the r01 packing the efficiency was almost not affected by liquid flow rates while for the r02 packing, this effect was stronger. For the e01 packing, this effect was even stronger.

The volumetric overall mass transfer coefficients for the centrifugal bed were higher than those for the conventional column. The column volume of the centrifugal column is almost 5 times smaller than that of the conventional. Centrifugal force was an important parameter, giving better mass transfer results than the conventional packed bed.

NOMENCLATURE

ATU_G	area of transfer unit for gas phase	
b	linear coefficient of equilibrium curve	
g	gravitational acceleration	
h_a	axial height of rotor, m	
$K_y a_e$	overall volumetric overall vapor-side mass transfer coefficient	
$L_{mol,0}$	molar flow rate of liquid at rotor inlet	
$L_{mol,1}$	molar flow rate of liquid at rotor outlet	
m	slope of equilibrium curve	
$\overline{Q}_L$	average liquid mass flow rate	
r_e	rotor outer radius	
r_i	rotor inner radius	
S	distillation factor	(mV_{mol} / L_{mol})
$V_{mol,1}$	molar flow rate of vapor at rotor outlet	
$V_{mol,2}$	molar flow rate of vapor at rotor inlet	
v	volume	
$x_{A,0}$	mole fraction of liquid at reboiler	
$x_{A,1}$	mole fraction of liquid at rotor outlet	
x_{AF}	mole fraction of liquid in the reboiler	
$y_{A,1}$	mole fraction of vapor at rotor inlet	
$y_{A,2}$	mole fraction of vapor at rotor outlet	

Greek Letters

ω	Rotating speed

Subscripts

cent	centrifugal
conv	conventional

REFERENCES

Chandra, A., Goswami, P. S., Rao, D. P., Characteristics of Flow in a Rotating Packed Bed (HIGEE) with Split Packing, Ind. Eng. Chem. Res., 44, 4051-4060 (2005).

Chen, Y. S., Lin, F. Y., Lin, C. C., Tai, C. Y. D., Liu, H. S., Packing Characteristics for Mass Transfer in a Rotating Packed Bed, Ind. Eng. Chem. Res., 45, 6846-6853 (2006).

Chen, Y. S., Lin, C. C., Liu, H.S., Mass Transfer in a Rotating Packed Bed with Various Radii of the Bed, Ind. Eng. Chem. Res., 44, 7868-7875 (2005a).

Chen Y. S, Lin, C. C., Liu, H. S., Mass Transfer in a Rotating Packed Bed with Viscous Newtonian and Non-Newtonian Fluids., Ind. Eng. Chem. Res., 44, 1043-1051 (2005b).

Gmehling, J., Onken, U., Vapour-Liquid Equilibrium Data Collection, Dechema Chemistry Data Series (1977).

Kelleher, T; Fair, J. R., Distillation Studies in a High-Gravity Contactor, Ind. Eng. Chem. Res., 35, 4646-4655 (1996).

Liu, H. S., Lin, C. C., Wu, S. C., Shu, H. W., Characteristics of a Rotating Packed Bed, Ind. Eng. Chem. Res., 35, 3590-3596 (1996).

Lin, C. C., Ho, T. J., Liu, W. T., Liu, H. S., Distillation in a Rotating Packed Bed, J. Chem. Eng. Jpn., 35, 1298-1304 (2002).

Onda, K., Takeuchi, H., Okumoto, Y., Mass Transfer Coefficients between Gas and Liquid Phases in Packed Columns, J. Chem. Eng. Jpn., 1, 56 (1968).

Ramshaw, C., “HIGEE” Distillation – An Example of Process Intensification, Chem. Eng., 13-14 (1983).

Rao, D. P., Bhowal, A., Goswami, P. S., Process Intensification in Rotating Packed Beds (HIGEE): An Appraisal, Ind. Eng. Chem. Res., 43, 1150-1162 (2004).

Reid, R. C., Prasnitz, J. M., Poling, B. E., The Properties of Gases and Liquids, New York: McGraw-Hill, Inc., 741 p. (1987).

Vargaftik, N. B., Tables on the Thermophysical Properties of Liquids and Gases, 2nd ed., John Wiley & Sons Inc. (1975).

Yang, H. J., Chu, G. W., Xiang, Y., Chen, J. F., Characterization of micromixing efficiency in rotating packed beds by chemical methods, Chem. Eng. J., 121, 147-152 (2006).