RESEARCH ARTICLE

Study of packed sieve tray column in ethanol purification using distillation process

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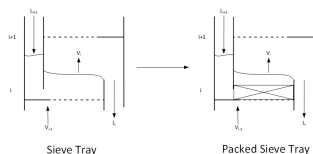
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Graphical abstract

Sieve Tray   Packed Sieve Tray

Abstract

Sieve tray becomes a popular contacting device in distillation process because of its relative simplicity and low cost. There is one way to improve the contact performance, especially mass transfer by modifying the sieve tray into packed sieve tray. This study was aimed to investigate the effect of adding packing in each tray to ethanol content on ethanol purification. This research was conducted via experiment and simulation approaches. The experiment used packed sieve tray that contained 3 cm and 5 cm bed of steel wool with 16 trays in the column, with operating pressure about 760 mmHg and performed in batch condition. The simulation used a reduced rated base model with some modifications for operation in the packed sieve tray column. It was found that the use of packed sieve tray gave better distillate in the batch distillation process than the use of sieve tray. The packed sieve tray raised distillate content about 8.89% when using 3 cm of packing and 23.31% when using 5 cm of packing when it was compared with sieve tray. The use of packed sieve tray could increase the mass transfer and reduce bubble diameter in the batch distillation process.

Keywords: Distillation, Ethanol, Packed sieve tray, Packing

INTRODUCTION

Distillation is a process of separation and purification that is widely used in the chemical industry. It is used to separate the components in solution which have different boiling points for partially miscible or immiscible solutions into their respective components. When a multicomponent solution is heated, the vapor will be dominated by volatile components. Today, distillation devices that widely used in the chemical industry were tray towers and packed towers.

Trays are used to enlarge the contacts between liquids and gases so that components can be separated in the form of gases or liquids. Trays are widely used in distillation columns because they are easy to design and require only a relatively small cost. There are a few types of trays such as sieve tray, valve tray, and bubble cap. Tray has a low efficiency in the process of mass transfer between vapor-liquid so it needs modification to improve efficiency.

Packing is the contact device between the vapor-liquid in the distillation column, the absorption column, and the stripping column. Packing provides low pressure drop, high mass transfer efficiency, and high capacity. Types of packing are structured packing and random packing. Recent research is directed by engineer in a packing structure that meets small pressure drop criteria but has high mass transfer efficiency.

Spagnolo and Chuang (1984) investigated sieve tray performance in combination with mesh packing. It was found that the tray efficiency increased from 5 to 40% with the addition of 30 mm mesh packing on one tray and packed sieve tray had low entrainment and high pressure drop. Chen *et al.* (1992) researched on hydraulic work

and mass transfer of a sieve tray with mesh packing for various heights in methanol-water system. Their study found that Murphree efficiency of the tray increased 40-50% for various concentrations and flow rates. Xu et al. (1996) found that the efficiency of the sieve tray was increased significantly with the addition of mesh packing. Packing would cause increased interfacial area and vapor-liquid contact time. Kachur et al. have been researched the use of structured packing as a substitute for dual flow tray. From the research, it was reported that structured packing tray has a larger capacity of 44% and has 100% greater efficiency when compared with dual low tray.

Khrishnamurthy and Taylor (1985) created a mathematical model for nonequilibrium stage used in countercurrent simulations of multicomponent separation processes. In this model, the relationship between the mass balance and the heat balance of each component for each phase, the mass transfer rate and energy equations and the equilibrium equations at the phase interface were solved in order to obtain the actual separation conditions. Bonilla *et al.* (2012) modified the mathematical model for nonequilibrium stage that previously developed by Khrishnamurthy and Taylor. In this study, reduced order rate based model has been successfully developed. To accommodate the operating conditions used in this study, modifications were made to the stage at the rate based model that was initially assumed to be packed sieve tray column.

Based on previous works, it can be concluded that the addition of packing on one tray can increase mass transfer and tray efficiency. This study investigated the effect of adding packing on each tray in the distillation process in order to improve purity. Packed sieve tray can increase the purity of ethanol due to the increase of mass transfer and the occurrence of bubble diameter reduction process in the packed

sieve tray. A model and simulation of the distillation column using a packed sieve tray were required to accommodate the phenomena occurring within the tray which would be validated by the experimental results.

EXPERIMENTAL

Materials

The chemicals used were ethanol 96% v/v and aquadest. Packing of steel wool type was also used in this research with wide area about 20.5 cm^2, as indicated in Figure 1 below.
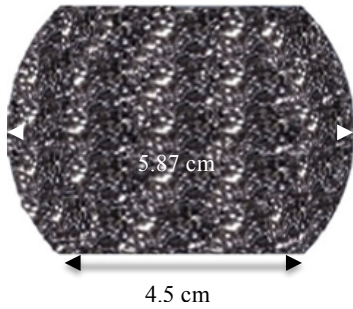

Figure 1 Dimension of steel wool packing.

Distillation process

The distillation process used a modified sieve tray columns with the addition of packing with the number of trays about 16. This research used packing of steel wool type. Figure 2 shows the schematic of the distillation column that used in this research. This particular modified sieve tray column is known as packed sieve tray distillation column. Packing of steel wool type was used in this study with high variation, in 3 and 5 cm. The procedure was started by preparing a feed solution consisted of 6 mol% of ethanol and 94 mole% water and pouring 13 L of feed into the reboiler under atmospheric conditions with a reflux ratio that maintained at 3.5. Cooling water was flowed to the condenser. The boiler was turned on and the temperature of boiler was kept at 100 °C. The distillation process was carried out for approximately 1 hour and the purity of ethanol was observed. The specifications of the distillation column were shown in Table 1.

Table 1. Specifications of the distillation column.

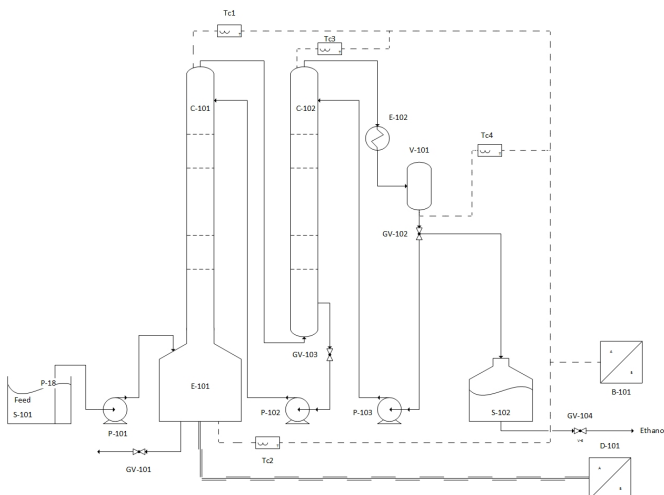| Information	Size
Column Diameter	6.2 cm
Tray Spacing	30 cm
Hole Diameter	2 mm
Column Height	2.5 m
Number of Tray	16
Tray Type	Sieve Tray
Weir Height	9 cm

Figure 2 The schematic of the distillation column

Analysis

The distillation product was analyzed using gas chromatography to determine the ethanol content obtained. The type of gas chromatography used in this research was Gas Cromatografi Thermo DSQ II Mass spectrometer.

Modelling and simulation of distillation process

The model developed was based on a reduced rated base model with some modifications for operation with a certain reflux ratio. Runge Kutta numerical solutions were used to solve the differential equations in this study. Some assumptions were used in the distillation simulation to facilitate the completion of the rate based model, as following:

- Using a binary mix.
- Bulk phases were assumed to be perfectly mixed.
- Vapor-liquid equilibrium was occurred only in the vapor-liquid interface.
- The relationship between heat flux and mass fluxes in the interface was ignored.
- Reboiler was assumed to be in a thermodynamical equilibrium state.

The mathematical equations mentioned in following paragraphs were used in dynamic and algebraic equations. Notation g in mathematic equation was reflected to algebraic equation.

Reboiler

The partial reboiler was assumed in a thermodynamical equilibrium state. The mass and energy balance was modeled in equilibrium stage.

$$\dot{M}_{1,1}^L = L_2 x_{1,2}^l - B x_{1,1}^l - V_1 y_{1,1}^l \tag{1}$$

$$\dot{M}_{2,1}^L = L_2 x_{2,2}^l - B x_{2,1}^l - V_1 y_{2,1}^l \tag{2}$$

$$\dot{E}_1^L = L_2 H_2^L - B H_1^L - V_1 H_1^V + Q_R - Q_1^L \tag{3}$$

$$g_{K_{1,1}} = y_{1,1}^l - K_{1,1} x_{1,1}^l \tag{4}$$

$$g_{K_{2,1}} = y_{2,1}^l - K_{2,1} x_{2,1}^l \tag{5}$$

$$g_{P_1} = P_2 + \Delta P_2 - P_1 \tag{6}$$

$$g_{M_{t_1}^L} = M_{1,1}^L + M_{2,1}^L - M_{t_1}^L \tag{7}$$

Non-equilibrium Stage

The mass and energy balance at the bulk phase for component i=1,2 and stage j=2,…,N-1

$$\dot{M}_{1,j}^L = L_{j+1} x_{1,j+1} - L_j x_{1,j} + F_j^L x_{1,j}^F + N_{1,j} \tag{8}$$

$$\dot{M}_{2,j}^L = L_{j+1} x_{2,j+1} - L_j x_{2,j} + F_j^L x_{2,j}^F + N_{2,j} \tag{9}$$

$$\dot{M}_{1,j}^V = V_{j-1} y_{1,j-1} - V_j y_{1,j} + F_j^V y_{1,j}^F - N_{1,j} \tag{10}$$

$$\dot{M}_{2,j}^V = V_{j-1} y_{2,j-1} - V_j y_{2,j} + F_j^V y_{2,j}^F - N_{2,j} \tag{11}$$

$$\dot{E}_j^L = L_{j+1} H_{j+1}^L - L_j H_j^L + F_j^L H_{f,j}^L - Q_j^L + \varepsilon_j^L \tag{12}$$

$$\dot{E}_j^V = V_{j-1} H_{j-1}^V - V_j H_j^V + F_j^V H_{f,j}^V - Q_j^V + -\varepsilon_j^V \tag{13}$$

The total hold ups were calculated based on the geometry of distillation column and the component hold ups were described as

$$g_{M_{t_j}^L} = M_{1,j}^L + M_{2,1}^L - M_{t_j}^L \tag{14}$$

$$g_{M_{t_j}^V} = M_{1,j}^V + M_{2,1}^V - M_{t_j}^V \tag{15}$$

$$g_{M_{t_j}^L} = M_{t_j}^L - \frac{\pi}{4} d^2 l h_{t_j}^L c_{t_j}^L \tag{16}$$

$$g_{M_{t_j}^V} = M_{t_j}^V - \frac{\pi}{4} d^2 l (\epsilon - h_{t_j}^L) c_{t_j}^V \tag{17}$$

$$g_{E_j^L} = E_j^L - \frac{\pi}{4} d^2 l h_{t_j}^L c_{t_j}^L H_j^L \tag{18}$$

$$g_{E_j^V} = E_j^V - \frac{\pi}{4} d^2 l (\epsilon - h_{t_j}^L) c_{t_j}^V H_j^V \tag{19}$$

The interface equation of distillation column was described in this following equation

$$g_{N_{1,j}^L} = N_{1,j} - a^l c_t^L k_j^L \left(x_{1,j}^l - x_{1,j} \right) - x_{1,j}(N_{1,j} + N_{2,j}) \qquad (20)$$

$$g_{N_{1,j}^V} = N_{1,j} - a^l c_t^V k_j^V \left(y_{1,j} - y_{1,j}^l \right) - y_{1,j}(N_{1,j} + N_{2,j}) \qquad (21)$$

$$\varepsilon_j^V = h_j^V a_j^l \left(T_j^V - T_j^l \right) + \sum_{i=1}^{2} N_{1,j}^V \overline{H_{i,j}^V} \qquad (22)$$

$$\varepsilon_j^L = h_j^L a_j^l \left(T_j^l - T_j^L \right) + \sum_{i=1}^{2} N_{1,j}^L \overline{H_{i,j}^L} \qquad (23)$$

$$g_{\varepsilon_j} = \varepsilon_j^V - \varepsilon_j^L \qquad (24)$$

$$g_{K_{1,j}} = y_{1,j}^l - K_{1,j} x_{1,j}^l \qquad (25)$$

$$g_{K_{2,j}} = y_{2,j}^l - K_{2,j} x_{2,j}^l \qquad (26)$$

$$N^V = \frac{L_f \rho_V K_V}{V_f \rho_L K_L} N^L \qquad (27)$$

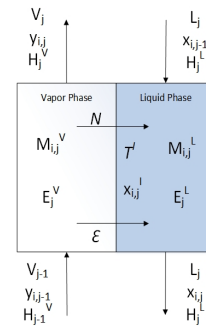$$N^L = k_L \frac{\rho_L V_f}{\rho_V L_f} a' t_G \qquad (28)$$

Equilibrium condenser

The total condensor was used to model the top stage in the column. Liquid was left the condenser with T_N^l temperature and amount of enthalpy H_C^L.

$$g_{E_{t,N}^L} = V_{N-1} H_N^V - L_C H_C^L + Q_C \qquad (29)$$

$$g_{K_{1,N}} = y_{1,N}^l - K_{1,N} x_{1,N}^l \qquad (30)$$

$$g_{K_{2,N}} = y_{2,N}^l - K_{2,N} x_{2,N}^l \qquad (31)$$

$$g_{y_{1,N}} = y_{1,N-1} - y_{1,N}^l \qquad (32)$$

Reflux Drum

The mass and energy balance for reflux drum was described in this following equation.

$$\dot{M}_{1,N}^L = L_C x_{1,N}^l - (L_r + D) x_{1,N} \qquad (33)$$

$$\dot{M}_{2,N}^L = L_C x_{2,N}^l - (L_r + D) x_{2,N} \qquad (34)$$

$$\dot{E}_N^L = L_C H_N^L - (L_r + D) H_N^L \qquad (35)$$

$$g_{M_{t_N}^L} = M_{1,N}^L + M_{2,N}^L - M_{t_{1N}}^L \qquad (36)$$

Figure 3 shows the system of the distillation column that used in this simulation. The packed sieve tray column simulated here was divided in four sections such as reboiler, packed sieve tray, condenser and reflux drum. Distillation column has 16 stages with added packing of steel wool type in each trays. The variation of heights used in this process was about 3 cm and 5 cm. Figure 4 illustrated the rate base model that occurred in the non-equilibrium stage. Mass and energy transfer took place in interface section because temperature and composition gradients, as respected by rate base model. The rate base model eliminated role of tray efficiency which was used in equilibrium model. The equation for each phase was related with mass and energy balance around interface section and interface was assumed at thermodynamic equilibrium.
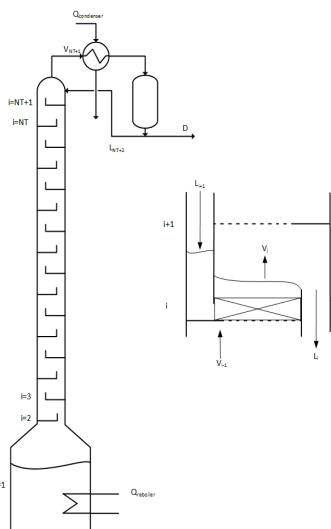

Figure 4 Non-equilibrium stage in the rate based model.

RESULTS AND DISCUSSION

This research used packed sieve tray consisted of 16 trays that arranged in series with atmospheric pressure. The experimental and simulation processes were carried out until the distillation process reached approximately 60 minutes. First, the distillation process was done with sieve tray which then continued with packed sieve tray which used packing of steel wool type with variation of packing heights of 3 cm and 5 cm.

Figure 5 shows the distillate content of the distillation time on a sieve tray. From Figure 5, it could be seen that the experimental results produced a maximum ethanol content of 76% mole and the simulation results produced a maximum ethanol content of 72% mole. These findings indicated the existence of deviations on the experimental and simulation results for about 6.08%.

Figure 6 shows the distillate content of the distillation time on a packed sieve tray using packing of steel wool type and 3 cm height. From Figure 6, it could be observed that the experimental results produced a maximum ethanol content of 83% mole while the simulation results produced a maximum ethanol content of 79% mole. These results proven the existence of deviations on the experimental and simulation results for about 5.02%.

Figure 7 shows the distillate content of the distillation time on a packed sieve tray using packing of steel wool type and 5 cm height. The experimental results produced a maximum ethanol content of 94% mole and the simulation results produce a maximum ethanol content of 94% mole, as presented in Figure 7. The obtained results referred to the existence of deviations on the experimental and simulation results for about 0.33%.
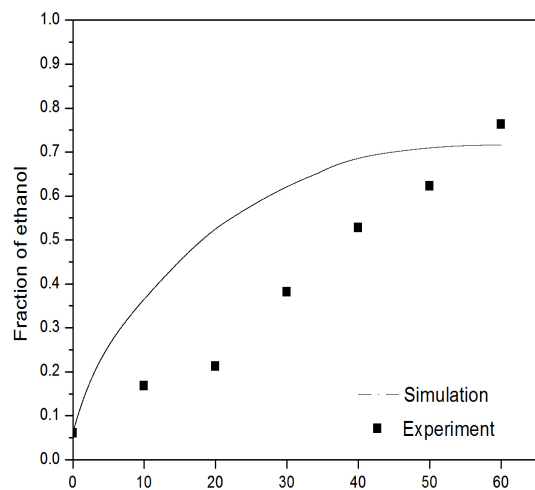

Figure 3 System of the simulation distillation process using packed sieve tray column.

Figure 5 The result of the distillate to the distillation time on the sieve tray consisting of 16 trays composed in series and atmospheric pressure for approximately 60 minutes of distillation operation.
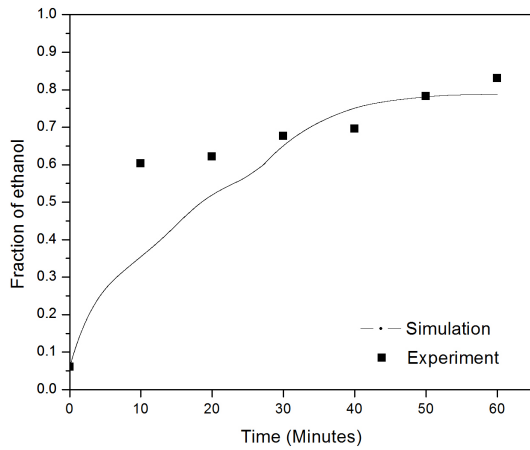
Figure 6 The result of the distillate to the distillation time on the packed sieve tray using packing of steel wool type and 3 cm height consisting of 16 trays composed in series and atmospheric pressure for approximately 60 minutes of distillation operation.

Figure 7 The result of the distillate to the distillation time on the packed sieve tray using packing of steel wool type and 5 cm height consisting of 16 trays composed in series and atmospheric pressure for approximately 60 minutes of distillation operation.
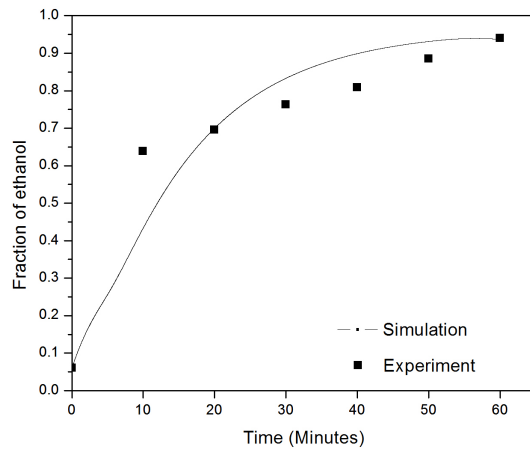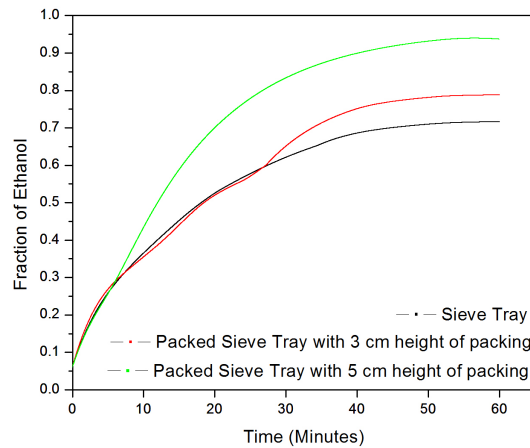

Figure 8 The comparison of ethanol content for various heights of packing consisting of 16 trays composed in series and atmospheric pressure for approximately 60 minutes of distillation operation.

The patterns of ethanol distillate content had the same tendency as shown in Figure 5 to 7 by observing experimental and simulation results. In general, ethanol distillate content would increase until the maximum distillate content that could be achieved was proportional to the duration of the distillation process for approximately 60 minutes. However there were some conditions where there was deviation between the ethanol distillate content by experimental method and the simulation method. There was a pattern in result of the distillate

content by the experimental method under the simulation result, such as in Figure 5 and 7. On the other hand, there was pattern in result of the distillate content by the experimental method above the simulation result, as observed in Figure 6. This might be happened because of several assumptions used in simulation methods such as binary mix was used, bulk phases were assumed to be perfectly mixed, vapor-liquid equilibrium was occurred only in the vapor-liquid interface, the relationship between heat flux and mass fluxes in the interface was ignored, reboiler was assumed to be in a thermodynamical equilibrium state.

Figure 8 shows the effect of using packed sieve tray to ethanol content in distillation. Packed sieve tray gave purer ethanol content than sieve tray. In other hand, the variation of packing heights gave the effect to bubble diameter in distillation process which was illustrated in Figure 10 to 12. The zone distribution in packed sieve tray was illustrated in Figure 9.
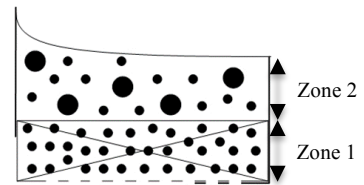

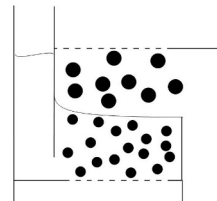**Figure 9** Hydraulic model of the dispersion on packed sieve tray.

Figure 10 Hydraulic model of the dispersion on sieve tray.
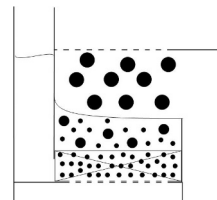

Figure 11 Hydraulic model of the dispersion on packed sieve tray with 3 height of packing.
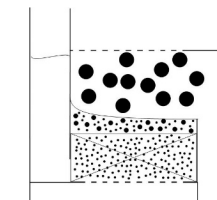
Figure 12. Hydraulic model of the dispersion on packed sieve tray with 5 height of packing

Figure 9 shows the hydraulic model of dispersion on packed sieve tray which was divided in 2 zone distributions. Zone 1 was zone enclosed by packing. In this zone, bubbles were constantly broken down by packing and caused the diameter of bubble to decrease. Zone 2 was zone above the packing. In this zone, bubbles were composed by large bubbles and small bubbles.

The distillation process using packed sieve tray with steel wool gave better tray efficiency than sieve tray. Packed sieve tray increased

the maximum ethanol content by 8.89% for variation of packing height of 3 cm with tray efficiency about 166.4% and 23.31% for variation of packing height of 5 cm with tray efficiency about 231.2% when compared with distillation process using sieve tray with tray efficiency about 77.9%. The increase in ethanol content was due to the use of packed sieve tray in the distillation process that caused the formation of small bubbles. Figure 10 illustrates the size and the number of bubbles which were occurred in sieve tray. The resulting bubbles had a larger diameter and fewer bubbles if they were compared with packed sieve tray which was illustrated in Figure 11 for packed sieve tray with 3 height of packing and Figure 12 for packed sieve tray with 5 height of packing. The decreasing diameter of bubbles could increase the interfacial area and improve the contact time between vapor and liquid in the tray. It also has an effect on increasing mass transfer in the tray so that the pure distillate level obtained was more pure~~ly~~. In addition, the use of packed sieve trays in the distillation process could maintain the mass transfer value and the bubble diameter since the bubbles were consistently broken down by packing of steel wool type.

The increase of mass transfer in each tray was supported by previous research conducted by Spagnolo and Chuang (1984, p 565) and Chen *et al.* (1992, p 213). In the previous study there was an increase in mass transfer in the tray that was influenced by the addition of packing on 1 tray. So the performance of the sieve tray could be increased with the addition of packing on 1 tray because there was an increase in the interfacial area inside the tray.

CONCLUSION

The use of packed sieve tray in the distillation process gave the better result when it compared to the usage of a sieve tray. The distillation process using packed sieve tray with steel wool increased the maximum ethanol content by 8.89% for variation of packing height of 3 cm and 23.31% for variation of packing height of 5 cm, compared with distillation process using sieve tray. The packed sieve tray caused the bubble diameter to form in decreasing manner in each tray. The small size of the bubble could increase the interface area, the contact time between vapour and liquid in the tray and mass transfer so that the ethanol content became purer.

NOMENCLATURE

a'	Geometrical interfacial area per volume of vapour (m²/m³)
a^I	Net interfacial area, (m²)
B	Bottom flow (mol/s)
c_t	Molar density (mol/m³)
d	Column internal diameter (m)
D	Distillate flow (mol/s)
E	Energy balance function
h_t	Specific liquid holdup
H	Molar enthalpy (kJ/mol)
k	Mass transfer coefficient (m/s)
K	Vapor–liquid distribution ratio
L	Liquid flow (mol/s)
L_f	Total liquid mass flow rate (gr/s)
M	Mass balance function
M_t	Total molar holdup (mol)
N	Mass transfer flux (mol/s)
P	Pressure (Pa)
Q	Heat (kW)
T	Temperature (K)
t_G	Mean residence time of vapour-phase in dispersion (s)
V	Vapor flow (mol/s)
G_f	Total vapour mass flow rate (gr/s)
x	Liquid composition
y	Vapor composition

Greek letters

ε	packing void fraction (%)
ρ	density

Subscripts

i	component index
j	stage index
R	reboiler
D	reflux drum
C	condenser

Superscripts

L	liquid related
V	vapor related
I	interface related

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REFERENCES

Afacan, A., Chuang, K. T., Bader, A. J., Sharp, D. 2015. Effect of liquid-phase properties on separation efficiency in a randomly packed distillation column. *The Canadian Journal of Chemical Engineering*, vol. 93, pp 1119-1125.

Burger, A. J., Uys, E. C., Preez., L. J. D., Knoetze, J. H. 2017. The influence of liquid physical properties on entrainment inside a sieve tray column. *Chemical Engineering Research and Design*, vol. 117, pp 205-217.

Bonilla, J., Logist, F., Degreve, J., De Moor, B., Van Impe J. 2012. A reduced order rate based model for distillation in packed columns: Dynamic simulation and the differentiation index problem. *Chemical Engineering Science*, vol. 68, no. 1, pp. 401-412.

Chen, G. X., Chuang, K. T., Chien, C., Ye, Y. 1992. Mass transfer and hydraulic of packed sieve tray. *Gas Separation and Purification*, vol. 6, no. 4, pp. 208-212.

Geankoplis, C. J. 2003. *Transport Processes and Separation Process Principles, 4th edition.* New Jersey: Pearson Education.

Górak, A., Olujić, Ž. 2014. *Distillation: Equipment and Processes.* Oxford, UK: Academic Press.

Kachur, P. J., Afacan, A., Chuang, K. T. 2004. Use of structured packing as a dualflow tray in distillation. *Chemical Engineering Research and Design*, vol. 82, no. 7, pp. 813-820.

Khrishnamurthy, R., Taylor, R. 1985. A nonequilibrium stage model for multi-component separation processes: Part I model description and method of solution. *AIChE Journal*, vol. 31, no. 3, pp. 449-456.

Kumar, S., Singh, N., Prasad, R. 2010. Anhydrous ethanol: A renewables source of energy. *Renewable and Sustainable Energy Reviews*, vol. 14, no. 7, pp. 1830–1844.

Olujić, Ž., Seibert, A. F. 2014. Predicting the liquid phase mass transfer of structured packings. *Chemical and Biochemical Engineering Quarterly*, vol. 28, no. 4, pp. 409-424.

Perry, R. H., Green, D. W. 2008. *Chemical Engineer's Handbook, 8th edition.* New York: Mc. Graw Hill Book Company.

Pilling, M., Holden, B. S. 2009. Choosing tray and packing for distillation. *Chemical Engineering Progress*, September, pp. 44-50.

Ramesh, K., Aziz, N., Abd Shukor, S. R., Ramasamy, M., 2007. Dynamic rate-based and equilibrium model approaches for continuous tray distillation column. *Journal of Applied Sciences Research*, vol.3, no. 12, pp. 2030-2041.

Richardson, J. F., Harker, J. H., Backhurst, J. R. 2002. *Coulson And Richardson's Chemical Engineering Particle Technology and Separation Processes Volume 2, 5ᵗʰ edition.* Oxford, UK: Elsevier.

Seader J. D., Henley E. J., Roper D. K. 2010. *Separation Process Principles, 3rd edition.* Hoboken, New Jersey: John Wiley & Sons.

Smith, R. 2005. *Chemical Process Design and Integration.* Hoboken, New Jersey: John Wiley & Sons.

Spagnolo, D. A., Chuang, K. T. 1984. Improving sieve tray performance with knitted mesh packing. *Industrial & Engineering Chemistry Process*, vol. 2, pp. 561- 565.

Spiegel, L., Knoche, M. 1999. Distillation columns with structured packings in the next decade. *Chemical Engineering Research and Design*, vol. 77, no. 7, pp. 609-612.

Spiegel, L., Meier, W. 2003. Influence of hydraulic conditions on separation efficiency of optiflow. *Chemical Engineering Research and Design*, vol. 81, no. 1, pp. 39-47.

Taufany, Fadlilatul., Soewarno, Nonot., Sutanto, M. E., Girsang, I. R. 2015. Performance of appended wire mesh packing in sieve tray distillation column of ethanol-water system. *Modern Applied Science*, vol 9, no. 7, pp. 148-153.

Treybal, R. E. 1981. *Mass Transfer Operation*. New York: McGraw-Hill Book Company.

Van Ness, H. C., Smith, J. M., Abbott, M. M. 2001. *Introduction to Chemical Engineering Thermodynamics, 6th editionn*. New York: Mc. Graw Hill Book Company.

Van Winkle, M. 1967. *Distillation*. New York: McGraw-Hill Book Company.

Xu, Z. P., Afacan, A., Chuang, K. T,. 1996. Prediction of packed sieve tray efficiency in distillation. *Chemical Engineering Research & Design*, vol. 74, no. A, pp. 893-899.