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Spray distillation model development for bioethanol downstream processing

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Abstract. Spray distillation process as a novel process was developed to overcome the energy requirement of separation and purification of bioethanol. Unlike conventional distillation method that utilizes difference in boiling point to provoke phase change, spray distillation column utilizes the gradient concentration of ethanol contained in the droplets and the surrounding air to provoke transfer of compound out of the droplets. Previous study has developed a basic model of spray distillation and proven that the separation process could be achieved at relatively low temperature of 40°C. However, the basic model, which was based on classical mass, energy, and momentum balances could not describe the process in higher scale. This study aims to develop the better modelling of spray distillation process by taking into consideration vapor flux and diffusion model of throughout the column. The information is required to develop larger scale equipment which is more suitable to be further increased to actual industrial capacity. Modelling show that ethanol vapor travels in the opposite direction of the spray with the affecting variables: Velocity of counter current air (u_{cc}), density of surrounding gas (ρ_g), volume of vapor (V_v), droplet velocity (u_d) and the amount of feed (m_0). Vapor flux model defined as the function of droplet radius (r_c), column radius (r_d), and number of droplets inside the column (N_D), which affecting the sizing of prototype of distillation column in combination with diffusion model. Testing and simulation at 15-80% v/v ethanol water mixture shows that spray distillation performance is comparable with microbubble distillation and pervaporation with better energy consumption.

1. Introduction

The production of bioethanol as greener replacement of fossil fuel is facing several problems. While the fermentation itself has been considered as mature technology, the development of separation and purification process of bioethanol experienced slower pace. The oldest and most common apparatus used in bioethanol purification is a distillation column. However, it holds some disadvantages, such as large installation cost, high operational cost caused by the high energy consumption [1].

Spray distillation process as a novel process was developed to overcome the energy requirement of separation and purification of bioethanol. Unlike conventional distillation method that utilizes difference in boiling point to provoke phase change, spray distillation column utilizes the gradient concentration of ethanol contained in the droplets and the surrounding air to provoke transfer of compound out of the droplets. Mixture of bioethanol and water from filtered and clarified fermentation broth were sprayed into a slightly heated column. The separation of bioethanol from water mixture occurred at the interface between the liquid micro droplets with surrounding air. Spray distillation apparatus utilizes diffusivity,

phase transformation, concentration gradient, and extremely large surface area to separate ethanol from the water mixture. It works at lower temperature compared to conventional distillation process [2].

Previous study has developed a basic model of spray distillation and proven that the separation process could be achieved at relatively low temperature of 40°C. However, the basic model, which was based on classical mass, energy, and momentum balances could not describe the process in higher scale. The flow pattern of the ethanol vapor and the effect of diffusion through the droplets during its journey from the top to bottom of column has not yet studied. The information is required to develop larger scale equipment which is more suitable to be further increased to actual industrial capacity.

This study aims to develop the better modelling of spray distillation process by taking into consideration vapor flux and diffusion model of throughout the column.

2. Materials and method

The model developed for the spray distillation enhancement was divided into vapor flow modelling, vapor flux model development, and diffusion model. Model testing was done in a developed prototype with various ethanol-water mixture concentrations. The material used in this experiment was ethanol 70% v/v technical grade. The experiment was tested against different feed concentration and feed flow rate.

2.1. Vapor flow direction

In order to model the direction of ethanol vapor released from the droplets, momentum balance was generated. Since the prediction was based on the movement of a falling droplet through a specific height, the forces that affect the movement of vapor released from the surface of the droplet were also the same. Said forces could be elaborated as drag force, buoyancy force, and gravitational force. The starting equation for the model is described in Eq 1.

$$\frac{d(mu_v)}{dt} = F_g - F_b - F_d \text{ (eq. 1)}$$

where m is the mass of feed, u_v is vapor velocity, while F_g , F_b , and F_d are gravitational force, buoyancy, and drag force respectively.

2.2. Vapor flux model

Vapor flux is used to represent the velocity of vapor movement through the top outlet of the column. The equation was generated in order to support the flow of vapor through the outlet for vapor collection. It was necessary to determine the factors that influenced vapor flux inside the column. With greater vapor flux, it was predicted that more vapor would go out from the outlet and more chance on collecting the vapor. The vapor flux modelling was based on the evaporative mass transfer equation created by Lupo and Duwig (2017)[3], as presented in Eq 7.

$$\dot{m}_{of \ a \ droplet} = 4\pi\bar{\rho}_g\bar{D}_{v,g}r_d \ln(1 + B_M) \text{ (eq. 7)}$$

Where m is mass evaporative rate of a droplet, $\bar{\rho}_g$ is the gas density, $\bar{D}_{v,g}$ represents diffusivity between the vapor and gas, r_d represents droplet radius, while B_M is Spalding mass transfer number.

2.3. Diffusion model

When droplets of ethanol-water mixture fall in the column, the concentration gradient between the droplet surface with the surrounding air promotes the diffusion or evaporation. Due to the continuous evaporation, droplet radius will decrease from time to time [4]. The previous momentum balance developed assumes that the droplet is free-falling inside the column [2]. However, it is suspected that there is a force that contributes to the velocity of a droplet falling. The force comes from the pressure where the feed is sprayed at the beginning and it will affect the rate of evaporation itself.

3. Results and discussion

3.1. Vapor flow direction

Vapor flow direction is an important aspect because it determines in which section of the column should the outlet be installed. The momentum balance of vapor direction was done by referring to the momentum balance of droplet. According Cai (2018)[5], the momentum balance of a droplet is influenced by several forces. However, due to the extremely small size of droplets, it was assumed that there were only three forces influences the droplet movements as presented in Eq 1.

$$\frac{d(mu_v)}{dt} = F_g - F_b - F_d \quad (eq. 1)$$

Neglecting the small gravitational force surrounding the droplet, the mathematical modelling for predicting vapor direction generated the following equations:

$$\frac{d(mu_v)}{dt} = F_g - f(u_{cc}, \rho_g, V_v) - \frac{A\rho_v C_D u_d^2}{2} \quad (eq. 2)$$

$$\frac{d(m\theta)}{dt} = -f(u_{cc}, \rho_g, V_v) - \frac{A\rho_v C_D u_d^2}{2} \quad (eq. 3)$$

$$m_0 \frac{d(\theta)}{dt} + \theta_0 \frac{d(m)}{dt} = -f(u_{cc}, \rho_g, V_v) - \frac{A\rho_v C_D u_d^2}{2} \quad (eq. 4)$$

$$m_0 \frac{d(\theta)}{dt} = -f(u_{cc}, \rho_g, V_v) - \frac{A\rho_v C_D u_d^2}{2} \quad (eq. 5)$$

$$\frac{d\theta}{dt} = -f(u_{cc}, \rho_g, V_v) - \frac{A\rho_v C_D u_d^2}{2m_0} \quad (eq. 6)$$

The Eq 2 – 6 above stated that the vapor travels in the opposite direction of the spray with the affecting variables as mentioned: Velocity of counter current air (u_{cc}), density of surrounding gas (ρ_g), volume of vapor (V_v), droplet velocity (u_d) and the amount of feed (m_0). Therefore, the large-scale prototype column must be designed in such a way that the vapor outlet is located on the top section of the column.

3.2. Vapor flux model

Vapor flux (θ) plays a huge role in determining the characteristic of vapor flow inside the column. It was done by considering the evaporation rate of droplet surrounded by gas phase. According to Lupo and Duwig (2017)[3], mass evaporation rate of a droplet is as stated in Eq.7.

$$\dot{m}_{of\ a\ droplet} = 4\pi\bar{\rho}_g\bar{D}_{v,g}r_d \ln(1 + B_M) \quad (eq. 7)$$

The mass evaporative rate of a droplet was altered in order to suit the case of spray distillation column and to predict the velocity of vapor flowing inside the column. Since vapor flux equation aims to predict the rate of vapor that travels inside the column and there would be more than one droplet produced by the nozzle, further transformation of the equation could be observed below:

$$\dot{m}_{of\ a\ droplet} = 4\pi\bar{\rho}_g\bar{D}_{v,g}r_d \ln(1 + B_M) N_D \left[\frac{kg}{s} \right] \quad (eq. 8)$$

$$V = 4\pi \frac{\bar{\rho}_g}{\bar{\rho}_v} \bar{D}_{v,g} r_d \ln(1 + B_M) N_D \left[\frac{m^3}{s} \right] \quad (eq. 9)$$

$$\theta = 4 \frac{\bar{\rho}_g}{\bar{\rho}_v} \bar{D}_{v,g} \ln(1 + B_M) N_D \frac{1}{\pi r_c^2} \left[\frac{m}{s} \right] \quad (eq. 10)$$

$$\theta = 4 \frac{\bar{\rho}_g}{\bar{\rho}_v} \bar{D}_{v,g} \frac{r_d}{r_c^2} \ln(1 + B_M) N_D \left[\frac{m}{s} \right] \quad (eq. 11)$$

Where θ represents vapor flux, $\bar{D}_{v,g}$ represents diffusivity between the vapor and gas, r_d represents droplet radius, r_c shows the column radius, $\bar{\rho}_v$ shows the density of vapor species, $\bar{\rho}_g$ is the density of the gas surrounding the droplet, and N_D shows the amount of droplets. Vapor flux itself, could be defined as the function of droplet radius (r_c), column radius (r_d), and number of droplets inside the column (N_D) as shown in Eq.12.

$$\theta = f(r_d, r_c, N_D) \quad (eq. 12)$$

High vapor flux is desired because with higher flux means that a faster and higher rate of vapor flow took place in the system. It is beneficial for condensation because with more flux means the possibility of condensation would also increase. The modelling also resulted knowing in three factors contributing to vapor flux, name: droplet radius, column radius, and the number of droplets.

3.3. Diffusion model

Diffusion model took into account the effect of water evaporation in the modelling of droplet diffusion, thus changing the momentum balance by studying the initial velocity of the feed. It was then established that the height of the spray distillation column was influenced by the droplet concentration; meaning changes in droplet radius over time was taken into consideration. The diffusion model also consents a relationship between initial velocity, feed rate, and number of sprays. In addition, because there were observable changes in the effect of water evaporation towards the process, the diffusion model has established that the critical process and design parameters were column height and number of sprays.

With the additional effect of water evaporation, the modelling for ethanol and water fraction can be calculated as follow:

$$Vles_{t+\Delta t} = \frac{\frac{bVles_t r_t^3 - ar_t \left(\frac{Yves_t}{\sum iYvis_t} \right)}{\rho_e}}{bVles_t r_t^3 - ar_t \left(\frac{Yves_t}{\sum iYvis_t} \right) + \frac{c(1-Vles_t) r_t^3 - ar_t \left(\frac{Yvws_t}{\sum iYvis_t} \right)}{\rho_w}} \quad (\text{ethanol fraction})$$

$$Vlws_{t+\Delta t} = 1 - Vles_{t+\Delta t} \quad (\text{water fraction})$$

Thus, contributing to the change in ethanol and water evaporation rate over time as:

$$\frac{dF}{dt} = \frac{d(u_d)}{dt} = \frac{\frac{4}{3}\pi r^3 \rho g - \frac{4}{3}\pi r^3 p_g g - 4\pi r^2 p_g C_D \frac{u_d^2}{2} - \frac{\dot{V}_F}{\pi r_d^2 N_s} \frac{m_{end} - m_{start}}{\Delta t}}{\frac{4}{3}\pi p_0 r_0^3}$$

both variables affected the modelling in terms of its momentum balance as both affects the height of the column. The diffusion model assumes that there is an optimum time for evaporation where ethanol fraction is greater than water fraction, which is the ideal condition for achieving high purity and high yield outcome. Therefore, predicting that with different feed ethanol concentration required a different height of column.

3.4. Model simulation

Several feed concentrations were tested, which were 15%, 30%, 45%, 60%, and 70% (all in v/v %). Observation of the prototype shows that condensate was formed on the collection area despite its low volume thus making direct measurement of ethanol purity from the top product still a problem to decipher. It was observed that higher feed concentration led to less condensate; on the other hand, a higher feed rate is the opposite, it contributes to the likelihood of more condensate formation.

Measuring the decrease of ethanol concentration of bottom product in comparison to the feed concentration, shows that separation has been achieved. Comparison of separation factor between spray drying as modelled in this study with other separation method is presented in Table 1.

Table 1. Performance comparison of spray distillation to other distillation method [1,6]

Feed concentration (% v/v)	Spray distillation (36 ml/ min, 40°C)	Microbubble distillation (150°C)	Pervaporation (50.88 mmHg, 60°C)
15	9.71	11.2	9.70
30	5.47	8.40	4.50
45	3.55	6.00	N/A
60	2.38	3.60	N/A
70	1.81	N/A	N/A

From Table 1 it is observable that the performance of the prototype developed based on the modeling done in this study provide comparable performance to other downstream processing method used for bioethanol separation. The fact that spray drying is operating at much lower temperature compared to microbubble distillation shown the potential of further development of the process into low energy method for separation. Spray drying process is also in comparable performance with more advance pervaporation process, but without the need to create the vacuum condition.

4. Conclusions

The diffusion model which takes into account the effect of evaporation of water results in prototype design of column size and number of sprays installed. The diffusion model established as well that the critical processing parameters of spray distillation are feed rate and concentration. The model developed for the spray distillation process was successfully resulting in working prototype with comparable performance with other ethanol-water separation process.

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