HETP measurement using industrial-scale batch distillation

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ABSTRACT

The height equivalent to a theoretical plate (HETP), representing the mass transfer efficiency, allows designers to determine the packed bed height. In this study, a novel methodology was proposed to evaluate the HETP using an existing industrial-scale batch distillation when designing the height of an industrial-scale continuous distillation column. Specifically, an existing batch distillation used for the reclamation of dichlorodifluoromethane CCl2F2 (R-12 or Freon-12) from a waste refrigerant mixture was employed to determine the HETP value. Prior to the simulation, a literature survey was investigated to determine the suitable property package for simulation. The simulation results were then compared with the experimental data to determine whether REFerence fluid PROPERTIES can be used as a suitable package for this study. Several batch distillation runs were carried out using industrial structured packing and random packing, which are candidates for the design and optimization of a continuous distillation column in this specific separation. The batch distillation was then rigorously simulated for a comparison with the experimental data, enabling us to calculate the HETP value. The simulated results agreed well with experimental data, demonstrating that the proposed methodology is effective and easy to apply.

1. Introduction

The distillation process, which is the most commonly employed process in the chemical and petrochemical industries, is normally conducted in either continuous or batch mode [1]. Installation of the equipment can be achieved using a tray or packing, which is the main aspect contributing to the performance of such equipment [2,3]. Lower pressure drops can be achieved when using columns packed with random and structured packing as compared to trayed distillation towers. Their generally shorter height, mechanical simplicity, ease of installation, and ability to be fabricated in a cost-effective manner from corrosion-resistant materials are other advantages of packed distillation columns [4].

Random packing is known for its easily installation, blocking resistance, and acceptable-efficiency devices for distillation columns [5]. Currently, approximately 200 different types and sizes of random packings are available from different manufacturers worldwide [6]. The larger the random packing, the higher the capacity, but at a cost of lower efficiency [7]. A smaller packing possesses a higher efficiency but a lower capacity and higher cost. The appropriate packing should be selected with the most economical balance between capacity and efficiency. Structured packing can be considered for a lower pressure drop. In addition, other advantages of a structured packing are usually realized at a high surface area and high separation efficiency [8–11]. However, they are considerably more expensive per unit volume than random packing and are susceptible to corrosion [10].

The performance of packed distillation columns is frequently
expressed in terms of the height equivalent to a theoretical plate (HETP) [12]. HETP representing the mass transfer efficiency is an empirical but extremely practical parameter [13]. The number of theoretical stages required for a specific separation and the HETP for a particular type of packing are both used to determine the actual height of the packing required to achieve the desired separation. Note that height of transfer unit (HTU) can also be considered to estimate the packed-height, although this HETP approach is usually preferred. This is because the HETP approach is suitable for multicomponent systems, can use the stage-by-stage computer programs, and enables easier comparison with plate columns, while the HTU approach is more complex and more difficult to use [8].

Three approaches are commonly used for HETP prediction, namely, mass transfer models, rule of thumb, and data interpolation [8]. The mass transfer models first determine the height of a transfer unit (HTU), which is then used to calculate the value of the HETP [14,15]. The accuracy of the model clearly depends on the accuracy of the correlations used for predicting the basic parameters, i.e., the mass-transfer coefficients for the gas and liquid phases, respectively, and the effective interfacial area [16]. In addition, because there are only a few variables that significantly affect a random packing HETP, and owing to the unreliability of even the best mass transfer model, a rule of thumb for the HETP competes successfully with mass transfer models [2,8,17]. Furthermore, an interpolation of experimental HETP data is another means of obtaining the designed HETP values [18].

2. Problem statement

The HETP is essential for designers to estimate the height of a packing column. HETP is determined as

\[
\text{HETP} = \frac{H}{n}
\]

(1)

where \(H\) is packed bed height and \(n\) is the number of actual stages.

Despite the significant amount of data available, no method yet exists for HETP prediction with a high degree of confidence [17]. The HETPs are different for different packing sizes and types and for different chemical systems [19]. The HETP of structured packing is also affected by the pressure (high pressure), vapor, and liquid load [8]. Thus, it would be better if the HETP can be estimated experimentally for the same mixture and similar operating conditions.

In this study, a novel methodology is proposed to estimate the HETP used to design the packing height of a continuous distillation unit using the existing industrial-scale batch distillation unit. An existing industrial-scale batch distillation unit used for reclaiming dichlorodifluoromethane \(\text{CCl}_2\text{F}_2\) (R-12 or Freon-12), which is the most common representative chlorofluorocarbon (CFCs), from a waste refrigerant mixture was used to estimate the HETP value of the desired packing. Prior to the simulation, a literature survey was carried out to determine the suitable property package required for simulating the refrigerant mixtures. Based on a comparison between data from simulated Pxy diagram and the experimental data, the thermodynamic model for evaluating the vapor-liquid equilibrium (VLE) behaviors can be selected. An experimental operation using the existing industrial-scale batch distillation unit was carried out, and the obtained results were compared with the simulation results to determine the HETP value of two industrial structured packings and random packing NMTP 15.

3. Methodology

3.1. Materials

An illustrative example is the purification of R-12 from a waste refrigerant mixture. A mixture including R-12 and 1,1,1,2-tetrafluoroethane \(\text{CH}_2\text{FCF}_3\) (R-134a) was obtained from OunR2tech (Korea). The waste refrigerant was recovered in a liquid state and injected into the evaporator; the liquid refrigerant was then evaporated in the evaporator to treat the impurities such as oil and water in the mixture.

3.2. Apparatus

3.2.1. GC apparatus

The samples collected for the batch distillation run were analyzed through a GC analysis, which was carried out using an Agilent 490 equipped with a flame ionization detector (FID) and fitted with a 20 m \(\times\) 0.25 mm column programmed at 120 °C. The sample injection volume is 10 μL. Helium was used as the carrier gas under a pressure of 0.6 MPa and a flow rate of 0.5 ml/min.

3.2.2. Batch distillation system

Batch processing is commonly used in the pharmaceutical, biochemical, and specialty chemical industries. This process is preferable to continuous distillation when small quantities of chemicals and biochemicals need to be separated [20,21]. The most prominent characteristic of batch distillation is its flexibility, allowing one to deal with uncertainties in the feedstock or product specifications. Another advantage of batch distillation is that it is able to separate a multi-component mixture with only a single column.

In this study, an industrial-scale batch distillation used for R-12 reclaimation was employed for determining the HETP. After the refrigerant was separated and purified from other heavy components using the evaporator, the components were fed into the batch distillation (shown in Fig. 1). An industrial-scale batch distillation unit constructed at Ounr2tech was tested using wire gauze structure packing HMP 500 W G or wire gauze structure packing HMP 700 W G, or random packing NMTP 15 as suggested by the vendor. Table 1 shows the existing hydraulics and product specifications of the industrial-scale batch distillation column. In this system, the control panel allows establishing and controlling the operating parameters such as the hot water temperature, cold water temperature, and collector level during the entire experiment.

3.3. Simulation

Aspen Hysys was used to select suitable property package through a comparison with the experimental data in the Pxy equilibrium phase diagram. Furthermore, to estimate the HETP value of the specified packing, an Aspen Batch Modeler V10 was employed to simulate the batch distillation, the results of which were compared with those of an experiment on a real industrial-scale batch distillation.

4. Results and discussion

4.1. Phase equilibrium

Because a property package was required for the simulation of R-12 and R-134a, a literature survey was carried out. As a result, the REferencE fluid PROPerties (REFPROP) method was found, which provides the thermodynamic and transport properties of industrially important fluids and their mixtures with an emphasis on refrigerants and hydrocarbons [22,23]. The literature data [24] on a Pxy equilibrium phase diagram were used to compare with the simulation results using REFPROP in Aspen Hysys DB. As shown in Fig. 2, the Pxy curves matched the experimental data closely. Thus, REFPROP was selected as the property package for the prediction of the vapor-liquid equilibrium and all simulations conducted in this study. All simulations were applied using Aspen Hysys and Aspen Batch Modeler V10 simulators.

A mixture of R-12 (boiling point of -29.8 °C) and R-134a (boiling point of -26.3 °C) has an azeotrope with a composition of approximately 60 mass% R-12, and an azeotropic temperature of approximately -34.5 °C. Although it is easier to separate at 1 atm (1.013 bar), the
temperature is then too low, as shown in Fig. 3. Thus, the column should be operated at a higher operating pressure. A column pressure of 8 bar was selected to allow the available heating and cooling water to be applied together. Please note that the cooling water and hot water available together are at 13 °C and 48 °C, respectively.

4.2. Batch distillation operation

The feed contains 1280 kg of 91.31 wt% R-12 and 8.69 wt% R-134a (shown in Table 2). The required product purity is 99.5 wt% R-12 in the generator or reboiler. In addition, 320 kg for the collector, which can balance the reboiler duty and recovery of R-12, was selected. Since the density of this mixture is 1242 kg/m³, the volume of liquid in the collector was 0.2576 m³, which is equivalent to the 0.407 m height of the liquid in the collector. This value was used to set the collector level controller. The existing batch distillation unit has the following reflux

<table>
<thead>
<tr>
<th>Tray type</th>
<th>Packing</th>
</tr>
</thead>
<tbody>
<tr>
<td>Bed height (mm)</td>
<td>2550 / 2668</td>
</tr>
<tr>
<td>Column diameter (mm)</td>
<td>102</td>
</tr>
<tr>
<td>Max flooding (%)</td>
<td>84</td>
</tr>
<tr>
<td>Energy requirement of reboiler (W)</td>
<td>5500</td>
</tr>
<tr>
<td>R12 purity (mass%)</td>
<td>99.5</td>
</tr>
</tbody>
</table>

Fig. 1. (a) Existing industrial-scale batch column and (b) simplified flowchart illustrating the existing batch column for R-12 reclamation.

Fig. 2. R-134a + R12 VLE phase diagram compared with values from the literature at (a) -15 °C, (b) 5 °C, and (c) 25 °C.
operating and vapor loading policies:

Fig. 4 shows the operation procedure. Before starting the batch distillation, the column and collector were prepared under vacuum conditions, whereas at the start, the generator was filled with a charge (1280 kg of liquid at ambient temperature (18 °C), 4 bar). The operation was then started by opening the valve in the vapor line of the reboiler, followed by turning on the pumps for cooling and hot water. Initially, the heat transfer rate is high because the temperature difference between the generator liquid and hot water is high. The liquid then starts to boil. The concentration of the more volatile component and the volume of the liquid decrease, and the column fills up with the liquid hold-up.

There was no reflux during the first period (approximately 4–5 h). During this period, all vapor was contained in the collector after being totally condensed. When the height of the collector was 0.407 m, which occurs on a case-by-case manner, as estimated above, the reflux pump started to operate to generate a reflux flow returning to the column. The collector level was then controlled and maintained by the reflux pump (shown in Fig. 1). Thus, the top vapor flow rate is equal to the reflux flow rate. Another important variable during batch distillation is the vapor loading of the reboiler. The reboiler was operated under a constant duty cycle and was set at the highest capacity without flooding in the column.

Because there is no liquid withdrawn from the column, the light component is gradually accumulated in the collector, while the concentration of less volatile component increases in the generator. Thus, during the operation, the generator temperature increases. Samples of the generator were taken for further mass quantification of R-12 using the previously described analytical methods. As time proceeds, the material in the generator becomes less rich in the more volatile components, and its vapor flow rate reduces because the reboiler duty remains constant. This process is continued until the specifications for the residue in the generator are satisfied. After achieving the target purity of R-12 in the generator, the batch distillation is stopped and all remaining components inside the column and collector are removed. Vacuum conditions are then generated in the column and collector before another batch can be started.

4.3. Batch distillation simulation

The batch distillation was then simulated using Aspen Batch Modeler. Table 3 shows all parameters needed for simulation of the existing batch distillation. All operating policies used in experiments can be implemented in Aspen formats. REFPROP was used as the property package. The zero distillate rate can be set in Aspen Batch Modeler. The operation includes two steps. The first step is a feed charging step, while the heat is supplied in the second step. During the second step, the collector level of 0.407 m was kept constant by setting in Aspen Batch Modeler. Several simulation runs were applied with

![Fig. 3. Temperature vs. composition plot at (a) 1 and (b) 8 bar.](image)

Table 2
Feed mixture conditions.

<table>
<thead>
<tr>
<th>Component</th>
<th>Mass Fractions (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>R-12</td>
<td>91.31</td>
</tr>
<tr>
<td>R-134a</td>
<td>8.69</td>
</tr>
<tr>
<td>Feed weight (kg)</td>
<td>1280</td>
</tr>
<tr>
<td>Temperature (°C)</td>
<td>18</td>
</tr>
<tr>
<td>Pressure (bar)</td>
<td>4</td>
</tr>
</tbody>
</table>

![Fig. 4. Operation procedure.](image)
different numbers of stages using the Aspen Batch Modeler.

Notably, in the Aspen Batch Modeler, if a start-up of the batch column or the use of an initial charge is required to simulate, a partial condenser is needed because the column is initially filled with nitrogen or air, and it is unrealistic to completely condense air or nitrogen which enters the condenser during the start-up.

4.4. Comparison between simulation and experiment

4.4.1. HMP 500 W G structured packing

In the first case study, because wire gauze structure packing HMP 500 W G shown in Fig. 5a is a candidate for continuous distillation in this project, it was tested to evaluate the HETP used to estimate the designed column height. Structured packing has a well-defined geometrical structure, which consists of an alternating arrangement of corrugated sheets forming intersecting open channels for a vapor flow [13]. Wire gauze structured packing is generally used in medium- to small-diameter columns for separations requiring a maximum number of theoretical stages at the minimum column height [25].

The samples collected for batch distillation were analyzed using GC. Fig. 6 shows the history of the composition purity. For a given same packed bed height, each kind of packing has its own HETP value or mass transfer efficiency, which leads to different number of stages and different operating time to achieve target purity. Several simulation runs were then carried out with different numbers of stages using the Aspen Batch Modeler. The experimental variations of the generator composition and temperatures based on the operating time were compared with the simulation results. The number of stages having the purity history and operation time matched well with that of real operation was finally selected. As the results indicate, the purity history curve under eight stages matched with that of the real operation most closely. Thus, the HETP, which was calculated by Eq. (1), was approximately 319 mm under these operation conditions. Furthermore, the minimum number of stages can be found to be seven under these conditions.

Fig. 7 shows the temperature histories at the generator of the column for both the simulation and experimental results, which demonstrate extremely good agreement. Furthermore, it took 28.7 and 28.3 h during the simulation and real operation, respectively, to achieve 99.5 % R-12, which indicates that simulated results are in agreement with those of the experimental results.

4.4.2. HMP 700 W G structured packing

Wire gauze structure packing HMP 700 W G, shown in Fig. 5b, was developed by Hanbal Masstech Co. with the aim to increase the efficiency, which can lead to a reduction of the HETP. Fig. 8 shows the variation of the composition of R-12 during both the experiment and simulation. The experimental curve is located near the curves when the number of stages is 9 and 10. Thus, the HETP is approximately 267–296 mm for this specific separation. It took 24.2 and 23.6 h during the simulation and real operation, respectively, to achieve 99.5 % R-12. The real batch distillation column allowed an R-12 product to be achieved at the generator of the column.

Fig. 9 shows the temperature histories of the generator of the column for both the simulation and experimental results, which have extremely good agreement. It took 28.7 and 28.3 h for the simulation and real operation, respectively, to achieve 99.5 % R-12. The real batch distillation column allowed R-12 to be obtained at the generator of the column. In general, a good agreement was obtained between the

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**Table 3**

Parameters for the batch distillation model.

<table>
<thead>
<tr>
<th>Species</th>
<th>R-12 and R-134a</th>
</tr>
</thead>
<tbody>
<tr>
<td>Number of stages</td>
<td>Varied</td>
</tr>
<tr>
<td>Distillate flow rate</td>
<td>0 kg/hr</td>
</tr>
<tr>
<td>Generator geometry diameter</td>
<td>1 m</td>
</tr>
<tr>
<td>Collector geometry diameter</td>
<td>1 m</td>
</tr>
<tr>
<td>Pressure</td>
<td>8 bar</td>
</tr>
<tr>
<td>Initial conditions</td>
<td>Empty</td>
</tr>
<tr>
<td>Operating steps</td>
<td></td>
</tr>
<tr>
<td>Step 1</td>
<td>Charging (1280 kg)</td>
</tr>
<tr>
<td>Step 2</td>
<td>Duty 5 kW</td>
</tr>
<tr>
<td>Collector level controller</td>
<td>0.407 m</td>
</tr>
</tbody>
</table>
simulation and experimental results, demonstrating the capability of REFPROP for modeling the thermodynamic behavior of the VLE between R-12 and R-134a. Note that the flooding is 87.1% under these operating conditions (from the Aspen Batch Modeler).

4.4.3. NMTP 15 - random packing

NMTP 15 packing (Fig. 5c), which is applicable in a broad range of services, was also investigated to measure the HETP. NMTP tower packing has a high void fraction providing a superior high liquid rate...
capacity as well as a lower pressure drop compared to other random packings of similar size [25,26]. This packing combines the advantages of a saddle-shape packing with those of modern high-performance ring-type packings [27]. The vendor has suggested using this packing for the separation of R-12 and R-134a. Thus, it was tested to measure the HETP for this specific separation.

At the beginning of the operation, the generator is filled with R-12 and R-134a. As soon as the operation starts, R-134a tends to move toward the collector, leading to a low composition in the generator, where R-12 is dominant during the final period. The pressure drop was 0.1-0.2 bar. The heat loss was ignored because the batch column system is well insulated. Very good agreement exists between the experimental and simulation results when nine stages are applied, leading to an HETP of 296 mm. Fig. 10 shows the composition and flooding profiles at the end of simulation. Note that, under atmospheric conditions, the HETP value from the vendor is 250 mm (shown in Table 4) because, in addition to the packing type and size, the HETP values are complex functions of the pressure, vapor and/or liquid flowrates, distribution, and physical properties, among other factors. For greater reliability, it is therefore recommended to estimate the HETP for a specific separation using a rigorous methodology.

Because the batch distillation and designed continuous distillation have almost same pressure to separate the same system, HETP estimated using batch distillation has counted the effects of pressure. In other words, with this novel methodology, the effects of pressure can be eliminated when estimating HETP and the height of the continuous column.

5. Conclusions

This study proposed a simple and effective methodology for estimating the HETP using an existing industrial-scale batch column. The REFPROP method has demonstrated a good capability to predict the VLE of R-12 and R-134a. The proposed methodology was tested using HMP 500 W G and HMP 700 W G structured packings and random packing NTMP 15 through a study on the current industrial separation of R-12 and R-134a. Experimental results of a commercial batch distillation column showed good agreement with the simulation results. This novel methodology can be accomplished easily and efficiently using Aspen Hysys, Aspen Batch Modeler, and an existing industrial-scale batch distillation unit. For reliability, it has been recommended to apply a rigorous methodology when estimating the HETP for a specific separation. The effects of pressure, distribution and physical properties can be eliminated when estimating HETP and the height of the continuous column.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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