Pressure-driven batch distillation optimal control for ethanol-water separation

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Abstract

Ethanol-water separation by pressure-swing batch distillation in a double-batch rectifier system is a process under investigation. In this work, a complete global solution to the optimal control problem in the form of a sequential synthesis of controlled trajectories is derived. During this study, the optimal reflux strategy through cyclic operation was extended to the separation of a second non-ideal minimum boiling azeotrope of industrial significance. Under the pressure force, the process driven by the increasing pressure from different "directions", ie. from atmospheric to high-pressure, is researched.

Keywords

pressure-swing, batch distillation, ethanol-water, optimal control, direct method, Pontraygin’s Maximum Principle

1. INTRODUCTION

Dussel and Stichlmair (1995), investigated the separation of different binary mixtures by extractive distillation: among them, ethanol-water separation by ethylene glycol, whereas, a hybrid process proposed replacing one distillation step by the absorption operation, at the top of the column high boiler is fed in the first step in order to absorb the water from the vapor arising from the water-rich mixture place in the decanter, enriched in glycol, whereas, in the second step, water and ethylene glycol are rectified, presented the course of distillate concentrations and boundaries on ternary diagrams. In the same year, Watson et al (1995), detailed the industrial operating cyclic method to separate a quaternary mixture of methanol-cyclohexane-ethanol-water: providing the possibility to concentrate binary heteroazeotrope of methanol-cyclohexane at very high reflux ratios: two periods of “drainage” interconnected by the total reflux period were necessary to concentrate the maximum of the heteroazeotrope, which is to disappear in the next step of finite reflux. This is possible only due to the fact that the initial mixture is poor in methanol, afterwards, even four infinite reflux periods intertwined with periods of drainage, were necessary to concentrate the binary homoazeotrope of ethanol-water, finally to produce ethanol, started after 10.3h, and last till 15 h, ie. 4.7 h in total. It is worth noting, that “periods of drainage” were gradually increased, but one should note that the second drainage period’s total duration is longer than the first by 46.3088%, consecutively, the third drainage period is longer than the second by 31.5468%, and, the fourth longer than the third for 24.0698%. A particular step duration decrease (%) , (third-second) vs (second-first) is 14.7620%, but, (forth-third) vs
(third-second) only 7.4770%. Furlonge (2000), detailed the comparison of breaking the binary homoazeotrope of ethanol-water, in regular extractive batch column vs non-conventional configurations (middle vessel), according to the author unlike the regular column the operating policy involves a relatively long initial total reflux period, whereas ethanol composition in distillate vs time remained well above the azeotropic point for more than 30% of the total time, thanks to the high solvent feed rate. Tavan and Hosseini (2014), investigated the continuous reactive distillation process for the system containing even 4 azeotropes, one of them heterogeneous, two feeds are introduced into the column, ethanol/water, and ethane: interesting to note is that the total duty (en.) via stages, for the reflux ratio ranged [3-4], keeps fluctuating in the interval (13000-17000 [kW]), also, these functions are different with the respect to the azeotropic feed inlet stage - reflux ratio interval ranged [0.5-2.5], the total duty is ranged in the even broader interval (2000-14000 [kW]), in particular reboiler duty up to 10000 [kW]. Esteban-Decloux et al. (2014), tempted to propose the best startegy for the separation of the particular complex industrial mixture issued from spirit plants: important was to “track” four components present initially in traces, to observe that D-limonene reached the maximum during the production of approx. 0.2 or up to 20 times increase, but, an even greater jump could be stated for linalool oxide, as approx for a hundred times. Iqbal and Ahmad (2016), detailed the professional software designed (HYSYS) optimal operation results, whereas, defined the optimal reflux ratio for HP (column) as greater than for LP (column) for approx. 13.5%, whereas, the ratio of distillate to feed flowrate (like “liquid ratio for batch distillation”), greater for even 43.75%. Heras-Cervantes et al. (2019), a special case of fuzzy-logic control is introduced, ie. Takagi-Sugeno controller, significantly, among the three premise variables the reflux valve opening percentage is placed as recognized to influence all the states of the system together with light component compositions in condenser/reboiler. Most recently, Putri et al. (2022), showed from normalized product concentration output from a particular neural network approach, observed the “quasi zero-bang” sequence in the very beginning, fluctuating further around a mean value (of 0.8), subsequently dropped for “quasi-zero” almost instantaneously to switch to the instantaneous increase in the next moment, however, the combined control made up from neural network and Kalman filter, (“smoothened”) resulted in lowering the “quasi-bang-zero” sequence (from previous maximum 1 or 0.99) to less than 0.95.
2. MATERIALS AND METHODS

2.1 Optimal control problem

In this short communication, the author aims to expand the optimal control strategies to the separation of the other mixtures with minimum boiling azeotrope, by pressure-swing batch distillation. In particular, a case of ethanol–water is taken into examination, due to its significance in the beverage and food industry, as well as the possibility to expand further the study to the more complex mixtures of alcohols. The optimal control problem can be described as follows: For a given pressure-swing batch distillation configuration \((N)\) total number of trays, \((P)\) working pressure, batch composition, the distillation task, and overall time horizon \((t_f)\), determine the optimal reflux ratio so as to maximize the distillate, subject to any constraints (model equations, bounds on the optimization variables).

![Figure 1: A regular double-rectifier scheme for pressure-swing batch distillation](image_url)

The pressure-swing process scheme presented in Figure 1, assumes “one-pass” through the column at the time, in the other words it is assumed that columns work consecutively (“two-step” process), in double batch rectifier configuration. But, the maximum achievable distillate concentration at lower pressure \(P_1 = 1\) atm is 95.5% of ethanol, but, at \(P_2 = 10\) atm only 82% of water. Moreover, in Tables 1–3, the parameters corresponding to thermodynamics model Wilsin, and, originated from Gmehling et al. (1981), are tabulated, and verified with the work of the authors Binous and Al-Hathi (2014).

**Table 1. Binary interaction parameters for Wilson model**

<table>
<thead>
<tr>
<th>Component i</th>
<th>Component j</th>
<th>(A_{ij}) [cal/mol]</th>
<th>(A_{ji}) [cal/mol]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ethanol</td>
<td>Water</td>
<td>-2.5035</td>
<td>-0.0503</td>
</tr>
</tbody>
</table>

**Table 2. Antoine parameters for ethanol from Dechema**
Table 3. Antoine parameters for water from Dechema

<table>
<thead>
<tr>
<th>n°</th>
<th>A</th>
<th>B</th>
<th>C</th>
<th>T_{\text{min}}, [°C]</th>
<th>T_{\text{max}}, [°C]</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>8.20417</td>
<td>1642.89</td>
<td>230.3</td>
<td>-57</td>
<td>80</td>
</tr>
<tr>
<td>2</td>
<td>7.68117</td>
<td>1332.04</td>
<td>199.2</td>
<td>77</td>
<td>243</td>
</tr>
</tbody>
</table>

Table 4: Working conditions

<table>
<thead>
<tr>
<th>Parameter</th>
<th>$P$ [atm]</th>
<th>$U_N^0$ [l]</th>
<th>$U_f^0$ [l]</th>
<th>$V$ [mol/h]</th>
<th>$t_f$ [h]</th>
<th>$\chi_N^0$</th>
<th>$y^*$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Predefined/initial value</td>
<td>LP: 1</td>
<td>10</td>
<td>0.1</td>
<td>4</td>
<td>1.6</td>
<td>0.1</td>
<td></td>
</tr>
<tr>
<td></td>
<td>HP: 10</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>LP: 1.0.85 or 2.0.95</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>HP: 0.82</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

** 1. purity set at 95%, 2. purity set at 85%

Table 5: The optimal results by BOCOP solver

<table>
<thead>
<tr>
<th>Product recovered</th>
<th>$N$</th>
<th>$U_0(t_f)$</th>
<th>Recovery rate</th>
<th>Discr. scheme, nb. of points</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ethanol</td>
<td>21</td>
<td>1. 1.8340</td>
<td>1. 66.26% (single period) against 93.11% (quadruple, 2. 47.19% (single period) against 90.29% (double period)</td>
<td>Gauss, 2200</td>
</tr>
<tr>
<td></td>
<td></td>
<td>2. 1.1138</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Water</td>
<td>10</td>
<td>0.748</td>
<td>1. 72.20% (quadruple) 76.46% (5 periods)</td>
<td>Gauss, 2200</td>
</tr>
</tbody>
</table>

(*) 1. 95 %, 2. 85 %
Discussion for the interesting findings for the desired product purity: first strategy seeking 94%, yielded more than 90\% only after a quadruple period of time, 4 consecutive runs, ie. final time of 3.2 [h], whereas the second proposed strategy where only 85\% of purity seeking, yielded its maximal only after a double period (1.6 [h]).

The HP run, for the first strategy proposed, could last for effectively 0.2 [h], whereas all the control trajectory is constituted from two consecutive bang arcs, concatenated by a very short singular arc, lasting not more than 3E-3 [h]: it seems that for this short interval, the instantaneous switching to the at least two extremely short piques reaching their minimums in the period of 1.8-1.4 and maximums at approx 2.2-2.4. The second bang arc is of about double more duration compared to the very first one, whereas the final zero arcs appears for not more than 5\% of the total batch time.

Moreover, then the HP step should be repeated for even four times, ie. “quadruple period” in order to achieve more than 70\% of the recovery, and even five periods necessary to achieve more than 75\%, for the desired purity of 82\%, ie. slightly more than an azeotropic point.

Otherwise if again, the relaxation on the purity constraint is introduced, 95\% for the first case, but, 85\% for the second case, then it is suggested for the minimum batch time, to be 1h, (LP+HP), whereas for the satisfactory recovery, It is suggested for the minimum batch time of the two-step process, (LP+HP), 1) in the case of the first proposed strategy: (3.2+0.2 *4 = 4.0 [h]), 2) for the second proposed strategy: (1.6 + 0.2*4 = 2.4 [h]). It is to examine, which strategy would bring more in terms of overall performance, (ie. yield, productivity, desired purity, and most importantly energy requirement). As longer running the operation, the cost of power utilities will be higher heating, and electric energy), assuming they are constant with time. Otherwise, a cyclic strategy should be additionally proposed for a particular case.

The optimal control policies for the azeotropic mixture of ethanol-water are depicted in Figure 2, as the LP and HP step, both in the batch rectifier, respectively.
Moreover, in Figure 3, the output for the temperature evolutions along with the optimal solution is presented. Above, the temperatures on all the stages of the column for the rectifier (LP step) are depicted. From here, the interval of temperature increases along with the first rectifier, going from the bottom to the top (batch to accumulator), following the optimal trend from the minimal temperature of 78.3223°C to the maximal one at 100.0962°C, at the specified working pressure of \( P_1 = 1 \) atm. As previously stated, verify that the ethanol is recovered in the product tank. Below, the optimal temperature evolutions are presented for the HP step of the process, ie. the second rectifier, at the specified working pressure of \( P_2 = 10 \) atm, temperature rise, in the same sense, following the trend within the temperature interval from the minimal temperature of 151.5540°C to the maximal temperature of 177.2374°C. Hereby, previously written goes in favor of the fact that ethanol is recovered (again) in the first rectifier column product tank. As a consequence, it is verified, that the ultimate goal of the designed two-step process, ie. consecutive production of ethanol and water, is achieved.

![Figure 3: The optimal temperature evolution for the: 1) left: LP step, 2) right: HP step](image)

### 3.1 The influence of vapor boil-up

The influence of vapor boil-up on the structure of the optimal control pattern is observed, as well. In Figure 4, on the left/right, respectively, the optimal control patterns for purity constraint set at 85%/95%, respectively, are plotted for varied values of vapor boil-up. One can observe that, with the increase of the vapor boil-up parameter, the structure becomes more complex: firstly, the singular arcs are to appear more and more frequently, ie. from a total number of 1 to 3, for both the desired purities; moreover, the unique zero arcs tends to last more, as it triples for an increase of only 25% and becomes even 5 times longer for an increase of 2.5 times in vapor boil-up; the last mentioned change, invokes also the introduction of the additional bang arc for all desired purities.
Figure 4: The influence of vapor boil-up variation on the control structure: 1) left: purity set at 85%, 2) right: purity set at 95%.

4. CONCLUSIONS

The optimal cyclic reflux policy used, solved the problem of optimizing a multi-cut pressure-swing batch distillation campaign using variable reflux ratios with the “recycling” of cuts at predetermined times, whereas, the gap between compositions in reboiler and recycled cut(s) is minimized. The additional analysis of process constraints, i.e. their possible influence on the duration of the process, is investigated also. Last, but not least, the author reported the details on productivity. Furthermore, future studies should focus on more complex mixtures originating from industrial waste, especially those treated after the effluents were trashed from beverage and food industry, containing complex compounds such as alcohols, esters, aldehydes (Esteban-Decloux et al. (2022)). Consequently, the primary goal should be direct implementation into the industrial environment after the pilot-plant checking for predesigned control platform structures (Lara-Montanoa et al. (2019)).

REFERENCES


