Optimal Heat Integration and Transfer Distribution in an Extractive Distillation System Using Columns with Internal Heat Exchangers

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We have optimized the heat integration and energetic load distribution in two extractive distillation systems for ethanol dehydration. The first system employs conventional (adiabatic) columns, whereas columns with internal sequential heat exchangers (SHE) are used in the second one. The optimal heat load distribution is found by minimizing the entropy production in the whole system fixing the product composition and number of stages in both processes. The results show that extractive columns are the main entropy generator (~80% of total), the dehydration system using SHE columns reduces the entropy production in 17% compared with the conventional one and the largest entropy saving is obtained in extractive column. The optimal operational condition for the system with SHE column implies an important reduction on heating utility.

1. Introduction

The conscience about the fossil fuels depletion as well as their impact on the environment has promoted the search for renewable energy sources, new processes and improving of energy efficiency. So, the use of ethanol as fuel either in blends with gasoline or pure has become attractive among the renewable fuels.

The ethanol dehydration via extractive distillation continues being competitive, compared with other technologies, such as molecular sieves, pervaporation or azeotropic distillation, because of its simplicity, flexibility, throughput and energy consumption (Knight and Doherty, 1989).

The increasing cost of the energy has promoted the design of distillation columns weighting up the reversible distillation concept. In a reversible distillation there is a differential composition change throughout the column and the heat transfer is carried out reversibly in each stage, this kind of column cannot be built in practice, but some features of the reversible distillation have been applied to design diabatic distillation columns with heat transfer in each stage (Kjelstrup et al., 2006).

The most efficient diabatic column requires heat transfer fluids (utilities) at different temperatures, close enough to the stage temperature. But this condition can be hardly achieved in a real process plant, a more realistic alternative are the columns with sequential heat exchanger (SHE) where the utilities go sequentially from one stage to another (see Figure 1), this configuration have the advantage of using only one heating and one cooling utility instead of one for each stage and requires to estimate a suitable heat transfer profile that minimize the entropy production due to heat transfers.

The aim of this work is assessing the potential reduction on the entropy production in ethanol dehydration via extractive distillation by using SHE and heat integration.

Figure 1. Extractive distillation system with SHE columns and heat integration.

2. The Distillation Systems

Two flowsheets are assessed. The first flowsheet (FS1) uses adiabatic columns, whereas in the second one (FS2) the adiabatic columns are replaced by SHE columns.

In FS2 (Figure 1), a near-azeotrope water/ethanol mixture, F_1, and the extractive agent (high purity ethylene glycol), F_U, are fed into the extractive column, C2, to obtain high purity ethanol as distillate, D2, and a blend of glycol, water and ethanol traces in the bottom stream, B2. The column C1 (recovering column) dehydrates the stream, B2, obtaining water as distillate, D1, and high purity ethylene glycol as bottom, B1. The stream B1 is mixed with the makeup stream, F, in the mixer MIX2.

The heat integration comprises two heat exchangers, HX1 and HX2, the condenser of the recovering column, and the reboilers of both columns. The heating utility H1 enters the reboiler of column C1, flows through the internal heat exchangers and leaves the column at the condition, H11. Fresh heating utility H, mixes with the stream H11 forming the stream H2 which flows through the reboiler and the internal heat exchangers of the column C2. The heating utility coming out from C2, H21, is used to cool down the recycle stream B1 up to the temperature T_{B11}. The recycle B11 is cooled down further up to the temperature T_{B12} using a cooling utility (water) coming from an external supply, W2.

The cooling utility coming out from the heat exchanger HX2, W21, flows into the condenser and the upper internal heat exchangers of the column C1. For the condenser and upper internal heat exchangers of column C2 fresh cooling utility is used.

3. Calculations

3.1 Flowsheet modeling

The flowsheets consider three main units, viz.: distillation columns, heat exchangers and mixers. The mathematical modeling of distillation columns is a modification of the boundary value method (Doherty and Malone, 2001), and allows taking into account the heat transfer at internal stages of the distillation columns. A detailed derivation of the model as well as its solving methodology can be found in a previous work (Mendoza

and Riascos, 2010). Entropy production in distillation columns is calculated from the entropy balance around the each column:

$$\left(\frac{ds_{irr}}{dt}\right)_{DC} = D\left[s_D - s_F - \frac{\dot{Q}_c}{DT_c}\left(1+\sum_{l=1}^{J}\frac{q_l}{\tau_l}\right)\right] + B\left[s_B - s_F - \frac{\dot{Q}_B}{DT_B}\left(1+\sum_{b=1}^{M}\frac{q_b}{\tau_b}\right)\right] + \left(\frac{ds_{irr}}{dt}\right)_{HXDC} \qquad (1)$$

where Q_c, T_c and Q_B, T_B are the heat loads and temperatures in condenser and reboiler respectively, and D and B are distillate and bottom products. The summations terms represent the heat load in the internal stages of the column $l =1$ to J are the stages of the rectifying section and the $b = 1$ to M are the stages of the stripping and extractive zones; τ_l and τ_b are stage dimensionless temperatures ($\tau_l=T_l/T_c$, $\tau_b=T_b/T_B$) and q_l, q_b are stage dimensionless heat loads ($q_l=Q_l/Q_c$, $q_b=Q_b/Q_B$); s_D, s_B, s_F are the molar entropies of distillate, bottom product and feed streams. The entropy of the feed is $s_F=(F_r s_{Fu}+s_{Fl})/(1+F_r)$ where s_{Fu} and s_{Fl} are molar entropies of the upper and lower feeds and F_r is the feed ratio: quotient between upper and lower feed, i.e. $F_r=F_u/F_l$. The last term of the right hand side is the entropy production due to heat exchange into distillation column between utilities and stages (HXDC), calculated as the product of the heat flow and its thermal force (Kjelstrup et al., 2006):

$$\left(\frac{ds_{irr}}{dt}\right)_{HXDC} = \sum_{l=1}^{J}\dot{Q}_l\left(\frac{1}{T_l}-\frac{1}{T_{f,l}}\right) + \sum_{b=1}^{M}\dot{Q}_l\left(\frac{1}{T_b}-\frac{1}{T_{f,b}}\right) \qquad (2)$$

where T_l, T_b are the temperature of the stage and $T_{f,l}$, $T_{f,b}$ are the average temperatures of the cooling and heating utilities in the stage.

Mass, energy and entropy balances in heat exchangers as well as in mixers do not take into account any internal detail of the apparatus, therefore the balance equations are based only in the material streams that enter and leave the equipment. Heat exchangers are counter-current because this configuration favors savings in entropy production (Kjelstrup et al., 2006). Entropy production in heat exchangers and adiabatic mixers is:

$$\left(\frac{ds_{irr}}{dt}\right)_{HX1} = B_1(s_{B11}-s_{B1})+H_2(s_{H22}-s_{H21}) \;;\quad \left(\frac{ds_{irr}}{dt}\right)_{HX2} = B_1(s_{B12}-s_{B11})+W_2(s_{W21}-s_{W2}) \qquad (3)$$

$$\left(\frac{ds_{irr}}{dt}\right)_{MIX1} = H_2s_{H2}-H_{12}s_{H12}-Hs_{H} \;;\quad \left(\frac{ds_{irr}}{dt}\right)_{MIX2} = F_Us_{FU}-Fs_F-B_{12}s_{B12} \qquad (4)$$

where s is the molar entropy and the index identifies the stream.

3.2. Case studies

The process is intended for dehydrating ethanol in the column C2, with a purity of 99.8 mol% in the distillate D2. The ethylene glycol obtained in the bottom of the recovering column, C1, is 99.9 mol%. The fractional recoveries of ethanol, in column C2, and of ethylene glycol, in column C1, are 99.999% in both cases.

The lower feed that enters the extractive column, F_l, is 0.278 mol s^{-1} with 85 mol% ethanol and 15 mol% water and it´s at bubble temperature. The upper feed, F_U, is 0.139 mol s^{-1} with 99.9 mol% ethylene glycol (~0.1 mol% water and traces of ethanol). The makeup stream, F, has a temperature of 298.1 K and a composition of 99.9 mol% ethanol and 0.1 mol% water.

Temperatures of streams H1 and H2 are 490 and 435 K respectively, whereas for streams, F, W1 and W2 is 298.15 K. The minimum temperature approach allowed in the

heat exchangers, reboilers and condensers is 10 K. The pressure of the columns, heat exchangers and mixers is, in all cases, 1 atm. The number of stages and the separation task developed in distillation columns are equal for the both systems.

3.3. Model solution

The calculation sequence employed in this work avoids any convergence problem for the recycle streams. The calculation order is as follows:
1. Flows and compositions of streams B2 and D1 are calculated from the global material balances in the columns C2 and C1. The temperatures of steams D1, D2, B1 and B2 are calculated as bubble point temperatures at the column's pressure.
2. Set the temperature of the stream F_U 5 to 10 K below the temperature of distillate D2 (Knight and Doherty, 1989) and solve the column C2.
3. Solve the material and energy balances from mixer MIX2 to calculate the flow of makeup stream, F, and the temperature of the stream B12 and set the temperature of the stream B11 between the temperatures of the streams B1 and B12.
4. Set the outlet temperature of cooling utility W2 and calculate its flow using the material balance in the heat exchanger.
5. Solve the recovering distillation column C1 and calculate the flow and temperature of the fresh heating utility H, solving the mass and energy balances in mixer MIX1.

3.4. Optimization

The objective function, *OF*, comprises two terms, the first one is the entropy production of the system given by the summation of the entropy generated by each process unit, Eqs. (1), (3) and (4); the second one is a penalty term which takes into account the fulfillment of the design feasibility of the distillation columns (β_1), the minimum temperature approach in heat exchangers, condensers and reboilers (β_2 to β_7). β has a binary nature (zero if the constraint is fulfilled and one if it is not).

$$OF = \left(\frac{ds_{irr}}{dt} \right)_{process} + \alpha \sum_{r=1}^{7} \beta_r \qquad (7)$$

where α is a weighting factor ($\alpha = 100$). Optimization variables and their limits were defined based on a preliminary analysis by modeling distillation columns and the economic optimization done by Knight and Doherty (1989) for ethanol dehydration using extractive adiabatic distillation. These variables and limits are: upper feed temperature ($339 \leq T_{Fu} \leq 351.5$ K), total heat transfer conductivity of SHE in each stage ($0 \leq UA_{stage} \leq 2$ kW K^{-1}), molar external reflux ratio of recovery column ($0 \leq RR \leq 0.3$) and extractive one ($0 \leq RR \leq 0.5$), outlet temperature of recycle in the heat exchanger HX1, ($395 \leq T_{B11} \leq 410$ K), flow of cooling utility in extractive column ($4.0 \leq W1 \leq 5.0$ mol s^{-1}) and recovery one ($0.5 \leq W2 \leq 1.5$ mol s^{-1}), and flow of heating utility in extractive column ($0.4 \leq H2 \leq 4.0$ mol s^{-1}).

3.5. Thermodynamic properties and computational methods

In liquid phases the activity coefficients as well as the enthalpies and entropies of mixing are calculated using the NRTL model (Renon, 1969), whereas vapor phases are considered as ideal solutions described by the ideal gas law. Liquid and vapor heat capacities and heats of vaporization were taken from Poling et al. (2008). The heat capacity of heating utility, DowthermT™, can be found in the Dow's web page (Dow, 2004). The model was programmed in Matlab™ using the functions *fsolve* and *patternsearch* to solve and optimize the model respectively. A derivative free method was chosen for optimization due to the presence of binary variables.
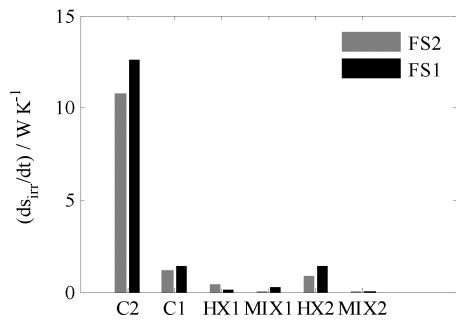
Figure 2. Entropy production distribution in extractive distillation systems.

4. Results and discussion

The outcome show using SHE columns overall entropy production is reduced about 17%. In both systems, the main entropy generators are extractive columns (Fig. 2), they account for 80% and 82% of total entropy production in the systems FS1 (15.84 W K^{-1}) and FS2 (13.21 W K^{-1}). So, the greatest reduction in the entropy production was also achieved in these column, from 12.6 (adiabatic) to 10.8 W K^{-1}(SHE column).

The entropy production profile in columns C1 and C2 (Fig. 3) shows the main reduction is located in the stripping zone and coincides with the heat load distribution of the SHE column. Differences in entropy production profile are generated by both the separation itself and the heat exchange with utility, including reboiler and condenser.
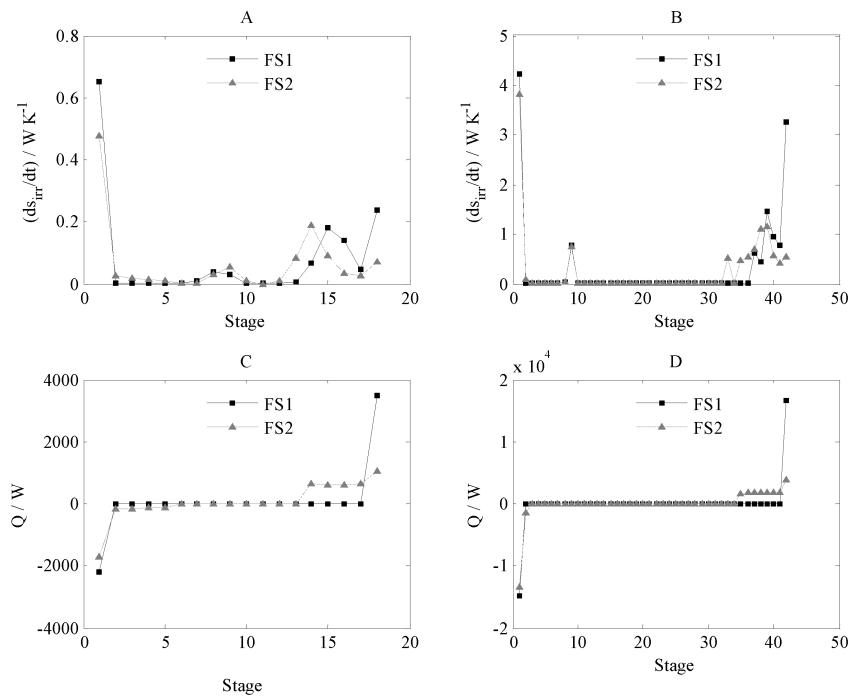

Figure 3. Entropy production and heat distribution. Left, recovery columns, Right, extractive ones.

Due to the internal heat exchange, entropy production in stages with exchangers is greater than in adiabatic columns, but it allows a larger scale reduction of entropy generation in the reboiler and condenser. The condensation-evaporation patterns obtained in the SHE columns produce greater variations in the internal flows compared to adiabatic ones, this fact requires special attention on hydraulic of the column, to avoid problems such as irregular pressure drop, flooding and/or drying-up (Kjelstrup et al., 2006). So trays are preferable than packing in SHE columns because the former have a better turndown ratio and used to be cheaper than packing (Lockett, 1986).

Savings in entropy production found in the current work represents a reduction in the loss work (calculated as the product of the ambient temperature, T_0=298.15 K, and the entropy production saving 2.64 W K^{-1}) of 787 W (68 MJ per day). In practice this saving is appreciated in the external heating requirements of the process (viz., the stream H changes from 3.54 mol s^{-1} and 426.1 K in FS1 to 0.36 mol s^{-1} and 430.3 K in FS2, likewise the flow in H1 reduces from 0.43 to 0.13 mol s-1), and ultimately in a lower fuel consumption, that means a reduction not only in the operational costs but also in the carbon dioxide emissions at the expense of incrementing capital costs by including sequential heat exchangers in distillation columns.

5. Conclusions and Future Works

Using distillation columns with internal sequential heat exchangers in extractive distillation systems has a significant effect on reduction of entropy production in the process (17 % less than process using adiabatic columns). Extractive column is the main entropy generator (~80 %), in order that efforts for improving energy efficiency must be directed to the design and operation of this column. The improvements in energy efficiency and the environmental advantages showed by the distillation columns with internal sequential heat exchangers are an alternative to be considered not only in the case currently presented but also in other distillation processes, and it is worthy of future studies.

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