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ABSTRACT

Distillation columns are generally operated very inefficiently from an energetic point of view. At the bottom, a boiler is required to vaporize the mixture while at the top heat is rejected to the environment. Vapor recompression heat pumps have been very successful in the realization of extremely large energy savings. For larger temperature lifts between top and bottom, the temperature glide in the absorber and desorber of compression-resorption heat pumps reduce the thermodynamic temperature lift and make these heat pumps a competitive solution.

In this paper the performance of a diabatic column with the resorber of an ammonia-water compression-resorption heat pump integrated in the stripping section of the column and the desorber in the rectification section will be compared with the performance of an adiabatic column. The conventional boiler / condenser option and a vapor recompression option will both be investigated. The impact of ammonia-water concentration on performance and operating conditions will also be discussed.

1. INTRODUCTION

Distillation is one of the main operations in the process industry. Distillation columns are intrinsically very inefficient and form the major energy consumers in the process industry. To separate the top and bottom products, large amounts of heat are added to the reboiler at the bottom of the column while similar amounts of heat are rejected to the environment in the condenser at the top of the column. The efficiency, defined as the ratio of the separation energy and the energy input to the reboiler, is generally very low. Integration of heat pumps with distillation columns can significantly improve their energetic performance. Vapour recompression heat pumps have been preferred, see Fonyo and Mizsey (1994), since the temperature driving force on the evaporator side vanishes: the distillate product is taken as the working fluid for the heat pump. In this way large COP (ratio between reboiler heat and shaft power consumption of the heat pump) values can be attained. Fonyo and Benko (1996) concluded that when the lift (temperature difference between reboiler and condenser of the distillation column) becomes too large the performance of vapor recompression heat pumps starts being less attractive. When a vapor recompression heat pump is implemented, it is installed between condenser and reboiler of the distillation column. In this way the column still operates as an adiabatic column. Rivero (2002) investigated the possibility of transferring heat at each column tray instead of externally. In this way the column becomes diabatic and the local entropy production is reduced since heat transfer takes place at reduced temperature driving forces. In compression-resorption heat pumps heat transfer takes place at gliding temperatures. Further these cycles can be designed to show temperature glides comparable to the temperature glides in the distillation column. In this way the average heat removal temperature increases while the heat addition temperature reduces. The result is that the actual temperature lift for this cycle reduces. In this way the COP can be expected to become competitive with the COP for the vapour recompression cycle.

The objective of this study is to verify if compression-resorption heat pumps are attractive alternatives to vapor recompression heat pumps in distillation columns with reasonable temperature lift. For this purpose a toluene-benzene distillation column with a temperature lift of ca. 30 K between condenser and reboiler has been selected. It is expected that distillation columns with larger lifts will be more attractive than the case considered here while the columns with smaller lifts will be less attractive. Models for column and heat pump are introduced so that the performance of the integrated system can be predicted. Finally results are presented and discussed.

2. MODEL

2.1 Distillation Column

In the past among others Schaller *et al.* (2001) and Rivero (2001,2002) have investigated diabatic distillation columns. The model presented here is based on these previous studies. Figure 1 shows schematically an adiabatic column with F the feed flow with (light component) concentration z_F, D the distillate product flow with (light component) concentration x_D and B the bottom product flow with (light component) concentration x_B. Mass conservation gives:

$$F = D + B \tag{1}$$

$$Fz_F = Dx_D + Bx_B \tag{2}$$

It is assumed that the condenser is a total condenser so that

$$V_1 = D + L_0 \tag{3}$$

with V_1 the vapor that leaves the top of the column and enters the condenser and L_0 the condensate flow that is sent back to the column. Since the condenser is a total condenser

$$y_1 = x_D = x_0 \tag{4}$$

Figure 1. – Adiabatic column

with y_1 the light component concentration of the vapor. L_0 is the external reflux that is given by the external reflux ratio, R:

$$R = L_0/D \tag{5}$$

The quantity of heat removed by the external condenser is

$$\dot{Q}_{condenser} = V_1(h_1^V - h_D^L) \tag{6}$$

At the bottom the following equations apply:

$$L_N = B \tag{7}$$

$$x_N = x_B \tag{8}$$
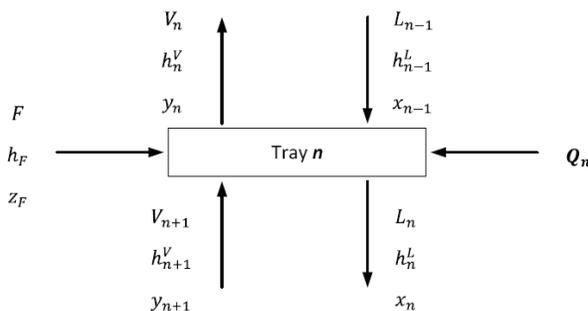
Figure 2. – Conservation balances for n[th] tray

where N stands for the bottom tray of the column.
In the further discussion only a two component mixture is considered, the pressure drop is neglected, the processes are considered to be in steady state, the liquid and vapor phases are considered to be in equilibrium in each tray. Further it is assumed that there is a single tray to which the feed is added, that both distillate and bottom products are liquids at their boiling points and that the column does not exchange heat with the surroundings. Mass and energy balances for each tray, schematically represented in Figure 2, give the following equations.

$$V_n + L_n = V_{n+1} + L_{n-1} + F \tag{9}$$

$$V_n y_n + L_n x_n = V_{n+1} y_{n+1} + L_{n-1} x_{n-1} + F z_F \tag{10}$$

$$V_n h_n^V + L_n h_n^L = V_{n+1} h_{n+1}^V + L_{n-1} h_{n-1}^L + F h_F + \dot{Q}_n \tag{11}$$

with $\dot{Q}_n$ the heat transferred in the nth tray with the component of the compression resorption cycle. As stated above, equilibrium has been assumed between liquid and vapor at each tray. The equilibrium equations have been provided by the software library FluidProp (Colonna and van der Stelt, 2004):

$$y_n = f(x_n) \tag{12}$$

Further from a conservation balance for the rectification section of the column, the part of the column above the feed tray, and the stripping section, the part of the column below the feed tray, the following equations apply:

$$V_{n+1} - L_n = D \text{ if } 1 <= n <= n_F - 2 \tag{13}$$

$$V_{n+1} - L_n = D - F \text{ if } n = n_F \, orn = N \tag{14}$$

$$V_{n+1} - L_n = D - (1-q)F \text{ if } n = n_F - 1 \tag{15}$$

$$V_{n+1} y_{n+1} - L_n x_n = D x_D \text{ if } 1 <= n <= n_F - 2 \tag{16}$$

$$V_{n+1} y_{n+1} - L_n x_n = D x_D - F z_F \text{ if } n = n_F \, orn = N \tag{17}$$

$$V_{n+1} y_{n+1} - L_n x_n = D x_D - (1-q)F z_F \text{ if } n = n_F - 1 \tag{18}$$
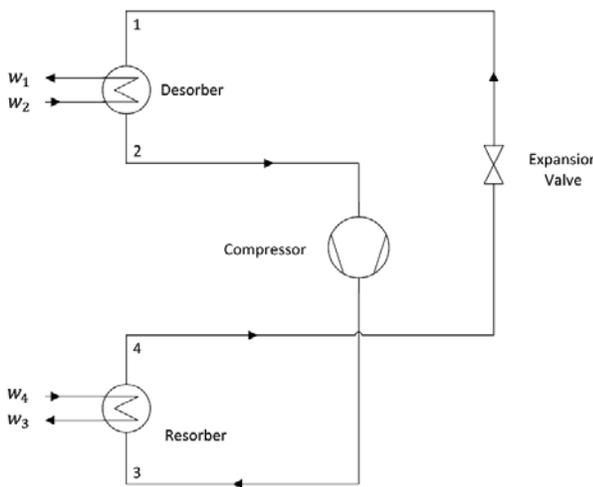

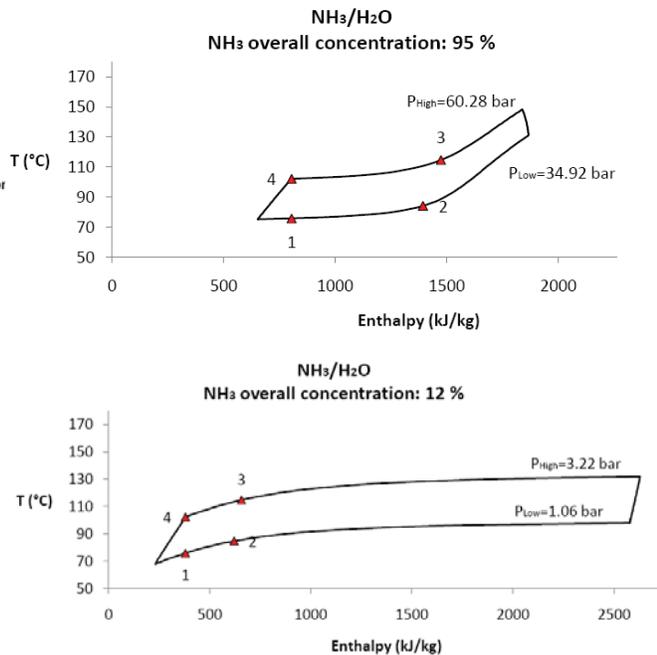Figure 3. – Schematic of compression resorption heat pump cycle

Figure 4. – Compression resorption heat pump cycle in ammonia-water diagram for an ammonia overall concentration of 95% (left) and 12% (right).

with n_F the tray in which the feed flow is added and q the liquid fraction of the feed flow. The energy balance gives:

$$\dot{Q}_n = V_n h_n^V + L_n h_n^L - V_{n+1} h_{n+1}^V - L_{n-1} h_{n-1}^L \text{ if } 1 <= n <= n_F - 2 \tag{19}$$

$$\dot{Q}_n = V_n h_n^V + L_n h_n^L - V_{n+1} h_{n+1}^V - L_{n-1} h_{n-1}^L - qF h_F^L \text{ if } n = n_F \tag{20}$$

$$\dot{Q}_n = V_n h_n^V + L_n h_n^L - V_{n+1} h_{n+1}^V - L_{n-1} h_{n-1}^L - (1-q)F h_F^V \text{ if } n = n_F - 1 \tag{21}$$

For the bottom tray (the resorber) the balance gives

$$\dot{Q}_N = \dot{Q}_{reboiler} = V_N h_N^V + B_n h_n^L - L_{N-1} h_{N-1}^L \tag{22}$$

2.2 Compression Resorption Heat Pump

Figure 3 shows a schematic of the compression resorption cycle. Compression resorption cycles are discussed among others in Itard and Machielsen (1994). Although different working fluids can be selected for application in this cycle, the working mixture considered here is ammonia-water. This mixture appears to be quite suitable for the applications under study. The cycle is quite comparable to the conventional vapor compression cycle. Difference is that the heat rejection takes place when ammonia vapor is absorbed in the ammonia-water solution. So instead of a condenser, a resorber is applied. In the low pressure side instead of evaporation at constant temperature, desorption takes place under gliding temperature. This is further illustrated in Figure 4 that illustrates the cycle in T,h diagrams.

Process 3 to 4 represents the resorption process and takes place at high pressure. In the present study the pressure drop during resorption and desorption is neglected so that heat transfer takes place at high and low constant pressures. Since desorption is incomplete, the compressor increases the pressure of both ammonia-water solution and ammonia vapor.

Figure 4 indicates that the operating pressures will significantly depend on the overall ammonia concentration in the ammonia-water solution. Both cycles show similar temperature glides. However the enthalpy changes at higher ammonia concentration are higher while the compression work remains comparable. From this can be seen that the COP of cycles with higher ammonia-water concentrations will be higher but at the same time the operating pressures will also be higher. The ammonia-water solution concentration is selected to fit the glide of the column to the glide of the heat pump. Heat pump cycle and distillation column are integrated as illustrated in Figure 5.

The mass flow of solution through the resorber results from the energy to be delivered to the column:

$$\dot{m}_{sol} = \frac{\dot{Q}_{reboiler}}{h_3 - h_4} \tag{23}$$

It is assumed that the temperature at the outlet of the resorber, T_4, is $\Delta T_{resorber}$ Kelvin higher than the
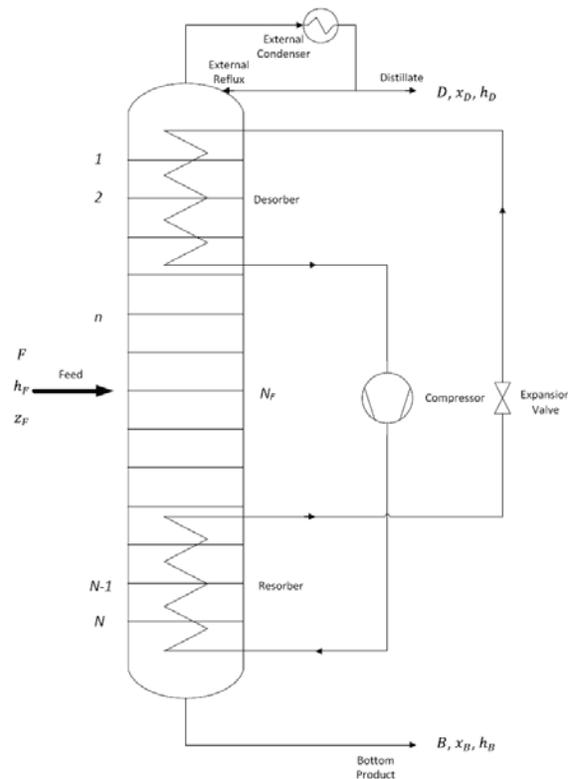

Figure 5. – Schematic of compression resorption heat pump cycle integrated with a distillation column

temperature in the respective column tray. Notice that an extra degree of freedom has to be considered in relation to the number of trays equipped with part of the resorber. For the results presented in this paper a value of 5 K has been assumed for $\Delta T_{resorber}$ while a variable number of trays with part of the resorber has been considered.

When an average ammonia water concentration is selected then, since saturated liquid is assumed at the outlet of the resorber, this implies that the operating pressure of the resorber and enthalpy h_4 are known. The same temperature difference is assumed at the resorber inlet:

$$T_3 = T_B + \Delta T_{resorber} \tag{24}$$

In this way enthalpy h_3 is also known. The amount of heat removed by the desorber follows from

$$\dot{Q}_{desorber} = \dot{m}_{sol}(h_2 - h_1) \tag{25}$$

Since a throttling process takes place between states 4 and 1,

$$h_1 = h_4 \tag{26}$$

The operating pressure for the desorber is obtained from h_1 and T_1 where T_1 is obtained from

$$T_1 = T_{tray1} - \Delta T_{desorber} \tag{27}$$

This $\Delta T_{desorber}$ has been taken so that for all parts of the desorber the temperature difference between tray process fluid and desorber temperature is not lower then a fixed value. For the results presented here 5 K has been assumed. Notice that this requires an iterative procedure.
An isentropic efficiency is assumed for the (wet) compressor so that

$$h_2 = \frac{h_{3s} - \eta_{is} h_3}{1 - \eta_{is}} \tag{28}$$

with η_{is} the isentropic efficiency of the compressor. It has taken equal to 0.70 in the further discussion.

3. RESULTS AND DISCUSSION

First of all the ammonia-water average concentration has been varied. In this way it has been identified that the best performance (expressed as the coefficient of performance, COP) was obtained for low (12%) or large concentrations (95%). Low ammonia concentrations will lead to low pressures (around atmospheric pressure) while high ammonia concentrations will lead to high pressure, up to 70 bar. When implementing the resorber in the distillation column, making it diabatic, the heat addition takes place at a lower temperature than in the case heat is added in a reboiler. To guarantee the same purity of the products, the total heat that has to be added is slightly larger than for the adiabatic case. This means that $\dot{Q}_{reboiler}$ in Eq. (23) can not be found from the adiabatic column results and that an iterative method is needed to identify $\dot{Q}_{reboiler}$ for diabatic operation. The second parameter to be considered is the number of trays in which the resorber is installed. This value has been varied between 4 and 8.

The original adiabatic column has 12 trays with 7 trays in the stripping section. The feed flow is 2.8 kg/s liquid at its boiling point 94.1°C (40% benzene and 60% toluene), the distillate flow is 1.1 kg/s liquid at its boiling point 80.8°C (97% benzene and 3% toluene) and the bottom flow is 1.7 kg/s liquid at its boiling point 109.6°C (2% benzene and 98% toluene). The temperature difference between bottom and top is 28.8 K. The required reboiler heat is 1.84 MW while 1.81 MW are removed at the condenser. For reference purposes it is assumed that the reboiler is gas fired with

an efficiency of 85% as proposed by Araujo et al. (2007). The energy consumption of the reference adiabatic column is thus 2.16 MW.

3.1 Mechanical Vapor Recompression Heat Pump

As previously reported by Fonyo and Mizley (1994), vapor recompression heat pumps form the state of the art technology for applications in combination with distillation columns. A schematic is given in Figure 6. In this case the distillate is the working fluid of the heat pump cycle. Under the assumption of 5 K temperature difference between bottom product and condensation temperature, the COP of the cycle is 6.7 so that the power required by the compressor shaft is 0.28 MW.

3.2 Compression Resorption Heat Pump

First the impact of ammonia average concentration has been investigated. Table 1 shows some model results from which it can be concluded that largest COP's are obtained with larger ammonia concentrations.

Table 1. Impact of the average ammonia concentration in the ammonia water solution.

Ammonia water concentration [%]	COP	p [bar] resorber	p [bar] desorber
12	4.03	3.96	1.00
13	3.95	4.23	1.07
34	3.06	13.7	4.05
90	5.43	64.2	32.2

The results in Table 1 apply for a column with 13 trays with the top 4 trays of the rectification section equipped with the desorber and the bottom 5 trays of stripping section equipped with the resorber. The largest COP values are obtained at larger ammonia concentrations. This could be expected from Figure 4 that shows that the technical work required by the compressor ($h_3 - h_2$) becomes much smaller than the enthalpy change in the resorber ($h_3 - h_4$) when the ammonia concentration increases. From Table 1 is also clear that the pressure ratio that has to be overcome by the compressor is the smallest for the large ammonia concentrations.
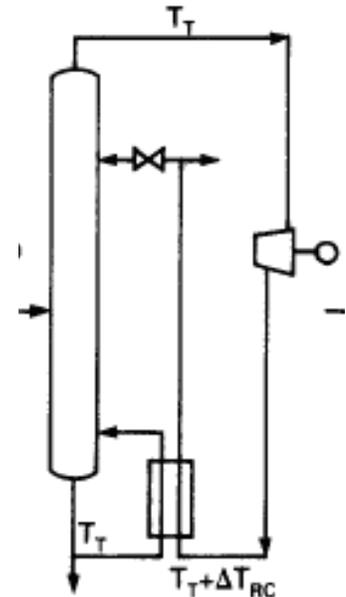

Figure 6. – Scheme for vapor recompression heat pump assisted distillation column, (Fonyo and Mizley, 1994).
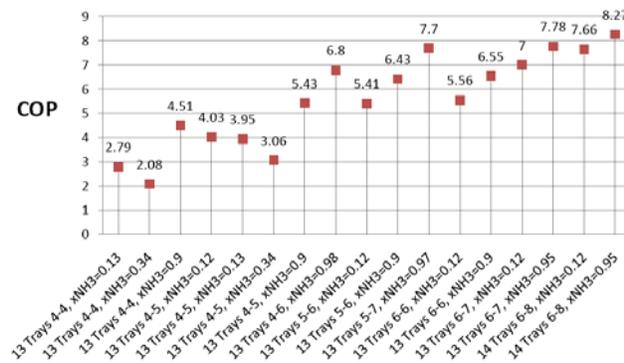

Figure 7. – COP of the compression resorption heat pump coupled with a diabatic distillation column
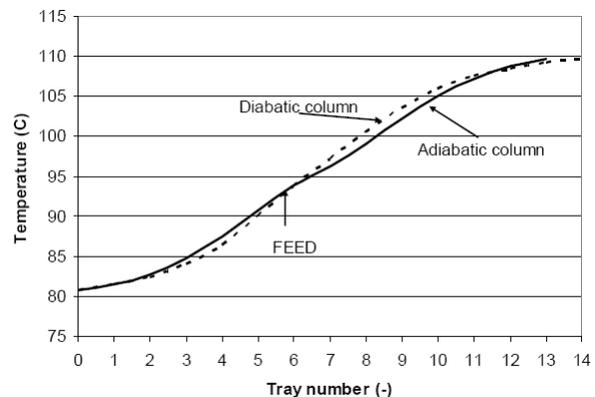

Figure 8. – Temperature profiles of adiabatic and diabatic distillation column

The impact of the number of trays equipped with the resorber as also been investigated. Figure 7 gives an overview of the situations that have been modeled. The different indexes listed in the figure indicate the number of trays of the column, the number of trays of the rectification section equipped with part of the desorber, the number of trays of the stripping section equipped with part of the resorber and the average ammonia concentration in the cycle. The two most right cases have 8 stripping section trays equipped with the resorber. In this case an extra tray is needed to maintain the purity of the products. The number of rectification trays with part

of the desorber follows from the temperature profile of the column and the specific heat pump cycle conditions. The best performance is found for the most right case and has a COP of 8.27. Here the operating pressure of the resorber is 60 bar while it is 35 bar for the desorber.

Figure 8 shows the impact of heat removal (rectification section) and addition (stripping section) on the temperature profile of the distillation column. In general, during diabatic distillation the temperatures in the rectification section are lower while the temperatures in the stripping section are higher than in the adiabatic case. The diabatic case given in Figure 8 concerns the profile obtained with 5 rectification section trays in which the desorber is installed (1 to 5) and 8 stripping section trays in which the resorber is installed (7 to 14). In total 1.8 MW are removed from the rectification section while 2.4 MW are added in the stripping section. In this case, an external condenser removes 0.6 MW.

3.3 Comparison with Alternatives

As indicated above, the conventional gas fired reboiler consumes 2.16 MW while the mechanical vapor recompression heat pump consumes 0.28 MW electrical energy. The compression resorption heat pump case with best performance requires a 20% larger heating power: 2.44 MW. The COP is larger than for the vapor recompression cycle but the heat requirement is here larger so that the net consumption of electrical energy is 0.29 MW. This value is practically the same as for the vapor recompression heat pump case.
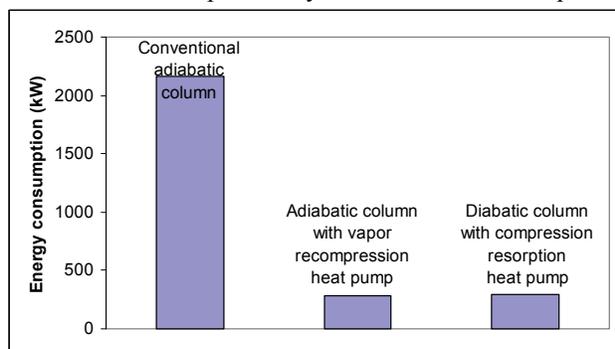
Figure 9. – Comparison between the energy consumption of a conventional distillation column and distillation columns with integrated heat pump
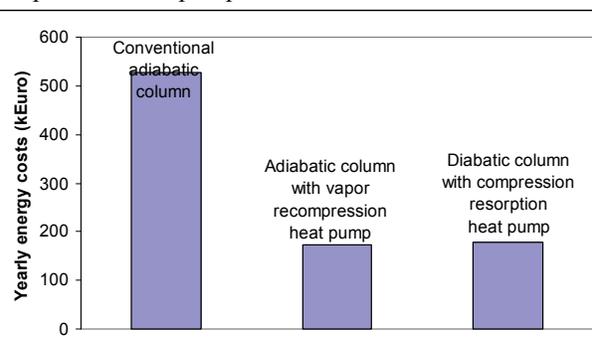
Figure 10. – Comparison between the yearly energy costs of a conventional distillation column and distillation columns with integrated heat pump

Figure 9 shows a comparison between a conventional adiabatic column and columns with integrated heat pumps. In the center the adiabatic column with external vapor recompression heat pump and right the diabatic column with compression-resorption heat pump integrated in the column trays.

The average energy costs for the European Union during the first semester of 2007 were: 0.0706 Euro/kWh for large industrial electricity consumers and 7.75 Euro/GJ for large industrial gas consumers (0.0279 Euro/kWh). Taking these prices into account and considering continuous operation, the yearly energy costs for the three alternative solutions are compared in Figure 10. From Figure 10 it appears that yearly 350000 Euros are saved when one of the two heat pump technologies is implemented. For the specific column considered here (benzene-toluene separation with ca. 30K temperature difference between top and bottom product) the compression-resorption heat pump shows no advantage when compared with the vapor recompression heat pump. The pressure ratio to be overcome by the compressor of the vapor recompression cycle is reasonably small (less than 3.0) making vapor recompression an attractive solution.

It can be expected that compression-resorption heat pumps become competitive alternatives only when the temperature difference between bottom and top products is larger than 30 K.

4. CONCLUSIONS

Compression-resorption heat pump cycles can be designed to have temperature glides in desorber and resorber that correspond to the temperature glides of the rectification and stripping section of distillation columns. This reduces the effective temperature lift that has to be bridged by the heat pump and allows for higher COP's than conventional vapor compression heat pumps would have.

The downside of these heat pumps is that since the heat is added at lower temperatures, larger amounts of heat are required to separate the column fluids. In this way the net energy consumption, for the case with 30 K between bottom and top products studied here, is similar to the energy consumption required for a vapor recompression heat pump with a significantly lower COP.

NOMENCLATURE

B   bottom product, kg/s
D   distillate, kg/s
F   feed, kg/s
h   enthalpy, kJ/kg
L   liquid mass flow, kg/s
$\dot{m}$ mass flux, kg/s
p   pressure, bar
$\dot{Q}$ heat flux, kW
R   external reflux ratio
V   vapor mass flow, kg/s
x   liquid weight concentration of light component, kg/kg
y   vapor weight concentration of light component, kg/kg

z   feed weight concentration of light component, kg/kg
η_{is} isentropic efficiency of compressor

Subscripts

0   reflux
1   tray one
B   bottom product
D   distillate product
F   feed
n n^{th} tray
N   bottom tray
sol   solution

Superscripts

L   liquid phase
V   vapor phase

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