

## The Influence of Heat Transfer Irreversibilities on the Optimal Performance of Diabatic Distillation Columns

M. Schaller<sup>1</sup>, K.H. Hoffmann<sup>1</sup>, R. Rivero<sup>2</sup>, B. Andresen<sup>3,4</sup>, P. Salamon<sup>4</sup>

<sup>1</sup>Department of Physics, Technical University of Chemnitz, Chemnitz, Germany

<sup>2</sup>Mexican Institute of Petroleum, Mexico City D.F., Mexico

<sup>3</sup>Ørsted Laboratory, University of Copenhagen, Copenhagen, Denmark

<sup>4</sup>Department of Mathematical Sciences, San Diego State University, San Diego, USA

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### Abstract

A distillation column with the possibility of heat exchange on every tray (a fully diabatic column) is optimized in the sense of minimizing its total entropy production. This entropy production counts the interior losses due to heat and mass flow as well as the entropy generated in the heat exchangers. It is observed that the optimal heating distribution, i.e. the heat exchange required on each tray, is essentially the same for all trays in the stripping and rectification sections, respectively. This makes a column design with consecutive interior heat exchanger and only one exterior supply for each of the two sections very appealing. The result is only slightly dependent on the heat transfer law considered. In the limit of an infinite number of trays even this column with resistance to transfer of heat becomes reversible.

### 1. Introduction

A number of recent works have examined optimal heating strategies for partially and fully diabatic distillation columns [1–9]. All of these works have focused on the entropy production *inside* the column while relegating to the environment the entropy production associated with getting the requisite heat exchanges to happen at desired rates. The present paper considers the question of how this heat exchange, when included in the entropy minimization, affects the optimal heating profile for fully diabatic columns. In the absence of counting irreversibilities due to heat exchange between the column and its surroundings, the optimal profile is independent of rate of operation in the sense that the optimal heating profile scales directly with the feed rate. This is no longer the case once the irreversibilities of the heat coupling to the outside of the column are included. For slow rate of operation the solution is essentially

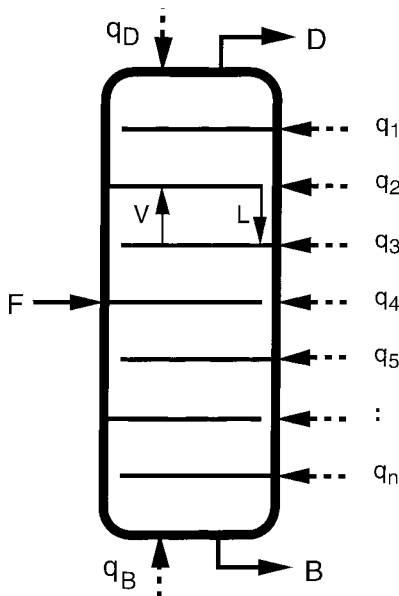
unchanged from the solution for the problem without irreversibilities due to the heat transfer. For fast operation, the entropy production inside the column becomes negligible compared to the irreversibilities due to the heat exchange which then dominates the behavior of the optimum.

## 2. Irreversible Heat Exchange

Consider an  $N$  tray completely diabatic distillation column (Fig. 1) separating an ideal binary mixture. The trays are numbered from  $n = 1$  at the top to  $n = N$  being the reboiler. The entropy production *inside* the column per unit time due to heat and mass flow can be written as

$$\Delta S^{\text{u, sep}} = \sum_{n=1}^N \frac{Q_n}{T_n} + \Delta S^{\text{streams}}, \quad (1)$$

where  $Q_n$  is the heat added to tray  $n$  of the column at temperature  $T_n$  and  $\Delta S^{\text{streams}}$  is the overall entropy flow associated with the three external mass streams, i.e. the feed  $F$ , the bottoms  $B$ , and the distillate  $D$ . Note that this expression only reflects the entropy production associated with the separation itself. It does not include irreversibilities due to external couplings like heat transfer. Previous studies have minimized  $\Delta S^{\text{u, sep}}$  [1–5] using the temperature profile  $T_n, n = 1, \dots, N$  as the control variables.



**Fig. 1.** Diabatic distillation column with possible heat exchange on all trays.

We assume that each tray is in thermodynamic equilibrium and thus the mole fractions  $x$  and  $y$  of the light component, 1 in the liquid and vapor phases, respectively, are related to the temperature by the Eqs. [10]

$$y = x \exp \left[ \frac{\Delta H^{\text{vap},1}(T)}{R} \left( \frac{1}{T_{b,1}} - \frac{1}{T} \right) \right] \quad (2)$$

$$1 - y = (1 - x) \exp \left[ \frac{\Delta H^{\text{vap},2}(T)}{R} \left( \frac{1}{T_{b,2}} - \frac{1}{T} \right) \right], \quad (3)$$

where  $T_{b,1}$  and  $T_{b,2}$  denote the boiling points of the two pure components. The enthalpies of vaporization  $\Delta H^{\text{vap},1}(T)$  and  $\Delta H^{\text{vap},2}(T)$  are calculated as

$$\Delta H^{\text{vap},i}(T) = \Delta H_b^{\text{vap},i} + (T - T_{b,i})(c_p^{\text{vap},i} - c_p^{\text{liq},i}), \quad (i = 1, 2), \quad (4)$$

where  $\Delta H_b^{\text{vap},i}$  are the heats of vaporization of the pure components at their respective boiling points, and  $c_p^{\text{liq},i}$  and  $c_p^{\text{vap},i}$  are their heat capacities in the liquid and vapor phases. Equation (4) requires the heat capacities to be temperature independent.

The solution of the column proceeds from the control variables  $T_n$  to the heat requirements  $Q_n$  by first calculating the concentration profiles  $x_n$  and  $y_n$  using Eqs. (2) and (3). We next proceed to use the mass balance conditions to find the flow rates  $L_n$  and  $V_n$  of liquid and vapor originating on tray  $n$ . Finally, we use the energy balance condition to find the  $Q_n$ . For the mass balance conditions it is easiest to start with the overall mass balance.

In steady state operation the feed, distillate, and bottoms obey the balance equations

$$F = D + B \quad (5)$$

$$x_F F = x_D D + x_B B, \quad (6)$$

where  $x_F$ ,  $x_B$ , and  $x_D$  denote the corresponding mole fractions of the more volatile component in the liquid phase. Usually the purities of the feed, distillate, and bottoms are specified so that a given feed rate  $F$  determines the rate of distillate and bottoms production through

$$D = \frac{x_F - x_B}{x_D - x_B} F \stackrel{\text{def}}{=} d F \quad (7)$$

$$B = \frac{x_F - x_D}{x_B - x_D} F \stackrel{\text{def}}{=} b F. \quad (8)$$

The last expression in each equation defines the constants  $d$  and  $b$ .

The amount of material flowing out of a tray must be equal to the amount of material flowing into a tray. Hence, vapor rate  $V_{n+1}$  coming up from tray  $n + 1$  and liquid rate

$L_n$  flowing down from tray  $n$  have to balance the distillate  $D$  above the feed and the bottoms  $B$  below the feed, respectively, both as total amount and componentwise,

$$V_{n+1} - L_n = \begin{cases} D & \text{above feed} \\ -B & \text{below feed} \end{cases} \quad (9)$$

$$y_{n+1}V_{n+1} - x_nL_n = \begin{cases} x_D D & \text{above feed} \\ -x_B B & \text{below feed.} \end{cases} \quad (10)$$

On the uppermost tray ( $n = 1$ ) the balance equations become  $V_1 = D + L_0$  and  $y_1 = x_D$ ; for the lowest tray ( $n = N$ , the reboiler) one obtains  $L_N = B$  and  $x_N = x_B$ . While for optimal diabatic columns the reflux  $L_0$  is zero, its purpose in an adiabatic column is to carry heat out of the column. This function is not needed in the diabatic column since heat can be taken directly from any tray.

The feed tray  $n$  is taken to be the tray on which the liquid light mole fraction  $x_n$  is equal to or immediately below the feed concentration  $x_F$  i.e.  $x_{n-1} > x_F \geq x_n$ .

The heat needed on each tray to give enthalpy balance is obtained from

$$Q_n = V_n H_n^{\text{vap}} + L_n H_n^{\text{liq}} - V_{n+1} H_{n+1}^{\text{vap}} - L_{n-1} H_{n-1}^{\text{liq}}. \quad (11)$$

The enthalpies  $H^{\text{vap}}$  and  $H^{\text{liq}}$  carried by the vapor and liquid flows are determined by

$$H^{\text{liq}}(T) = x c_p^{\text{liq},1}(T - T_{\text{ref}}) + (1 - x) c_p^{\text{liq},2}(T - T_{\text{ref}}) \quad (12)$$

$$H^{\text{vap}}(T) = y \left[ c_p^{\text{liq},1}(T - T_{\text{ref}}) + \Delta H^{\text{vap},1}(T) \right] \\ + (1 - y) \left[ c_p^{\text{liq},2}(T - T_{\text{ref}}) + \Delta H^{\text{vap},2}(T) \right] \quad (13)$$

with the zeropoint given through a reference temperature  $T_{\text{ref}}$ . Here we assume constant heat capacities, a noninteracting mixture of ideal gases for the vapor phase, and an ideal solution for the liquid phase [11].

Entropy carried to the surrounding through mass flow amounts to

$$\Delta S^{\text{streams}} = -F s_F + D s_D + B s_B, \quad (14)$$

where the entropies per mole of the mass flows are given by

$$s_i = x_i \left( s_{\text{ref},1} + c_p^{\text{liq},1} \ln \frac{T_i}{T_{\text{ref}}} \right) + (1 - x_i) \left( s_{\text{ref},2} + c_p^{\text{liq},2} \ln \frac{T_i}{T_{\text{ref}}} \right) \\ + R [x_i \ln x_i + (1 - x_i) \ln(1 - x_i)], \quad (i = F, D, B), \quad (15)$$

again with proper zeropoints given through  $s_{\text{ref},1}$  and  $s_{\text{ref},2}$ . Note that  $\Delta S^{\text{streams}}$  is fixed by the specifications of the process and is therefore not part of the optimization.

The summary above shows why the temperature profile is a particularly convenient set of control parameters since, given the  $T_n$ 's, straightforward calculations allow the evaluation of all the other column quantities. We now extend this calculation to

include the entropy production due not just to the separation as in Eq. (1) but also to the heat exchange. Note that all the flows in the column are proportional to the feed rate  $F$ . This makes it natural that current studies [1–5] all optimize the entropy production per unit feed rate. Letting

$$q_n = Q_n/F \quad (16)$$

we find using the definitions of  $d$  and  $b$  from Eqs. (7) and (8),

$$\Delta S^{\text{u, sep}} = \sum_{n=1}^N \frac{Q_n}{T_n} + \Delta S^{\text{streams}} = F \left( \sum_{n=1}^N \frac{q_n}{T_n} - s_F + ds_D + bs_B \right). \quad (17)$$

### 3. Entropy Production Due to Heat Exchange

To explore the effects of non-vanishing rate heat exchange which must proceed across a finite temperature difference and therefore produce entropy, we introduce two simple models for the heat transfer into the trays, the Newton and Fourier laws. For convenience we take the heat transfer coefficients to be the same for all trays in both models.

The entropy production due to heat exchange to the  $n$ -th tray is

$$\Delta S_n^{\text{u, hx}} = Q_n \left( \frac{1}{T_n} - \frac{1}{T_n^{\text{ex}}} \right). \quad (18)$$

For a given amount of heat transferred  $Q$ , the required external temperature  $T^{\text{ex}}$  depends on our assumed heat transfer law. This law relates the heat transferred to the tray to the temperature inside the tray and to the temperature  $T^{\text{ex}}$  in the heat exchange fluid outside the column. With our choice of the internal temperature profile as the control parameter, it is convenient to eliminate this dependence on the external temperatures by solving for the external temperature in terms of the heat transferred and the internal temperature and to use the resulting expression to eliminate  $T^{\text{ex}}$  in Eq. (18). This is carried out below for both of our heat transfer laws.

It is noteworthy that with this procedure the entire optimization, including the losses in the heat exchangers, is still parameterized solely by the internal temperature. The external temperatures do not add a new degree of freedom but are consequences of the internal profile.

#### 3.1. Fourier Heat Conduction

The first model is Fourier's law of heat transfer. Here heat transfer is taken to be proportional to the difference of the inverse temperatures, i.e. to the thermodynamic force,

$$Q_n = \tilde{\kappa} \left( \frac{1}{T_n} - \frac{1}{T_n^{\text{ex}}} \right), \quad \tilde{\kappa} > 0. \quad (19)$$

where  $\tilde{\kappa}$  is the conductance of the contact between tray  $n$  at temperature  $T_n$  and a bath at temperature  $T_n^{\text{ex}}$ .

Solving for  $1/T_n^{\text{ex}}$  in Eq. (19) we find

$$\frac{1}{T_n^{\text{ex}}} = \frac{1}{T_n} - \frac{Q_n}{\tilde{\kappa}}, \quad (20)$$

which is substituted into Eq. (18) to yield the very simple expression, independent of temperatures,

$$\Delta S_n^{\text{u,hx}} = \frac{Q_n^2}{\tilde{\kappa}}. \quad (21)$$

Expressing this in terms of the heats per mole of feed  $q_n$  so as to make the feed rate dependence explicit, we get

$$\Delta S_n^{\text{u,hx}} = \frac{F^2}{\tilde{\kappa}} q_n^2 \quad (22)$$

$$= F \tilde{g} q_n^2, \quad (23)$$

where we have introduced the quantity  $\tilde{g} = F/\tilde{\kappa}$  to describe the relative rate of mass to heat flow.

Summing this over our  $N$  trays, the total entropy production can be expressed as

$$\Delta S^{\text{u}} = \Delta S^{\text{u,sep}} + \Delta S^{\text{u,hx}} \quad (24)$$

$$= F \left( \sum_{n=1}^N \frac{q_n}{T_n} - s_F + ds_D + bs_B + \tilde{g} \sum_{n=1}^N q_n^2 \right). \quad (25)$$

Here it is evident that only the internal temperatures  $T_n$  are needed to parameterize the full operation of the column. Further note that since the total amount of heat exchanged,  $\sum_{n=1}^N q_n$ , is limited, the last sum in Eq. (25) vanishes for an infinite number of trays. This means that even *with* heat exchanger losses the separation becomes reversible in this limit.

### 3.2. Newtonian Heat Conduction

In the second more conventional model, Newtonian heat conduction, the heat transfer is taken to be proportional to the difference of the temperatures,

$$Q_n = \kappa(T_n^{\text{ex}} - T_n), \quad \kappa > 0. \quad (26)$$

Proceeding in the same fashion as in the previous subsection by eliminating  $T_n^{\text{ex}}$  in Eq. (18) we find

$$\frac{1}{T_n^{\text{ex}}} = \frac{1}{T_n + \frac{Q_n}{\kappa}} \quad (27)$$

and thus

$$\Delta S_n^{\text{u,hx}} = \frac{Q_n^2}{\kappa} \frac{1}{T_n \left( T_n + \frac{Q_n}{\kappa} \right)} \quad (28)$$

$$= Fg \frac{q_n^2}{T_n(T_n + gq_n)} \quad (29)$$

with  $g = F/\kappa$ .

This results in a total entropy production of

$$\Delta S^{\text{u}} = \Delta S^{\text{u,sep}} + \Delta S^{\text{u,hx}} \quad (30)$$

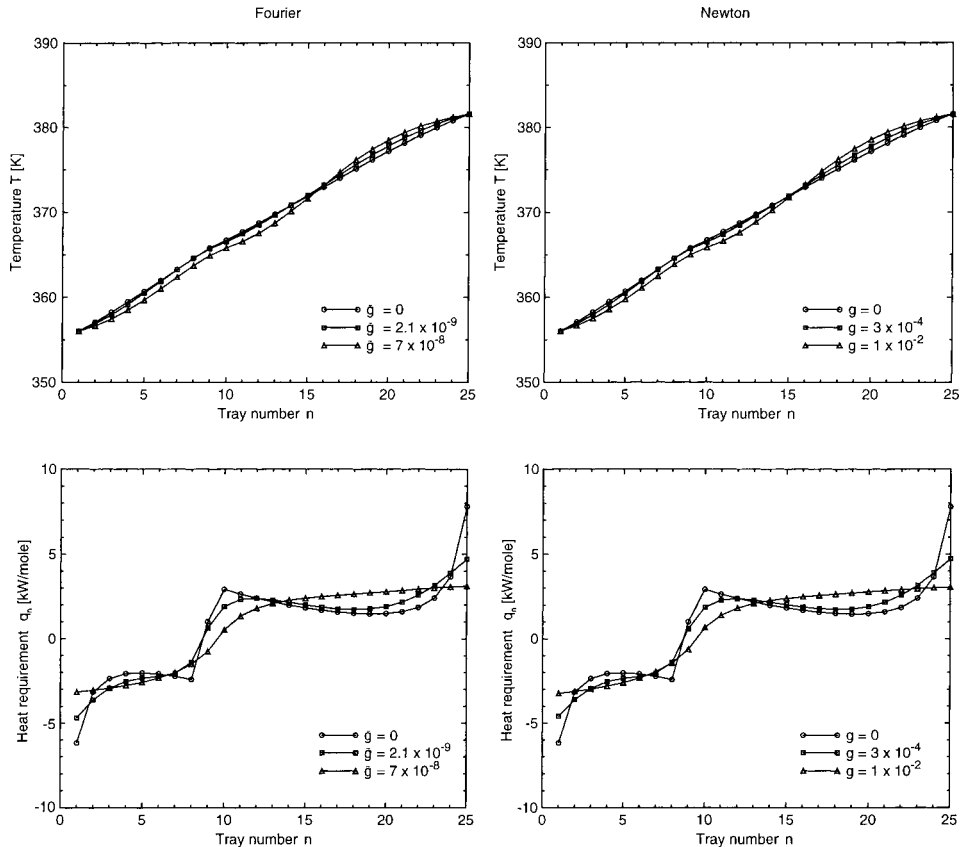
$$= F \left( \sum_{n=1}^N \frac{q_n}{T_n} - s_F + ds_D + bs_B + g \sum_{n=1}^N \frac{q_n^2}{T_n(T_n + gq_n)} \right). \quad (31)$$

Although a bit more complicated than Eq. (25), we see that the total entropy production is still a function of the internal temperatures  $T_n$  only. Note also that for  $q_n < 0$  (heat being withdrawn from the column), the value of  $g$  is limited to be in the range between 0 and  $T_n/q_n$ . The limit  $g = 0$  corresponds to vanishing feed rate or infinitely fast heat exchange, while the limit  $g = T_n/q_n$  corresponds to the highest possible rate of heat removal for which  $T_n^{\text{ex}} = 0$ .

#### 4. Results

As model systems, three columns of different length (25, 45, and 65 trays) are chosen to separate an ideal 50/50 benzene/toluene mixture. The required purities are 95% for the distillate and 5% for the bottoms, respectively. From a practitioner's point of view these columns are excessively long for the purity prescribed, but control requires extra freedom, and our results are particularly evident for long columns. Powell's routine [12] is used to minimize the total entropy productions (31) and (25).

Figure 2 shows the optimal temperature profiles and corresponding heating requirements for a 25-tray column with Fourier heat conduction (left frames) and Newtonian heat conduction (right frames). The three curves in each frame are calculated for  $g = 0$  and for two increasingly severe transfer resistances. The value of zero represents perfect heat conduction and is therefore identical to the previous studies of optimal distillation considering only internal losses [1–5]. The intermediate value corresponds to realistic heat conductance in a commercial heat exchanger, while the largest value of  $g$  is included to show the strongly resistive regime. The values of  $g$  and  $\tilde{g}$  are chosen to correspond to roughly the same temperature differences across the conductances and thus differ by a factor of  $1.4 \times 10^5 \text{ K}^2$  due to the forms of the two transfer Eqs. (26) and (19). The required temperature profiles are almost linear regardless of heat resistance. It is also clear that the form of the transfer law has very little effect on the optimal temperature sequence and hence the heating demand.

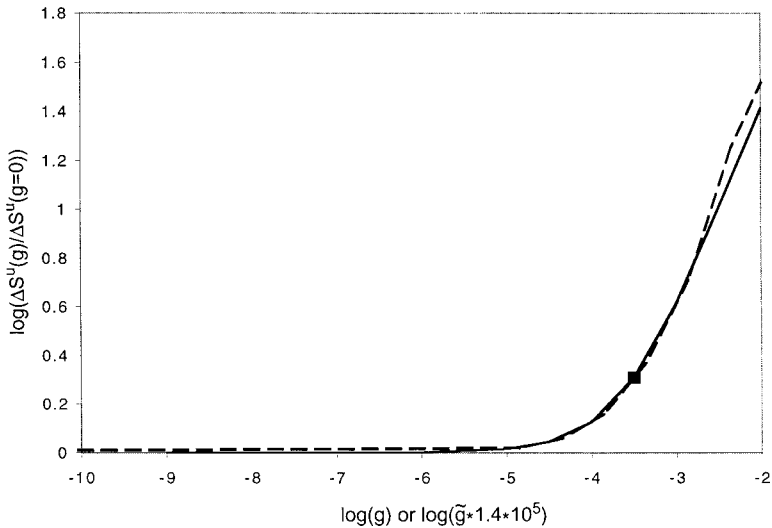


**Fig. 2.** Optimal temperature (upper frames) and heat duty profiles (lower frames) for a 25-tray column separating a 50/50 mixture of benzene-toluene. The left frames are for a Fourier heat law connecting the heat exchangers to the supply fluids, the right frames for a Newtonian conduction law. The relative flow parameters  $g$  and  $\tilde{g}$  are as indicated on the figure. The value zero corresponds to no entropy generation in heat exchange, i.e. reversible. The middle value is industrially realistic, while the lower value is strongly resistive.

This speaks in favor of using the simpler Fourier expression (25) for the entropy production.

A further comparison between the two heat transfer laws is provided by Figure 3, a log-log plot of the total entropy production relative to reversible heat transfer for a variation of  $g$  and  $\tilde{g}$  over 8 orders of magnitude. The realistic value of  $g = 3 \times 10^{-4}$  mole K/J also used in Figure 2 and the corresponding  $\tilde{g}$  is marked on the curves. The two curves are remarkably similar. We see that heat resistance is insignificant for  $g < 10^{-5}$  and dominant for  $g > 10^{-3}$  with normal operation right in the middle of the transition interval. The type of heat transfer (Newtonian or Fourier) is immaterial.

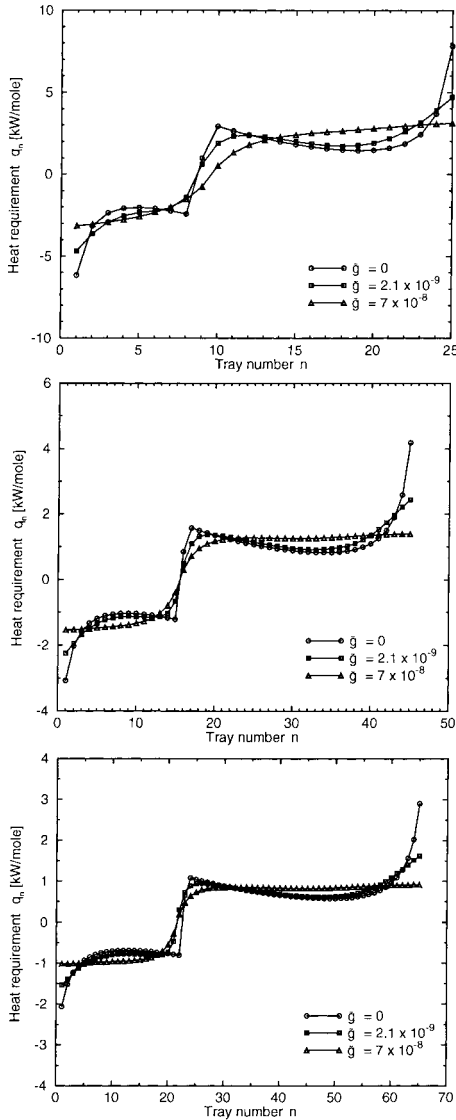




**Fig. 3.** Log-log plot of the total entropy production relative to reversible heat transfer for a variation of  $g$  and  $\tilde{g}$  over 8 orders of magnitude. Solid line: Newton's law of heat exchange; dashed line: Fourier's law of heat exchange. The special point marked corresponds to industrially realistic values of heat conductance.

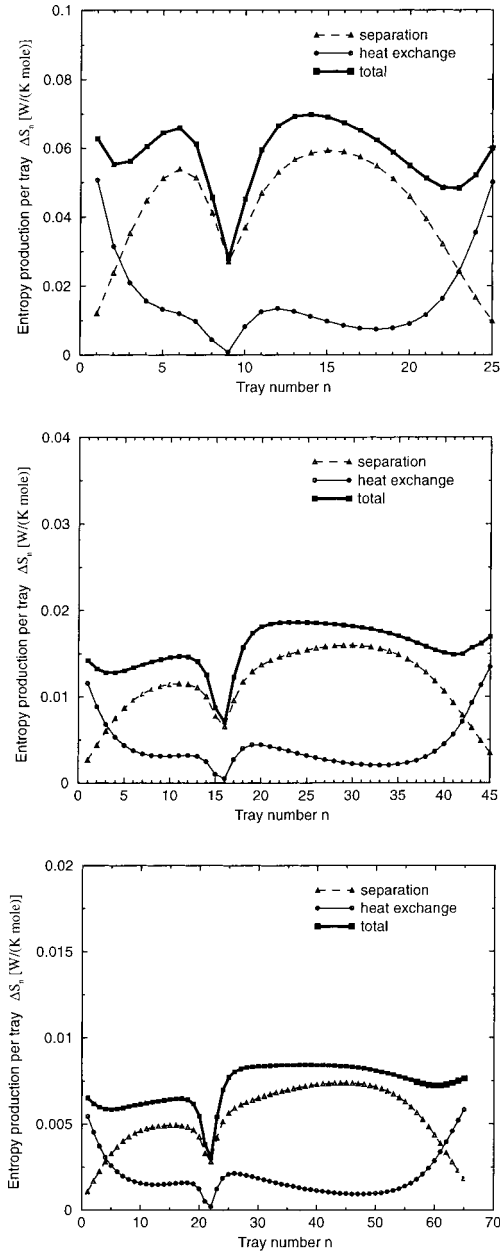
Figure 4 shows the optimal heating demands for columns of length 25, 45, and 65 trays using Fourier conduction. As indicated in Figure 2, the corresponding Newtonian results are indistinguishable and are therefore not shown here. The shorter column displays the characteristic 'inverted-U-U' shape of the heating curves observed in past studies for near reversible heat exchange. This is quickly smoothed out with increasing heat resistance where the internal losses, relatively speaking, become less significant. The longest column by contrast, even for very slight heat resistance, is operated optimally with an essentially flat heating profile: the same rate of heat transfer on every tray in each section of the column. This resistive effect is quite surprising, quickly washing out the structure of the optimal unconstrained column. It promises well for the simplified diabatic column design advocated by Rivero [8, 9] in which the external heat exchange medium is passed in sequence through heat exchangers on one tray after another in the stripping and rectifying sections, respectively. This design obviously alleviates the need for costly independent heat supply circuits for each tray. At the same time all heating curves show a marked reduction in heat duty for the reboilers and condensers, making it possible to include them inside the column rather than as separate exterior units.

The corresponding curves for the entropy production on each tray Figure 5, are even more illuminating for the effect of heat resistance. The entropy production due to heat and mass transfer on the trays alone is by no means flat, rather it follows the general shape originally developed in interior optimization. The entropy production due to

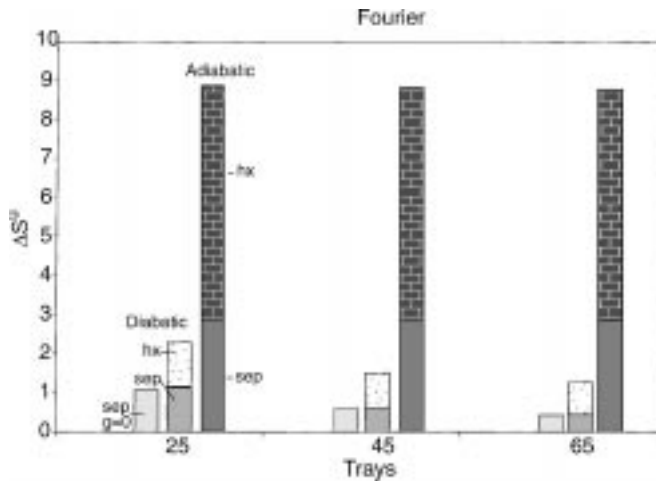


**Fig. 4.** Optimal heat duties for the individual trays in columns of length (from top to bottom) 25, 45, and 65 trays with Fourier heat transfer. The relative flow parameter  $\tilde{g}$  is as indicated in the figure. The value zero corresponds to no entropy generation in heat exchange, i.e. reversible. The middle value is industrially realistic, while the lower value is strongly resistive.

heat exchange, is not flat either, but together the internal and external entropy productions add up to an almost constant entropy production on each tray, a kind of equipartition often claimed to be a good design principle [13]. Optimal heat exchange on the feed plate is usually quite small and can be made zero by appropriately balancing of the vapor/liquid composition in the feed.



**Fig. 5.** Optimal distribution of entropy production on the individual trays in columns of length (from top to bottom) 25, 45, and 65 trays with Fourier heat transfer. Entropy production due to the internal heat and mass flows, the heat exchange and the sum are shown separately.



**Fig. 6.** Entropy production in adiabatic and diabatic columns of length 25, 45, 65 trays. Entropy production due to the internal heat and mass flows (sep) is shown in different shades of gray while heat exchange (hx) results in the hatched contributions. The ultimate lower limit is represented by the leftmost columns for a diabatic column with  $g = 0$ .

The entropy savings by using a diabatic column compared to a conventional adiabatic one are evident from Figure 6. Even for the shortest column of 25 trays, the adiabatic entropy production (including heat exchanger losses) is four times as large as the losses in the diabatic column, with the heat exchangers accounting for about two thirds of the losses. In the diabatic column, the contribution of heat exchange varies from about half for the shortest column to about two thirds for the longest column. To indicate the ultimate lower limit of entropy production without regard to heat exchange losses, the values for  $g = 0$  are shown as the leftmost columns in each group. In all the diabatic columns the internal separation losses hardly change by the introduction of heat exchanger losses, they are essentially additive, whereas in the adiabatic column even the internal losses are dramatically larger.

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Markus Schaller  
Prof. Karl-Heinz Hoffmann  
Department of Physics  
Technical University Chemnitz  
09107 Chemnitz  
Germany

Ricardo Rivero  
Mexican Institute of Petroleum  
07736 Mexico City D.F.  
Mexico

Prof. Bjarne Andresen  
Ørsted Laboratory  
University of Copenhagen  
Universitetsparken 5  
2100 Copenhagen  
Denmark

Prof. Peter Salamon  
Department of Mathematical Sciences  
San Diego State University  
San Diego, CA 92182-7720  
USA