

CONVECTIVE DRYING OF A MULTICOMPONENT FALLING FILM

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Abstract: The convective evaporation of a multicomponent falling liquid film into an inert gas with a co-current flow arrangement of the phases is studied. Simulations focused on the factors that influence the process controlling mechanisms and selectivity were performed using a mathematical model including liquid-side resistances. In absence of indirect heating, the process controlling steps change from a liquid-side-controlled process to a gas-phase-controlled one. Indirect heating makes evaporation less selective, and the process shifts from liquid-phase-controlled conditions to a process, in which neither the gas nor the liquid completely controls evaporation. Considering the evolution of the process controlling steps and its influence on selectivity, a model able to describe the whole evaporation process, must include the coexistence of several mechanisms.

Keywords: controlling steps, evaporation, multicomponent diffusion, phase equilibrium, selectivity

INTRODUCTION

Evaporation is defined as the removal of volatile substances from a solution or slurry. In some cases, the main purpose is to concentrate a solution. In other cases, the purpose may be to recover a solvent.

The uses of falling film evaporators are mainly in desalination, concentration of fluid foods, evaporation of temperature sensitive fluids, and in refrigeration. Some of the advantages of using falling film evaporation are: increased evaporation-side heat transfer, short contact time between liquid and heating surface, lack of static head (evaporation without elevation of boiling point), low temperature differences, and surface-only evaporation.

Most studies on falling film evaporation deals with the evaporation of pure liquids with emphasis on heat transfer and hydrodynamic phenomena (El-Genk and Saber, 2002; Du et al, 2002). These studies are mostly carried out in cylindrical and flat geometries arranged vertically or inclined. Others use plane or modified horizontal tubes to enhance heat transfer.

Concerning evaporation of multicomponent falling liquid films most studies found in the literature involve binary and ternary systems. Moreover, in most studies evaporation is into the vapours of the solvents contained in the liquid film mixture (Brotherton, 2002) and few ones address evaporation into an inert flowing gas (Baumann and Thiele, 1990; Agunaoun et al, 1998). Some studies focus on the influence of entrainment and deposition phenomena on mass transfer involving binary and ternary systems (Barbosa et al, 2003). Gropp and Schlünder (1985) study the influence of nucleate boiling and surface boiling on selectivity and heat transfer by liquid-side mass transfer resistances. They find that selectivity diminishes when increasing heat flux, and it is mainly controlled by thermodynamic equilibrium during surface boiling.

The purpose of this study is to present a qualitative analysis of the main factors that influence selectivity and control mechanisms during evaporation of multicomponent mixtures into an inert gas, that is, the inlet gas-to-liquid flow rates ratio, inlet liquid

temperature, gas velocity, and the supplying of indirect heat.

MATHEMATICAL MODEL

Mass and heat transfer depend on the transport in the gas phase, conditions at the interface, and diffusion as well as heat transfer in the liquid phase. Depending on the operating conditions, the process may be controlled by transport in the gas phase, in the liquid phase or by the equilibrium. Gas-phase-controlled evaporation usually occurs when the heat and molar fluxes in gas phase are low. In such cases, the process mainly depends on gas temperature, gas velocity and thermodynamic equilibrium, and not on diffusion and heat transfer in liquid phase.

Fig. 1 displays a differential volume element used to derive the model equations from the material and energy balances. Such a process usually involves indirect supply of heat through the supporting wall, heat and mass transfer through the liquid film, interface heat and mass transfer, complex diffusional interactions between the species in liquid and gas phase, as well as phase equilibrium. In addition, all these processes depend strongly on composition, temperature, flow regime of each phase, and the contact mode between the phases.

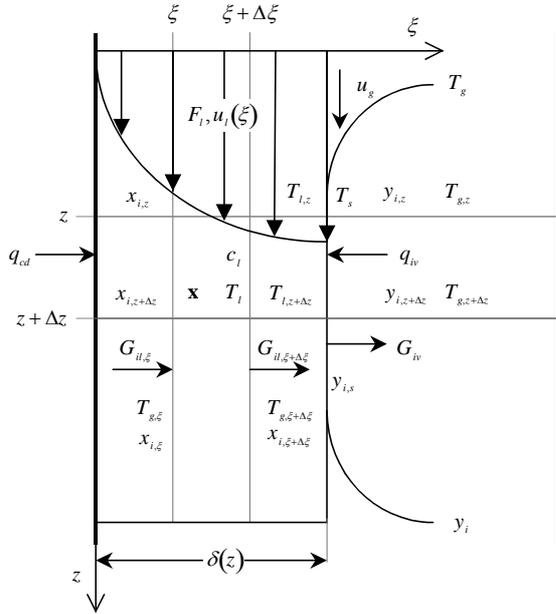


Fig. 1. Schematics of convective evaporation of a multicomponent falling liquid film in a differential volume element

This study focuses on the selectivity and the control mechanisms during evaporation. Therefore, the mathematical model is derived neglecting present interfacial tension effects, droplet entrainment and deposition, nucleate boiling and changes of film thickness. These factors, though important, are not species specific and are not expected to exert appreciable influence on the selectivity.

Mass and energy balances in the liquid phase

For each species in a mixture of n components, the mass balance over the liquid phase leads to the following differential equation:

$$u_l \frac{\partial(c_l x_i)}{\partial z} + \frac{\partial G_{i,l}}{\partial \xi} = 0 \quad i = 1, \dots, n \quad (1)$$

where u_l is the liquid velocity profile across the film thickness in the z direction, c_l is the total molar concentration of the liquid, x_i is the molar fraction of component i in a liquid mixture of n components, $G_{i,l}$ is the molar flux of component i in liquid phase and ξ is the space-coordinate in a direction perpendicular to the liquid flow.

A global mass balance can be written as:

$$u_l \frac{\partial c_l}{\partial z} + \frac{\partial G_l}{\partial \xi} = 0 \quad (2)$$

where G_l is the total molar flux. In matrix notation, the vector of liquid molar fluxes is given by:

$$\mathbf{G}_l = \mathbf{J} + G_l \mathbf{x} \quad (3)$$

where \mathbf{J} is the vector of diffusional molar fluxes referred to the average molar velocity.

For multicomponent mixtures the generalised Fick's law of diffusion gives the diffusion fluxes:

$$\mathbf{J} = -\mathbf{C}_l \mathbf{D} \frac{\partial \mathbf{x}}{\partial \xi} \quad (4)$$

where \mathbf{D} is the matrix of multicomponent diffusion coefficients, which is of order $n-1$ by $n-1$ since, according to the generalised Fick formulation, diffusion fluxes are functions of the gradients of the independently diffusing components. Introducing equation (3) and (4) into equation (1) results in:

$$u_l \frac{\partial(c_l \mathbf{x})}{\partial z} + \frac{\partial(G_l \mathbf{x})}{\partial \xi} = \frac{\partial}{\partial \xi} \left\{ c_l \mathbf{D} \frac{\partial \mathbf{x}}{\partial \xi} \right\} \quad (5)$$

If liquid flow is laminar, the velocity profile can be written as:

$$u_l = \frac{\rho_l g \delta^2}{2 \mu_l} \left\{ 2 \frac{\xi}{\delta} - \left(\frac{\xi}{\delta} \right)^2 \right\} \quad (6)$$

where ρ_l is the liquid density, g is the gravity acceleration, μ_l is the liquid viscosity, and δ is the film thickness.

The assumption that liquid flow in the film is laminar implies that mass and heat transfer, in the perpendicular direction to the liquid flow, is only diffusional. Equation (4) can then be rewritten as:

$$u_l \frac{\partial(c_l \mathbf{x})}{\partial z} = \frac{\partial}{\partial \xi} \left\{ c_l \mathbf{D} \frac{\partial \mathbf{x}}{\partial \xi} \right\} \quad (7)$$

The energy balance over the differential volume results in the following equation:

$$u_l \frac{\partial c_l C_{pl} T_l}{\partial z} = \frac{\partial}{\partial \xi} \left\{ \lambda_l \frac{\partial T_l}{\partial \xi} \right\} \quad (8)$$

where C_{pl} is the liquid molar heat capacity, λ_l is the liquid thermal conductivity, and T_l is the liquid temperature.

Mass and energy balances in the gas phase

The gas is considered to be inert and insoluble in the liquid and heat losses from the gas are neglected. A mass balance over the differential volume yields:

$$\frac{d\mathbf{F}_g}{dz} = a\mathbf{G}_v \quad (9)$$

where \mathbf{F}_g is the vector of molar flow of the vapours per cross section of the channel, \mathbf{G}_v is the vector of evaporation fluxes, and a is the specific contact area. An energy balance over the same differential element yields:

$$\mathbf{C}_{pg}^T \mathbf{F}_g \frac{dT_g}{dz} = -a q_{cv} \quad (10)$$

where \mathbf{C}_{pg} is the vector of molar gas heat capacities and q_{cv} is the convective heat flux. The superscript T denotes transposition.

Initial and boundary conditions

For the set of coupled partial differential equations (7) and (8), the following conditions may be applied:

Inlet conditions: $z = 0, \quad 0 \leq \xi \leq \delta$

$$\mathbf{x} = \mathbf{x}_0 \quad T_l = T_{l0} \quad (11)$$

Boundary conditions at the liquid-wall interface: $\xi = 0, \quad z > 0$

$$\frac{\partial \mathbf{x}}{\partial \xi} = \mathbf{0} \quad -\lambda_l \frac{\partial T_l}{\partial \xi} = q_{cd} \quad (12)$$

Boundary conditions at the liquid-gas interface: $\xi = \delta, \quad z > 0$

$$c_l \mathbf{D} \frac{\partial \mathbf{x}}{\partial \xi} = -\mathbf{G}_v$$

$$-\lambda_l \frac{\partial T_l}{\partial \xi} = q_{cv} - \{\mathbf{H}_g - \mathbf{H}_l\}^T \mathbf{G}_v \quad (13)$$

In the above expressions, q_{cd} is the heat flux supplied through the wall, \mathbf{H}_g is the vector of gas enthalpies, and \mathbf{H}_l is the vector of liquid enthalpies. The heat of mixing is neglected.

Equations (9) and (10) consist of a set of ordinary differential equations. For these equations only inlet conditions need to be supplied. Thus, for the gas

phase at the channel inlet ($z = 0$) the following conditions are applicable:

$$\mathbf{F}_g = \mathbf{F}_{g0} \quad (14)$$

$$T_g = T_{g0} \quad (15)$$

Interphase equilibrium

The coupling between the gas and liquid phases is given by:

$$\mathbf{y}_s = \frac{1}{P} \mathbf{P}^0 \boldsymbol{\gamma} \mathbf{x}_s \quad (16)$$

where \mathbf{y}_s is the vector of gas molar fractions at the interface, P is the total pressure, \mathbf{P}^0 is a diagonal matrix of liquid vapour pressures, $\boldsymbol{\gamma}$ is a diagonal matrix of activity coefficients, and \mathbf{x}_s is a vector of liquid molar fractions at the interface.

Mass and heat transfer rates

If diffusional interactions in gas phase are considered (the vector of evaporation fluxes for boundary conditions) may be written:

$$\mathbf{G}_v = \boldsymbol{\beta} \mathbf{K} \boldsymbol{\Xi} (\mathbf{y}_s - \mathbf{y}) \quad (17)$$

where \mathbf{y} is the vector of bulk gas molar fractions, \mathbf{K} is the matrix of zero-flux mass transfer coefficients, $\boldsymbol{\Xi}$ is the matrix of correction factors accounting for the influence of finite mass transfer rates upon the mass transfer coefficients, and $\boldsymbol{\beta}$ embodies a bootstrap relationship to calculate molar fluxes from diffusion fluxes (see Taylor and Krishna, 1993). The convective heat flux can be expressed by:

$$q_{cv} = \alpha \boldsymbol{\Xi}_h (T_m - T_l) \quad (18)$$

where $\boldsymbol{\Xi}_h$ is a correction factor for high mass transfer rates, α is a heat transfer coefficient at zero-mass flux. The indirect heat flux supplied by the heating medium is given by:

$$q_{cd} = h (T_m - T_l) \quad (19)$$

where h_m is a global heat transfer coefficient that describes heat transfer between the heating medium of temperature T_m and the liquid temperature at the wall.

Selectivity

For a multicomponent system selectivity is defined by (Martinez and Setterwall, 1991):

$$\mathbf{s} = \frac{\mathbf{G}_v}{G_v} - \mathbf{x} \quad (20)$$

where \mathbf{s} is a vector of the species selectivities, s_i . Species selectivity may be interpreted as follows:

$s_i > 0$ Species i is removed preferentially. The liquid molar fraction of the species decreases.

$s_i = 0$ The liquid molar fraction of species i remains constant.

$s_i < 0$ Species i is not removed preferentially. The liquid molar fraction of the species increases.

Controlling mechanisms

If the process is not controlled by equilibrium, conditions leading to a gas-phase or liquid-phase-controlled process can be estimated by means of a dimensionless group, known as the kinetic separation factor, according to the criteria developed by Schlünder (1988). The kinetic separation factor, modified here to account for a net effect of the species interacting in a multicomponent system, can be estimated by:

$$K_i = \exp\left(-\frac{G_i \delta}{C_i \|\mathbf{D}\|_\infty}\right) \quad (21)$$

where $\|\mathbf{D}\|_\infty$ is the infinity norm of the matrix of diffusion coefficients. The kinetic separation factor takes on values ranging from 0 and 1, i.e., from a liquid- to a gas-phase-controlled control respectively.

RESULTS AND DISCUSSION

The set of partial differential equations is solved through a Crank-Nicholson solution, and the mass and energy balances in the gas phase is solved using an Euler method. The system water-ethanol-acetone is used as a model mixture for simulations, and the activity coefficients are calculated using the Wilson equation with data from Gmehling and Onken (1982). Diffusion coefficients are calculated using Fuller's method (Reid et al, 1987) for the gas phase and the method of Bandrowski and Kubaczka (1982) for the liquid phase, using a correlation for the exponent ζ as a function of temperature estimated by Gamero and Martínez (2006). Other properties for pure components and their mixtures in both liquid and gas phases are calculated using methods reported by Reid et al (1987). The bootstrap matrix β is calculated assuming diffusion through stationary nitrogen. Mass and heat transfer coefficients at zero mass transfer rate are calculated, assuming a flat geometry, according to the boundary layer theory for streamline flow and the Chilton-Colburn analogy for turbulent flow. Numerical simulations are performed for the evaporation of a liquid mixture of fixed concentration and molar flow rate, high and low inlet liquid temperatures, and the following additional conditions:

- (a) Evaporation without heating, constant gas velocity, and high and low inlet gas-to-liquid flow rates ratios.
- (b) Evaporation with indirect heating, constant gas velocity, and high and low inlet gas-to-liquid molar flow rates ratios.

The inlet gas-to-liquid molar flow rates ratio is defined as:

$$R_0 = \frac{F_{g0}}{F_{l0}} \quad (22)$$

The following results are calculated for the following fixed conditions: $T_{g0} = 363.15$ K, $U_{l0} = 0.041$ m/s, $\mathbf{x}_0 = [0.15, 0.35, 0.50]$, $\delta = 1.5 \times 10^{-3}$ m. The liquid flow per width section is 1×10^{-3} kmol $\text{m}^{-1} \text{s}^{-1}$. The following parameters are used when adding indirect heat to the film: $T_m = 338.15$ K, $h = 1$ kW $\text{m}^{-2} \text{K}^{-1}$.

Influence of the inlet gas-to-liquid flow rates ratio

Since the inlet liquid flow rate is kept constant, a variation of the ratio R_0 , corresponds to a variation of the inlet gas flow rate. When evaporation proceeds without indirect heating at low R_0 , compositions in gas and liquid phases tend to reach equilibrium early along the contact path. Fig. 2a shows the composition in the liquid phase and Fig. 2b in the gas phase. Fig. 3a clearly shows that both liquid and gas temperatures decrease asymptotically to an equilibrium value due to the cooling effect caused by the evaporation.

As seen in Fig. 2a, under the conditions of the calculations, composition in the liquid is not affected considerably as in the case of the gas phase. When evaporation occurs at higher R_0 the compositions in the gas phase exhibit lesser changes, since there is a greater amount of gas compared to the evaporating liquid. Gas temperature is also less affected and displays small changes along the contact path (Fig. 3a). On the other hand, liquid temperature strives towards lower values. A taller channel would be needed to reach equilibrium for higher gas flow rates. With an increasing R_0 , evaporation loses the characteristics of a continuous process. In the extreme case when R_0 approaches infinity, evaporation tends to exhibit a batch-like behaviour for similar conditions and the establishment of equilibrium is not possible.

Fig. 4a shows selectivity curves when heating is not supplied. Calculations show that the evaporation is considerably more selective at the lower R_0 -value, and acetone is shown to evaporate preferentially. Since gas velocity is the same, the driving force mainly determines evaporation kinetics at the inlet, which is higher when the gas flow rate is higher. The higher evaporation rates cause a depletion of solvents on the surface and create a higher gradient in liquid phase to be overcome by diffusion. Consequently, a greater resistance in the liquid phase builds up, and the process becomes more liquid-phase-controlled and, therefore, less selective. After the entrance, the magnitude of selectivity increases along the contact path. This effect is more significant at low values of the flow rates ratio.

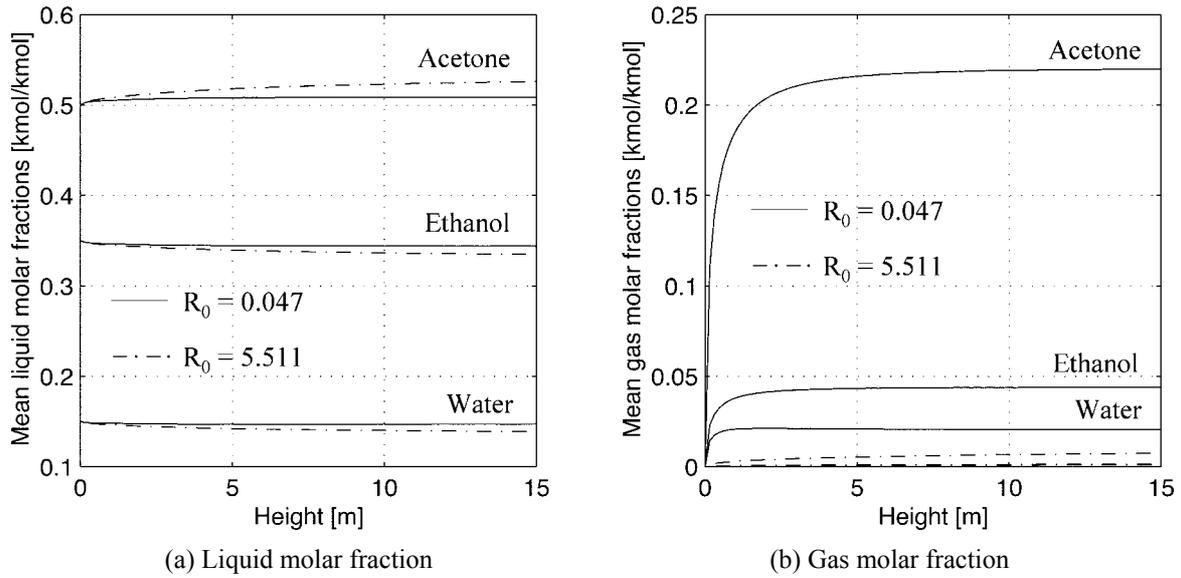


Fig. 2. Influence of R_0 on liquid and gas composition, without indirect heating, at $u_{g0} = 0.17$ m/s and $T_{i0} = 313.15$ K.

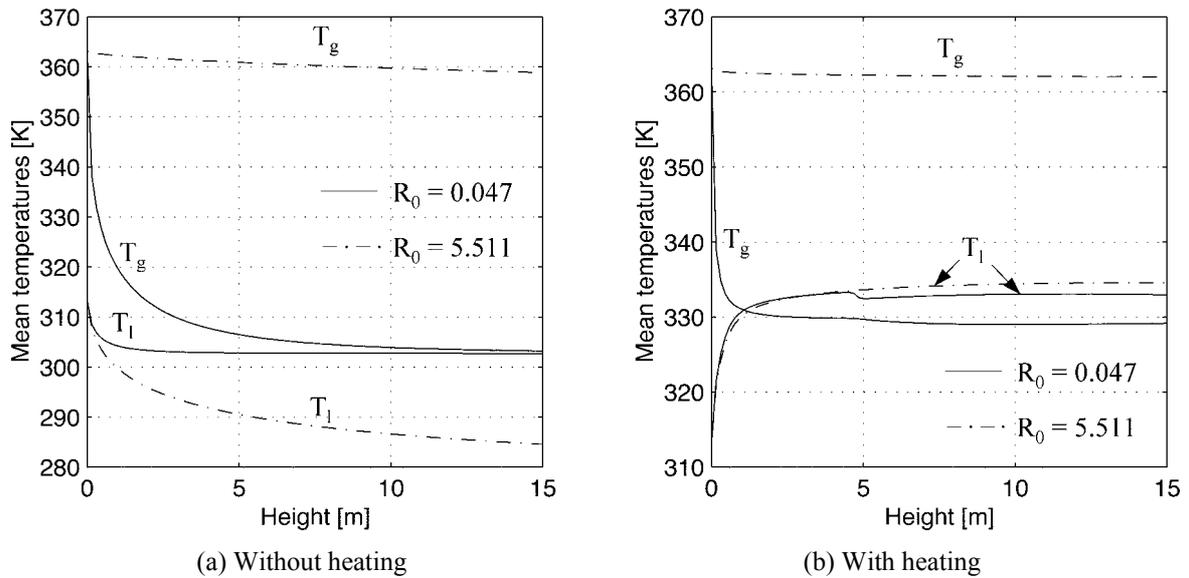


Fig. 3. Influence of R_0 on liquid and gas temperatures, at $u_{g0} = 0.17$ m/s and $T_{i0} = 313.15$ K.

At the conditions chosen for calculations, evaporation tends to be liquid-phase controlled within the section near the entrance, (see Fig. 5a). At lower R_0 -values and without indirect heating the process becomes rapidly gas-phase-controlled as the liquid film advances along the contact path. For higher flow rate ratios ($R_0 = 5.511$ in Fig. 5a) the kinetic separation factor is always less than unity and the process is never totally controlled by transport on the gas side. Thus evaporation consists of a combined phase-controlled process for most of the contact path length, which shifts from a dominating liquid side control at the entrance to an increased control by the gas phase.

The superficial depletion of components of higher relative volatility and the descent of liquid temperature results in that net evaporation rates (Fig. 5b) are high at the entrance, drop quickly within the entrance neighbourhood and eventually reach a zero mass transfer net flux at the equilibrium. High evaporation rates at the entrance result in high concentration gradients in the liquid. This makes evaporation liquid-phase-controlled. As the concentration of solvents in the gas enriches, evaporation rates diminish. Consequently, the gradients in the liquid phase decrease and the evaporation develops into a gas-phase-controlled process.

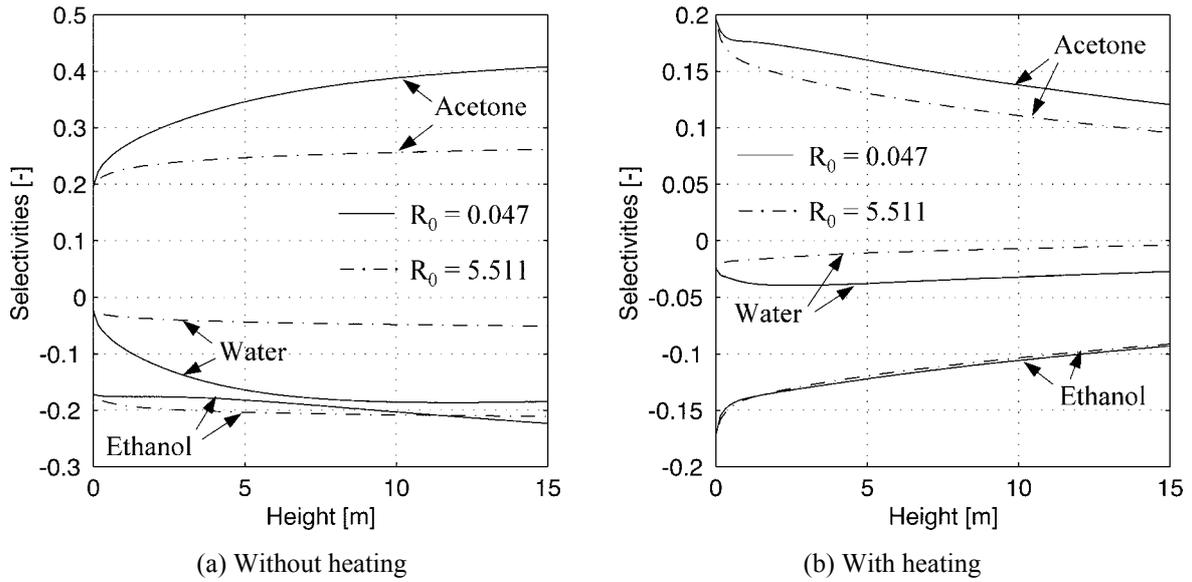


Fig. 4. Influence of R_0 on selectivity curves, at $u_{g0} = 0.17$ m/s and $T_{10} = 313.15$ K.

Influence of indirect heating

When the liquid film is heated indirectly, Fig. 4b (scale has been magnified), the flow rate ratio has a less significant influence on the selectivities. Indirect heating increases liquid temperature and evaporation rates making the process less selective.

If heat is indirectly supplied to the liquid film (Fig. 5a), the kinetic separation factor increases also rapidly at the entrance but tends later to a lower value compared to the case without indirect heating. As the gas enriches with vapours, the kinetic separation factor slowly increases striving towards a constant value with combined control of both gas and the liquid phases. Notice that within the entrance

section, at the lower flow rates ratio the kinetic separation factor quickly increases reaching a local maximum when gas temperature reaches the lowest value (see Fig. 3b).

Observe in Fig. 5a that when heating is supplied in the case of low gas flow rate, transition from streamline to turbulent flow regime takes place in the gas phase. On the other hand, when gas flow rate is high no transition is observed. Since the gas velocity is the same in both cases, only the effects of temperature on physical properties of the gas are responsible for the differences. The transition improves mass and heat transfer between the phases causing a higher evaporation rate. The sudden reduction of the kinetic separation factor reflects the

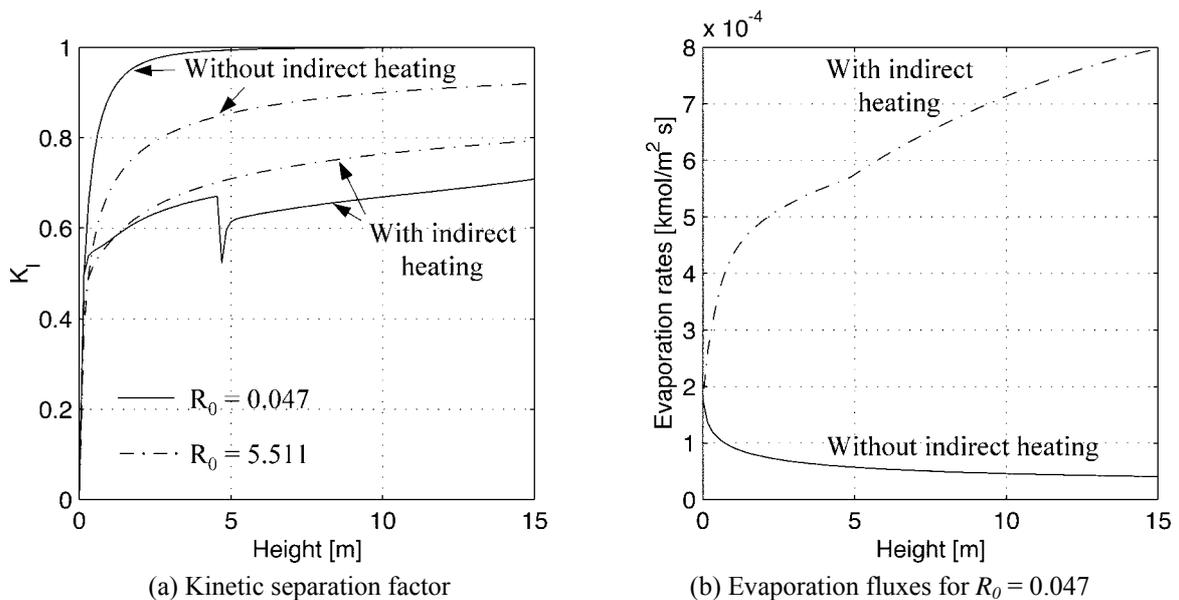


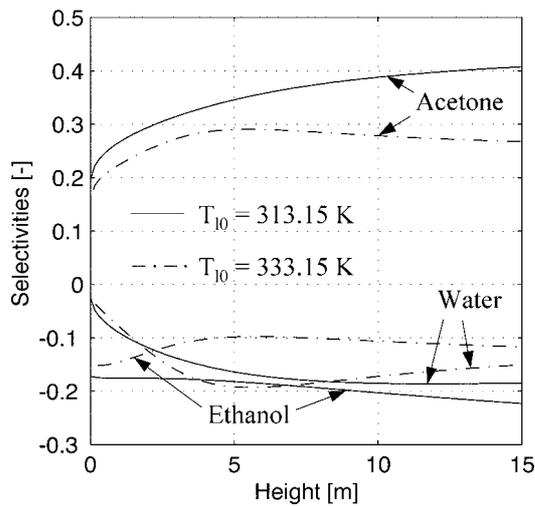
Fig. 5. Influence of R_0 , at $u_{g0} = 0.17$ m/s and $T_{10} = 313.15$ K.

sudden increase of the resistance in liquid phase.

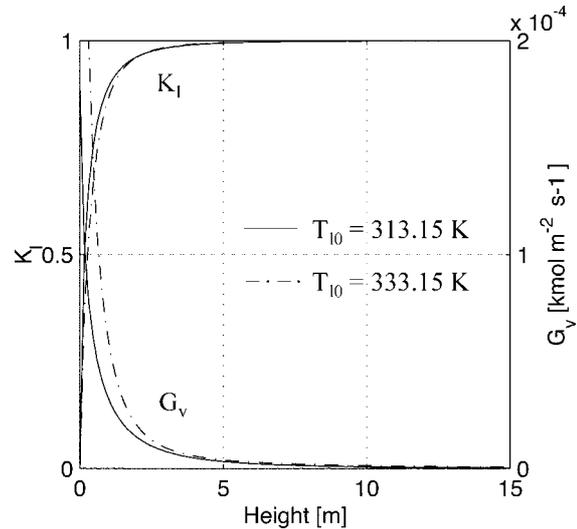
Influence of inlet liquid temperature

Fig. 6 displays the effect of the inlet liquid temperature on the evaporation process using a low R_0 -value without heating. Indirect heating makes the influence of other parameters on the control mechanism less significant and high values of R_0 make evaporation less selective. As noticed in Fig. 6a, evaporation becomes more selective at higher

phase controlled evaporation. The kinetic separation factor exhibits a similar behaviour with changes of the inlet gas-to-liquid flow rates ratio and the inlet liquid temperature. The process-controlling step develops from a process controlled by the liquid-side towards a process controlled by the gas side. The transition occurs faster and becomes more similar to a completely gas-side-controlled process at low values of these parameters. Consequently, the process becomes less selective at higher values of



(a) Selectivities



(b) Kinetic separation and evaporation flux

Fig. 6. Influence of liquid temperature, at $u_{g0} = 0.17$ m/s and $R_0 = 0.047$.

inlet liquid temperatures. A flash evaporation effect as soon as the liquid enters the channel contributes to high evaporation rates (Fig. 6b) and delays the transition of the kinetic separation factor to its maximum value. As a result, the film remains liquid-phase-controlled for a little longer contact path length. The fact that selectivity increases with liquid temperature is unexpected since high drying rates are associated to a less selective process. This effect is probably due to the influence of equilibrium at low R_0 .

CONCLUSIONS

This paper presents a qualitative study of the influence of the main parameters that affect the evaporation of a multicomponent falling liquid film in co-current flow with a gas stream. The study is performed through the simultaneous numerical solution of the mass and energy balances consisting of a set of partial and ordinary differential equations. The calculations show that in the absence of indirect heating, independent of the inlet conditions, it is difficult to maintain an evaporation process that is completely controlled by transport in the liquid phase when the liquid contains highly volatile components. The dynamic of the heat and mass transfer between the phases shifts the process inevitably towards gas-

inlet gas-to-liquid flow rates ratio. On the other hand, selectivity increases at higher inlet liquid temperatures. This is unexpected, since a higher resistance in the liquid phase is generally coupled to a lesser selectivity. A plausible explanation is the influence of the equilibrium, which under the condition of the calculations is not probably negligible. Addition of indirect heating to the liquid film makes evaporation less selective and lessens the effect of the other parameters. In all cases, evaporation goes from being liquid-phase-controlled at the inlet to rapidly reaching intermediate phase-controlling conditions where neither the gas nor the liquid completely controls evaporation. Considering the evolution of the process controlling steps and the coexistence of different mechanisms during important parts of the process, the knowledge of the asymptotic behaviour of selectivity is of very limited practical use to describe the process.

NOMENCLATURE

a	Specific area	$\text{m}^2 \text{m}^{-3}$
c	Molar concentration	kmol m^{-3}
C_p	Molar heat capacity	$\text{kJ kmol}^{-1} \text{K}^{-1}$
\mathbf{D}	Matrix of multicomponent diffusion coefficients	$\text{m}^2 \text{s}^{-1}$
\mathbf{F}	Vector of molar gas flow rate	

	per channel cross section area	$\text{kmol m}^{-2} \text{s}^{-1}$
G	Vector of molar fluxes with	$\text{kmol m}^{-2} \text{s}^{-1}$
H	Vector of molar enthalpies	kJ kmol^{-1}
<i>g</i>	Gravity acceleration	m s^{-2}
<i>h</i>	Heat transfer coefficient	$\text{kW m}^{-2} \text{K}^{-1}$
J	Vector of diffusion fluxes	$\text{kmol m}^{-2} \text{s}^{-1}$
K	Matrix of zero-flux mass transfer coefficients	$\text{kmol m}^{-2} \text{s}^{-1}$
K_l	Liquid side kinetic separation factor	-
<i>n</i>	Number of components	-
P⁰	Diagonal matrix of pure liquid vapour pressures	Pa
P	Pressure	Pa
<i>q</i>	Heat flux	$\text{kW m}^{-2} \text{s}^{-1}$
R_0	Inlet gas-to-liquid flow rates ratio	-
<i>s</i>	Selectivity	-
T	Temperature	K
<i>u</i>	Linear velocity	m s^{-1}
x	Vector of molar fractions in the liquid phase	kmol kmol^{-1}
y	Vector of molar fractions in the gas phase	kmol kmol^{-1}
<i>z</i>	Space dimension in the direction of liquid flow	m

Greek symbols

α	Heat transfer coefficient	$\text{kW m}^{-2} \text{K}^{-1}$
β	Bootstrap matrix, equation (17)	-
γ	Diagonal matrix of activity coefficients	-
δ	Film thickness	m
λ	Thermal conductivity	$\text{kW m}^{-2} \text{K}^{-1}$
μ	Viscosity	Ns m^{-2}
ξ	Space dimension perpendicular to liquid flow	m
Ξ	Matrix of correction factors in equation (17)	-
Ξ_h	Correction factor in equation (18)	-
ρ	Density	kg m^{-3}

Subscripts

<i>cd</i>	Conductive	<i>m</i>	Heating medium
<i>cv</i>	Convective	<i>s</i>	Interface
<i>g</i>	Gas	<i>v</i>	Evaporation
<i>l</i>	Liquid	<i>0</i>	Initial

Superscripts

<i>T</i>	Transposition
<i>0</i>	Saturation

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