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The Influence of Entrainment Separator on the Process of Molecular Distillation

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ABSTRACT

A model study of the influence of an entrainment separator on the efficiency of a molecular (short-path) distillation was performed. The temperature of the entrainment separator was determined from the balance equations for a one-dimensional model of molecular distillation. The initial period of distillation was investigated by using the direct simulation Monte Carlo method. The study of stream flows in an evaporator for a two-dimensional model of molecular distillation was also performed. The obtained results indicate that a sieve in the distillation gap of a molecular evaporator, acting as an entrainment separator, improves the composition of the distillate, although it also decreases the distillation rate. This effect is especially valuable when there is splashing, i.e., physical transfer of distilled liquid from the evaporator to the condenser. Hence, such a sieve stabilizes the distillate composition and muffles the effect of splashing.

INTRODUCTION

By placing a condenser in the immediate vicinity of an evaporator, which is a typical feature of molecular distillation, the probability of evaporated molecules returning to the evaporator is markedly reduced, thereby increasing the distillation rate (1). This geometrical layout, however, has one disadvantage. If the distilled liquid contains an easily volatile component (dissolved gas, traces of a solvent), this substance will cause splashing of the evaporating film as soon as it gets into the vacuum onto the evaporation surface. As a result, a portion of the distilled liquid falls in the form of droplets as far as the condenser, thus worsening the quality of the distillate (2). With the aim to eliminate this negative phenomenon a mechanical impediment, an entrainment separator, is located between the evaporator and the condenser to prevent splashed material from reaching the condenser. At the same time, the entrainment separator prevents evaporated molecules from falling directly onto the condenser, thus reducing the distillation rate (3).

The present study was designed to examine the consequences of the presence of an internal condenser on the separation power and rate of distillation with splashing. Since molecules hitting the internal condenser condense and reevaporate, an improvement of separation power can be expected. However, such an arrangement disturbs the ideal short-path flow of molecules—the basis of molecular distillation—a fact not without consequences on the material balance.

Three different mathematical approaches to molecular distillation (MD) modeling have been employed. The one-dimensional model of MD, based on balance equations, supplied data on the steady state of distillation, i.e., the temperatures and liquid composition on the internal condenser sieve.

The time-dependent description of temperature and composition on the sieve in the one-dimensional model was modeled by and on the direct simulation Monte Carlo (DSMC) method and gave another set of temperature and composition data for the steady state of the liquid film on the sieve.

Finally the two-dimensional model attempted to solve balance equations for both heat and mass transfer—a method which allows, unlike the one-dimensional models, an assessment of the downward flow on the sieve. An ideally mixed film without concentration and temperature gradients on the sieve was assumed in all three models.

PROBLEM ANALYSIS

Figure 1 shows the scheme of a molecular distillator where the evaporator and the condenser take the form of two plain, parallel, infinitely large surfaces, and an entrainment separator takes the form of a sieve situated between them. The scheme shows the most simple case when the temperature and the composition of the liquid on the evaporator are constant and the liquid on the sieve and on the condenser is ideally mixed (without gradient of temperature and composition).

Let the evaporated liquid consist of two components, A and B. The quantity J_{1A} of component A is evaporated from the evaporator. From

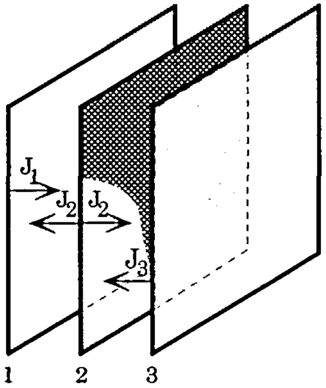

FIG. 1 Scheme of plane (one-dimensional) molecular evaporator with sieve: (1) evaporator, (2) sieve, (3) condenser.

the sieve, quantity J_{2A} is evaporated from both sides (toward the evaporator and condenser, respectively). If the temperature of the condenser is low enough, reevaporation the from condenser is negligible and $J_{3A} = 0$. In the steady state, when the collisions between molecules in the vapor state do not affect the process, the relationship:

$$J_{1A} = 2J_{2A} \tag{1}$$

also holds for component B:

$$J_{1B} = 2J_{2B} \tag{2}$$

Based on the Langmuir–Knudsen equation it is possible to express these flows as

$$J_{ij} = x_{ij}K_i(T_j); \qquad i = A, B; \qquad j = 1, 2, 3 \tag{3}$$

where

$$K_i(T_j) = p_i^0(T_j)(2\pi M_i RT_j)^{-1/2} \tag{4}$$

The composition and the temperature of the liquid on the sieve in the steady state can by determined by solving a set of nonlinear equations:

$$x_{A1}K_A(T_1) = 2x_{A2}K_A(T_2) \tag{5.1}$$

$$x_{B1}K_B(T_1) = 2x_{B2}K_B(T_2) \tag{5.2}$$

$$x_{Aj} + x_{Bj} = 1 \tag{5.3}$$

However, the solution of Eqs. (5.1)–(5.3) neither allows determination of the quantity of liquid on the sieve in the steady state nor does it supply information about the time development of the process to the steady state.

It is possible to obtain these data by using the direct simulation Monte Carlo method (4). By using this method, all velocity coordinates and those position coordinates of molecules in the direction of which the change of characteristics occurs are stored in the computer memory. The DSMC method enables modeling the time development of the system described in Fig. 1 from the initial state when liquid is only on the evaporator (its temperature and composition are constant during time development; the sieve and condenser are dry) to steady state. The temperature of the condenser is constant during the simulation, and the initial temperature of the sieve is determined by heat radiation between the evaporator and the condenser:

$$T_2 = \left(\frac{T_1^4 + T_3^4}{2}\right)^{1/4} \qquad (6)$$

At time $t = 0$, evaporation starts from the evaporator. Evaporated molecules condense on the sieve, and the released condensation heat causes an increase of the sieve temperature. As a consequence of this, reevaporation of the molecules from both sides of the sieve follows.

An industrial molecular evaporator with an entrainment separator is shown in Fig. 2. Evaporator, entrainment separator (represented by the

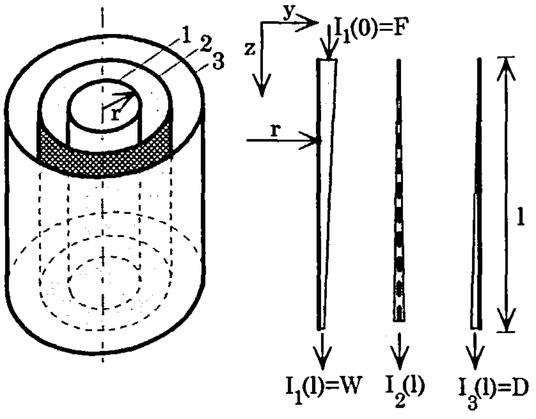

FIG. 2 Scheme of cylindrical (two-dimensional) molecular evaporator with sieve: (1) evaporator, (2) sieve, (3) condenser.

steel sieve), and condenser are arranged as coaxial cylinders. The feed F flows by gravity down the surface of the evaporator. Both the evaporated and the splashed molecules are trapped by the sieve. Reevaporation from the sieve occurs in both the direction to the evaporator and the direction to the condenser. The molecules reaching the condenser condense on it and flow down by gravity. Three streams are drained from the bottom of the evaporator: residue W, distillate D, and a stream from the sieve I_2. Stream I_2 is added either to the residue or to the feed.

The system of balance equations for the molecular evaporator, which is elaborated in Ref. 5, must be transformed to take into account a sieve as an entrainment separator.

The condition that all three films are ideally mixed and that reevaporation from the condenser does not occur has been assumed. The system of balance equations consists of equations of continuity for film on the evaporator, on the sieve, and on the condenser. The equation of continuity in the steady state is the same for all films:

$$\operatorname{div}(\vec{v}_j c_{Aj}) - \operatorname{div}(D_{AB} \operatorname{grad} c_{Aj}) = 0 \tag{7}$$

The velocities $\vec{v}_j$ for individual films are described by the Navier–Stokes equation whereby the following condition may be used:

$$\vec{v}_j(y, z, t) = (0, 0, v_y(0, y, z))$$

The evaporation rate from the surface of the jth film is for pure substance A as described by the Langmuir–Knudsen equation (4). When splashing occurs, the total rate of the evaporation of component A is described by

$$K_{ij}^{\text{tot}} = K_{ij} x_i(z) + K_j x_i(z) \tag{8}$$

where K_j is a constant expressing the rate of splashing from surface j. This value must be determined from experiment.

It is possible to express the flow on the evaporator for component i as

$$I_{ij} = 2\pi r \int_0^{h(y)} v_j c_{ij} \, dx \tag{9}$$

and then the total stream on the evaporator is

$$I_j = I_{Aj} + I_{Bj}$$

On the condition that the process of evaporation is isothermal and that collisions between molecules in the vapor phase are neglected, we obtain equations for all streams in the evaporator by using Eqs. (8) and (7):

$$\frac{d}{dz} I_{A1} = -2\pi r(K_{A1} + K_1)\frac{I_{A1}}{I_1} + 2\pi r K_{A2}\frac{I_{A2}}{I_2} \tag{10.1}$$

$$\frac{d}{dz} I_{B1} = -2\pi r(K_{B1} + K_1)\frac{I_{B1}}{I_1} + 2\pi r K_{B2}\frac{I_{B2}}{I_2} \qquad (10.2)$$

$$\frac{d}{dz} I_{A2} = -4\pi r K_{A2}\frac{I_{A2}}{I_2} + 2\pi r(K_{A1} + K_1)\frac{I_{A1}}{I_1} \qquad (11.1)$$

$$\frac{d}{dz} I_{B2} = -4\pi r K_{B2}\frac{I_{B2}}{I_2} + 2\pi r(K_{B1} + K_1)\frac{I_{B1}}{I_1} \qquad (11.2)$$

$$\frac{d}{dz} I_{A3} = 2\pi r K_{A2}\frac{I_{A2}}{I_2} \qquad (12.1)$$

$$\frac{d}{dz} I_{B3} = 2\pi r K_{B2}\frac{I_{B2}}{I_2} \qquad (12.2)$$

Reevaporation from the condenser back to the sieve has been neglected.

RESULTS AND DISCUSSION

We used an equimolar mixture of di-butyl-phthalate (DBP) and di-butyl-sebacate (DBS) as a model system [M_{DBP} = 278.35 g·mol^{-1}, log p^0_{DBP} = 12.7 − 4450/T Pa, $\Delta_{evp}H_{DBP}$ = 85.6 kJ·mol^{-1}, $c_{p\,DBP}$ = 583 J·mol^{-1}·K^{-1}, ρ_{DBP} = 1294.6 − 0.839T kg·m^{-3}, M_{DBS} = 314.46 g·mol^{-1}, log p^0_{DBS} = 13.22 − 4850/T Pa, $\Delta_{evp}H_{DBS}$ = 92.77 kJ·mol^{-1}, $c_{p\,DBS}$ = 632 J·mol^{-1}·K^{-1}, ρ_{DBS} = 1169.6 − 0.78T kg·m^{-3} (6)].

For the one-dimensional model (Fig. 1) we determined the sieve temperature by solving the set of Eqs. (5.1)–(5.3) by an iteration algorithm. Next, we performed a study of the initial phase of molecular distillation by the DSMC method. For the cylindrical evaporator (Fig. 2) we numerically solved Eqs. (10.1)–(12.2) and, as a result, we determined the dependence of streams on all surfaces on the position.

All computations were carried out on an IBM RS/6000 computer equipped with 64 MB RAM. All programs were written in the GNU C++ language ver. 2.7.2.

Plane One-Dimensional Molecular Evaporator

The set of Eqs. (5.1)–(5.3) was solved by an iteration algorithm. Table 1 collects temperatures and compositions of the liquid phase on the sieve as calculated for various evaporating temperatures T_1 and for splashing of up to 20%. The liquid phase on the sieve is generally cooler than that on the evaporator. Since the relative volatility decreases with the temperature, the volatile component is depleted and its content on the sieve is just marginally under that of the evaporator. Table 1 also shows a directly

TABLE 1
Temperature and Composition of the Film on the Sieve in Steady-State for Various Evaporation Temperatures and Splashing Relations

	Without splashing		Splashing 5%		Splashing 10%		Splashing 20%	
T_1 (K)	T_2 (K)	x_{2A}	T_2 (K)	x_{2A}	T_2 (K)	x_{2A}	T_2 (K)	x_{2A}
353.0	345.0	0.483	345.6	0.484	346.1	0.485	347.1	0.487
373.0	364.1	0.484	364.7	0.485	365.3	0.486	366.4	0.488
383.0	373.6	0.484	374.3	0.486	374.9	0.487	376.0	0.488
403.0	392.6	0.485	393.3	0.486	394.0	0.487	395.3	0.489
413.0	402.1	0.485	402.8	0.486	403.5	0.487	404.9	0.489
423.0	411.5	0.485	412.3	0.486	413.1	0.487	414.5	0.489

proportional rate of splashing to the sieve temperature T_2, i.e., a higher amount of splashing increases the T_2.

Figure 3 presents time development of the sieve temperature, and Fig. 4 presents time development of the molar fraction of DBP in the film on the sieve and condenser. The results obtained for steady state by the DSMC method are identical with the solution of Eqs. (5.1)–(5.3) summarized in Table 1.

Cylindrical Two-Dimensional Molecular Evaporator

The set of Eqs. (10.1)–(12.2) was solved numerically by the Runge–Kutt method of the 4th order. The value of K_1 was determined as

$$K_1 = \frac{n}{100}(K_{A1} + K_{B1}) \tag{13}$$

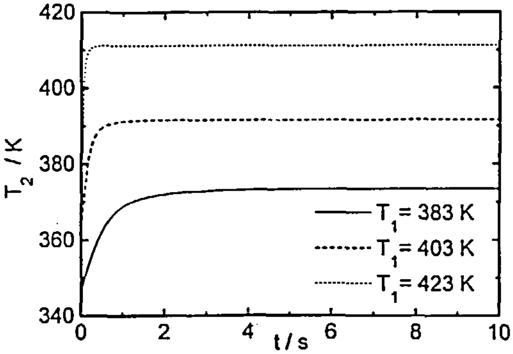

FIG. 3 Time development of the sieve temperature in plane molecular evaporator.

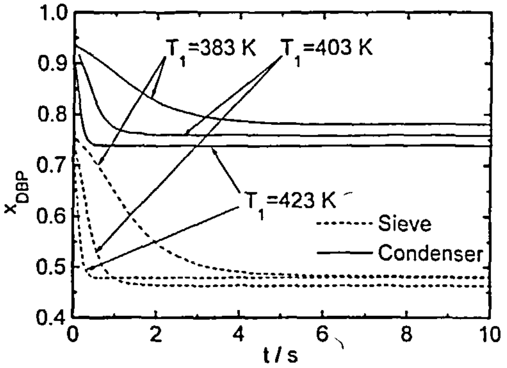

FIG. 4 Time development of film composition on the sieve and on the condenser in plane molecular evaporator.

where n = 5, 10, 20%. The values of n determined are in accord with the experimental results obtained by Malyusov et al. (2). In all cases $I_{1\ \mathrm{DBP}}(0) = 0.075\ \mathrm{mol{\cdot}s^{-1}}$, $I_{1\ \mathrm{DBS}}(0) = 0.075\ \mathrm{mol{\cdot}s^{-1}}$ ($\Gamma = 51\ \mathrm{kg{\cdot}m^{-1}{\cdot}n^{-1}}$), r = 0.05 m, the length of evaporator was 1 m, evaporating temperatures were 353, 373, and 383 K respectively, and the condensing temperature was 273 K, i.e., enough to render reevaporation insignificant. The temperature of the sieve was determined by its dependence on the evaporation temperature from Table 1.

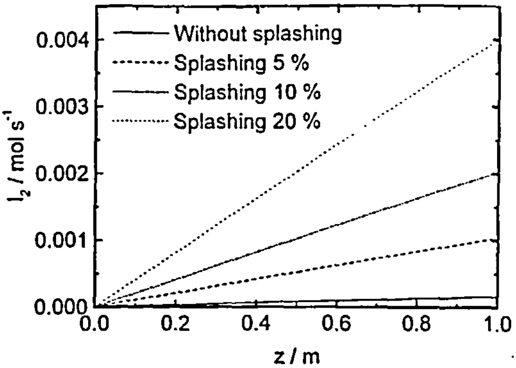

FIG. 5 Dependence of stream I_2 on the position on sieve with and without the existence of splashing.

Figure 5 shows a plot of the downward sieve flow l_2 against the increasing splashing K_1 at $T = 373$ K. The sieve flow is minimal at low splashing, but rises significantly at more intense splashing. The downward flow grows constantly, being replenished by vapors coming from the evaporator. However, the major part of l_2 originates from splashing, and consequently the rising temperature of the evaporator would cause more splashing according to Eq. (13).

The plot of the size of l_2 against the height of a molecular evaporator operating at zero splashing is shown in Fig. 6. The downward flow l_2 grows at higher temperatures because then there is more vapor coming from the evaporator.

Figure 7 depicts the relationship between the composition of liquid on the sieve and the height of the equipment at several values of K_1 and $T_1 = 373$ K. The content of the more volatile component in the liquid on the sieve was found to be the highest at $K_1 = 0$ and vice versa, but the absolute values of the difference calculated at extreme values of splashing are small and vary but little along the equipment. The concentration of the more volatile component in sieve flow l_2 decreases on going downward from the feed point, paralleling the same trend on the evaporator.

The dependence of the liquid composition on the sieve on the vertical coordinate (height) when operating at zero splashing and at various evaporating temperatures is shown on Fig. 8. As expected, the enrichment of vapors in the more volatile component is worst at highest temperatures.

From Fig. 9 the role of the sieve placed in the distillation gap as an entrainment separator becomes evident. Because it works as an internal

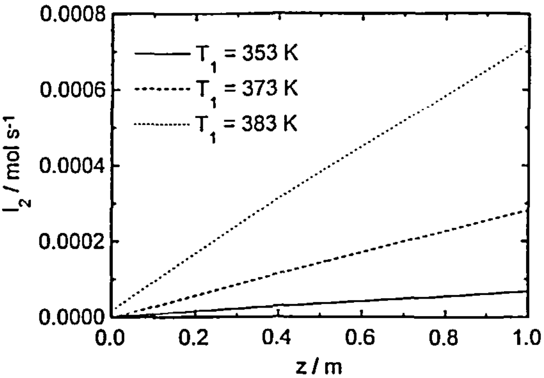

FIG. 6 Dependence of stream l_2 on the position on sieve for various evaporation temperatures.

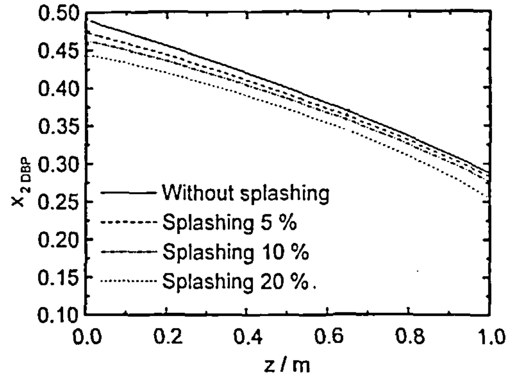

FIG. 7 Dependence of film composition on the sieve on the position in evaporator with and without the existence of splashing.

condenser with reevaporation, it intercepts some of the distillate, as well as the splashing.

Figure 10 illustrates two different situations. It shows the content of more volatile component on the condenser plotted against the vertical coordinate in experimental arrangements with and without the sieve. If no sieve is present, the splashing evidently spoils the composition of the condensate. The decay of $x_3 = f(z)$ when there is no sieve is more pronounced than with the entrainment in place. The presence of a sieve as

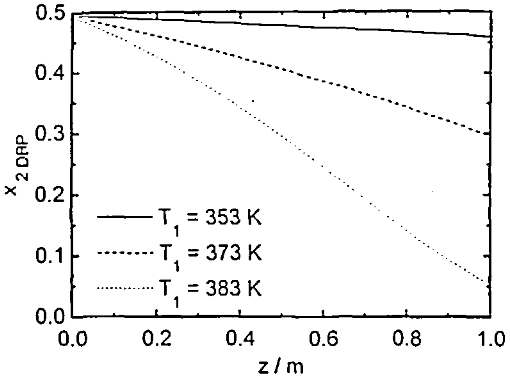

FIG. 8 Dependence of film composition on the sieve on the position in evaporator for various evaporation temperatures.

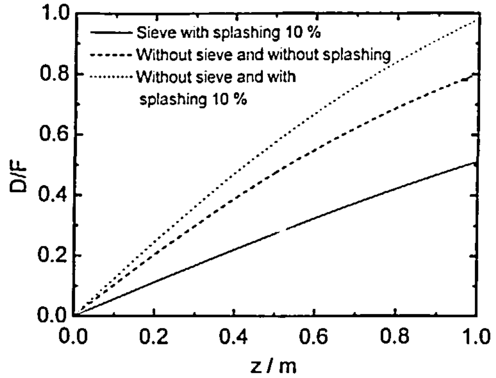

FIG. 9 Dependence of *D/F* on position in condenser for various modes of distillation.

an internal condenser introduces a significant change. The condensate is now generally richer in the more volatile component and its composition is almost independent of the vertical coordinate. This holds true for all values of splashing. Thus the difference in condensate composition at zero splashing and the highest splashing is small. In all four examined cases the temperature of the evaporator was constant at 373 K.

Figure 11 plots the composition of the distillate (condensate) against height z for four values of splashing and an evaporation temperature of 373 K. The dependence of x_3 on the vertical coordinate is relatively small

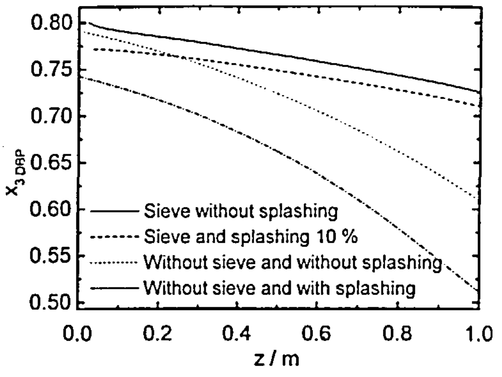

FIG. 10 Dependence of composition of the distillate on the position in evaporator for various modes of distillation.

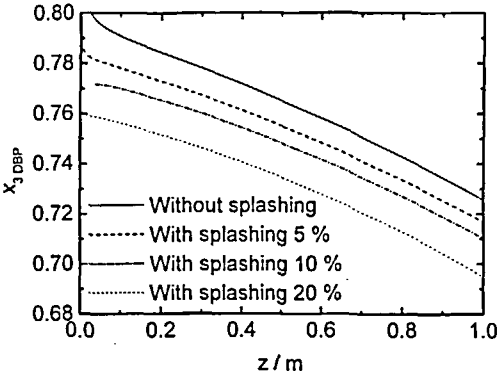

FIG. 11 Dependence of composition of the distillate on the position in evaporator with and without splashing.

for all selected values of splashing. Absolute values of differences in $x_3 = f(K_1)$ are nearly constant along the condenser. It is therefore evident that a sieve in the distillation gap has a stabilizing effect on the composition of the distillate.

Figure 12 depicts the dependence of the evaporated portion of the distillate on the vertical position as calculated for various values of splashing and an evaporation temperature of 373 K. Since the evaporated portion increases almost linearly with height, the D/F values do not vary much with splashing. Higher D values at higher splashing are caused by warmer

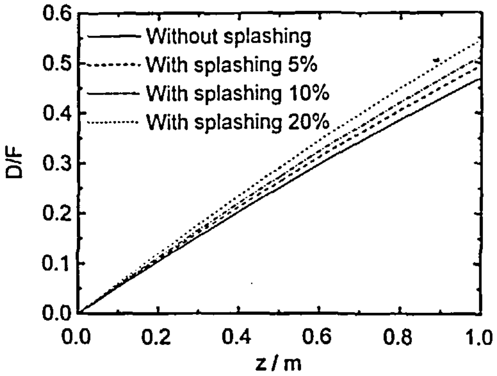

FIG. 12 Dependence of D/F on position in condenser for various values of splashing.

liquid on the sieve, the liquid being heated by the splashing itself (Table 1).

Thus, molecular distillation with an entrainment separator is the method of choice when high purity of distillate is required. The small decrease in distillation output can be compensated for by slightly increasing the temperature of the evaporator so that there is no danger of thermal decomposition. Of special value can be an entrainment in cases when the feed is colored and even a small splashing would taint the distillate and make it partially or completely unusable. Another good example is the distillation of electroinsulating materials where the desired low conductance of the distillate would be compromised by splashing.

CONCLUSIONS

The presence of a sieve as an entrainment separator in the distillation gap decreases the rate of distillation but improves significantly the separatory power of a molecular distillation. This effect is especially marked and therefore valuable when the distillation is accompanied by splashing. An entrainment separator mitigates the negative effects of splashing and stabilizes the composition of the distillate. Our mathematical model of an entrainment tallies well with the experimental evidence.

NOTATION

Variables and Constants Used

c_{ij}	molar concentration of component i on surface j ($\mathrm{mol{\cdot}m^{-3}}$)
c_j	molar concentration of the liquid on surface j ($\mathrm{mol{\cdot}m^{-3}}$)
D	distillate flow ($\mathrm{mol{\cdot}s^{-1}}$)
D_{AB}	diffusion coefficient ($\mathrm{m^2{\cdot}s^{-1}}$)
F	feed flow ($\mathrm{mol{\cdot}s^{-1}}$)
I_{ij}	molar flow of component i on surface j ($\mathrm{mol{\cdot}s^{-1}}$)
I_j	total molar flow on surface j ($\mathrm{mol{\cdot}s^{-1}}$)
J_{ij}	rate of evaporation of component i from surface j ($\mathrm{mol{\cdot}m^{-2}{\cdot}s^{-1}}$)
K_{ij}	evaporation constant of component i on surface j ($\mathrm{mol{\cdot}m^{-2}{\cdot}s^{-1}}$)
K_j	splashing constant on surface j ($\mathrm{mol{\cdot}m^{-2}{\cdot}s^{-1}}$)
K_{ij}^{tot}	total evaporation constant of component i on surface j ($\mathrm{mol{\cdot}m^{-2}{\cdot}s^{-1}}$)
M_i	molar mass of component i ($\mathrm{kg{\cdot}mol^{-1}}$)
p_i^0	saturation pressure of component i (Pa)
R	universal gass constant, 8.314510(70) $\mathrm{J{\cdot}K^{-1}{\cdot}mol^{-1}}$
r	radius of the evaporating cylinder (m)

T_j	temperature of surface j (K)
t	time (s)
v_j	velocity of the flow on surface j ($m \cdot s^{-1}$)
W	residue flow ($mol \cdot s^{-1}$)
x_{ij}	mole fraction of component i on surface j
y	axis direction radial to the evaporation cylinder (m)
z	axis direction parallel with the evaporation surface (m)

Greek Letters

Γ	peripheral liquid loading on the top of evaporator ($kg \cdot s^{-1} \cdot m^{-1}$)
π	3.141593

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