EFFECTS OF HEAT-TRANSFER CONDITIONS ON THE PERFORMANCE OF A LIQUID THERMAL-DIFFUSION COLUMN

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Theoretical concepts on the parasitic-correction mechanism [1] in thermal diffusion columns have been confirmed by experiment.

It is stated [1] that the density difference in a binary mixture along the column perimeter is a consequence not only of nonuniform temperature distribution but also of concentration asymmetry, which inevitably accompanies the latter.

We have checked the results of [1] on bromine isotopes in butyl bromide. The contents of the two bromine isotopes are almost identical, so it is possible to use relationships derived by considering the parasitic convection for the mixture characterized by the condition $c(1-c) \approx p$, where p is a constant.

In that case we have [1, 2]

$$q = \left[1 + \frac{1}{\varkappa}(1 - e^{-\varkappa y_e}) \right]^2 \left[1 - \frac{1}{\varkappa}(1 - e^{-\varkappa y_c}) \right]^{-2}, \tag{1}$$

$$\varkappa y_e = a \pm \frac{2pb}{\varkappa y_e} \left[y_e - \frac{1}{\varkappa}(1 - e^{-\varkappa y_e}) \right], \tag{2}$$

where $\varkappa$ is the parasitic-convection parameter, while a and b are parameters incorporating the thermal and concentration asymmetry.

TABLE 1. Column Operation with Steam Heating (P = 1.4 atm) in Relation to Water Flow Rate G

Parameter	G, ton/h								
	0,25	0,27*	0,65	1,0	1,83	2,25*	2,6	3,2	4,0*
Degree of separation q	1,17	1,165	1,23	1,275	1,32	1,34	1,34	1,30 1,37*	1,32
Hot couple reading, °C	108,0	109,0	107,0	106,0	107,0	107,0	107,0	104,0 107,5*	105,0
Cold couple reading, °C	33,5	28,0	19,5	19,0	16,5	16,2	15,5	15,0 14,2*	13,3
Temperature difference across gap ΔT, °C	71,5	77,8	85,0	83,5	87,0	87,5	87,8	86,0 89,5*	87,5
Re	2580	2780	6700	10300	18850	23200	26800	33000	41200
Nu	30	32,8	64,5	91	210	247	277	328	392
α_2, W/m²·deg	2010	2200	4300	6100	14000	16500	18500	21900	26300
h_2, W/m²·deg	95	101	150	178	235	244	249	257	265
Heat flux q_1, W/m	4400	4760	5210	5110	5330	5360	5370	5270 5480*	5360
$a_T = a$	1,5	1,77	3,31	3,80	4,47	4,60	4,60	4,70	4,75
$(\delta T)_T$, °C	0,195	0,23	0,43	0,5	0,58	0,60	0,60	0,61	0,62

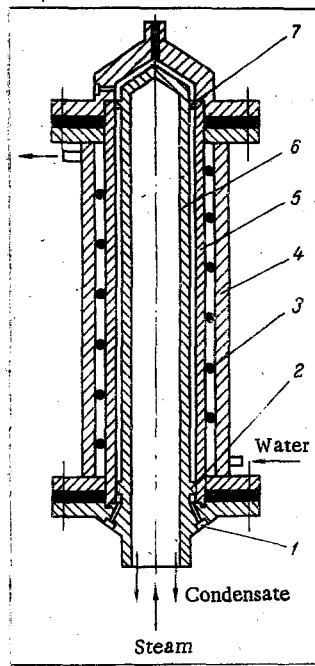Note. An asterisk denotes a value from the second series of runs.

Fig. 1. The thermal-dif-
fraction column: 1) sampler;
2) lower rim; 3) spiral in-
sert; 4) condenser body;
5) outer cylinder; 6) inner
cylinder; 7) upper rim

The value of a includes the joint effects of two factors, which deter-
mine the temperature nonuniformity around the perimeter: the nonunifor-
mity due to eccentricity of the internal cylinder relative to the outer one
and the nonuniformity due to variations in heat-transfer conditions in the
azimuthal direction, which may arise from variations in channel geometry,
scale deposits, uneven roughness in the heat-transfer surfaces, and non-
uniform thickness in the condensate film, which may be a consequence of
deviation from vertical in the column axis and also of effects produced by
the flow of steam in a direction opposite to that of the condensate.

Then

$$a = a_\varepsilon + a_T, \qquad (3)$$

where we have [1]

$$a_\varepsilon = 7.56 \cdot 10^3 \frac{\eta DL\varepsilon}{\rho g \beta \delta^5 \Delta T} \varphi_\varepsilon. \qquad (4)$$

The value of φ_ε is dependent only on the heat-transfer conditions,
and for cylindrical column geometry

$$\varphi_\varepsilon = \frac{(h_1/h_2) - 1}{1 + (h_1/h_2) + (h_1 \delta / d\lambda_L)}. \qquad (5)$$

We get for a_T from the standard relationships of heat-transfer theory
that

$$a_T = 2.88 \cdot 10^3 \frac{\eta DL}{\pi \rho g \beta \delta^4 \Delta T} \varphi_T,$$

where

$$\varphi_T = \left[\left(1 + \frac{2d\lambda_L}{\delta h_2} \right) \left(\frac{h_1'}{h_1''} - 1 \right) - \frac{h_1}{h_2} \left(1 - \frac{2d\lambda_L}{\delta h_1} \right) \left(\frac{h_2'}{h_2''} - 1 \right) \right]$$
$$\times \left(1 + \frac{h_1}{h_2} + \frac{\delta h_1}{d\lambda_L} \right)^{-1}$$

is also dependent on the heat-transfer conditions.

The quantities h_1', h_1'', h_2', h_2'' appearing in (7) are the reduced heat-transfer coefficients for regions
corresponding to the first parasitic flow (one prime) and the second one (two primes). We have
used a simplified model [1, 2], in which the temperature and concentration nonuniformities produce two
parasitic flows equal in magnitude but opposite in direction.

The unprimed reduced heat-transfer coefficients are means over the perimeter.

It follows from (4)-(7) that a_ε can be calculated to an error adequate for practical purposes if the
physical characteristics of the mixture are known together with the column geometry and the heat-transfer
conditions at both the working surfaces.

TABLE 2. Column Operation with Ethanol Vapor Heating (P = 3 atm)
in Relation to Water Flow Rate G

| Parameter	G, ton/h				
	0,40	1,0	2,0	3,0	4,0
Degree of separation q	1,07	1,11	1,17	1,17	1,20
Hot couple reading, °C	87,2	82,5	81,1	81,5	78,1
Cold couple reading, °C	20,9	17,1	13,7	13,9	13,1
Temperature difference across gap ΔT, °C	63,8	62,8	64,8	65	62,5
Re	4150	10300	20600	30900	41200
Nu	50	91	222	311	392
α_2, W/m^2·deg	3340	6100	14900	20800	26300
h_2, W/m·deg	131	178	238	255	265
Heat flux q_l, W/m	4060	4010	4130	4150	4000
$a_T = a$	—2,2	+1,525	+3,75	+4,1	+4,4
$(\delta T)_T$, °C	—0,16	+0,11	+0,273	+0,30	+0,32
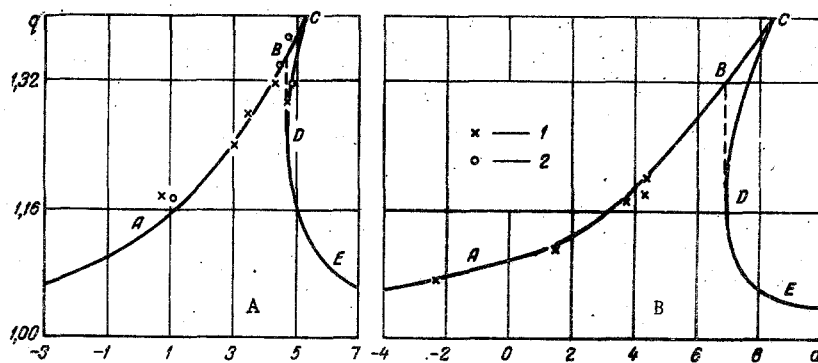
Fig. 2. Degree of separation q as a function of a for heating by: A) steam (1, first series; 2; second); B) ethanol vapor. Points from experiment, solid lines from (1) and (2)

However, this cannot be said about φ_T, which is seen from (7) to be dependent on the heating nonuniformity defined by h_1'/h_1'' and h_2'/h_2''.

The values of these quantities are determined by various factors that cannot be considered quantitatively, and this substantially hinders checking (1) and (2), since there is an element of uncertainty in calculating a. Nevertheless, if the experimental results agree satisfactorily with theory for some values of h_1'/h_1'' and h_2'/h_2'', this provides an indirect confirmation that the theory is correct.

It is clear from (4) and (6) that the temperature asymmetry is determined by the heat-transfer coefficient for given thermal conditions, and so it would appear necessary to perform a series of experiments in which the sole variable would be the heating or cooling conditions. The simplest approach is to vary the cooling-water flow, i.e., the reduced heat-transfer coefficient h_2.

The experiments were done with the column of Fig. 1, which has the following basic characteristics in mm: internal diameter of internal cylinder $d_{1i} = 34.0 \pm 0.1$, outside diameter of internal cylinder $d_{1o} = 49.472 \pm 0.003$, internal diameter of outside cylinder $d_{2i} = 49.984 \pm 0.008$, outside diameter of outside cylinder $d_{2o} = 68.0 \pm 0.1$, internal diameter of cooler jacket $d_3 = 78.0 \pm 0.1$, diameter of lower rim on internal cylinder 49.958 ± 0.002, diameter of upper rim of internal cylinder 49.942 ± 0.002, working height of column 350, pitch of final insert 30, and eccentricity of rims relative to internal cylinder 0.002.

The column was heated by saturated steam, whose pressure was measured with standard manometers with scale divisions of 0.016 atm when steam was used, or 0.06 atm when alcohol vapor was employed.

The cooling was provided by tap water, and the spiral insert intensified the heat transfer.

The water was supplied to the cooler by a centrifugal pump when the flow rate was high; the rate was measured with a flow gauge and was kept constant for each condition by manual control and monitoring from a pressure gauge.

The temperatures of the internal and outside cylinders were measured with copper—Constantan thermocouples in the usual way. Each cylinder had 6 thermocouples placed along two diametrically opposed vertical lines. The junctions were 2 mm from the working surfaces of the gap.

Butyl bromide boils at 101.6°C at atmospheric pressure, so an excess pressure of 2 atm was maintained in the working gap.

We performed three series of measurements, with steam heating in the first two and alcohol-vapor heating in the third. The heating was stopped on transferring from one series to another, and the working material was completely removed, with partial demounting of the column to check the state of the working surfaces. Reassembly inevitably led to minor changes in the working conditions. On the other hand, the
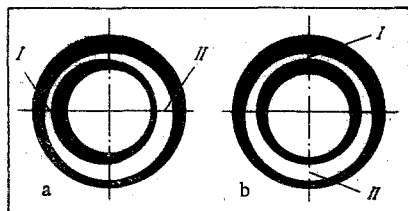

Fig. 3. Explanation of difference in thermal asymmetry with unchanged cooling conditions. The variation in thickness corresponds to that in heat transfer.

runs within a series were very closely identical. The pressure in the vapor generator was kept constant throughout a series, together with exactly reproduced geometrical dimensions in the channel, in order to keep conditions the same. The butyl bromide was of the same degree of purity in each run. Samples were taken at equal intervals in identical amounts.

The sealing of the column was checked with a GTI-2 halogen leak tester.

The length of a single run (24 h) ensured that the steady state was reached, since preliminary experiments [7] had shown that this took 12 h. After 24 h, samples were taken for isotope analysis, and then the column contents were displaced through the upper and lower samplers using the initial butyl bromide, and samples were taken to check the initial concentration. When it was clear that the replacement was complete, the next run was begun.

The samples were taken with devices of needle type, which enabled us to take reproducible small amounts of liquid into glass tubes. The sealed tubes were introduced into the vacuum lock of the MKh1303 mass spectrometer, and the bromine isotope composition was examined from the isotopic masses of $C_4^{12}H_9^1Br^{79}$ and $C_4^{12}H_9^1Br^{81}$, neglecting the possible separation of the carbon and hydrogen isotopes. The coefficient of variation in determining the bromine isotope composition was not greater than $\pm 0.5\%$.

Tables 1 and 2 give results characterizing the conditions and results.

We used the results of [3] for liquid flowing in curvilinear channels in order to calculate the heat-transfer coefficients for the cooling-water side, and there indicated that under our conditions with $34 < Re < 10300$ one can use the ordinary relationship for turbulent flow in a straight tube:

$$Nu = 0.023Re^{0.8}Pr^{0.4}. \tag{8}$$

For $Re > 10300$ we introduce the usual correction coefficient for flow curvature, i.e.,

$$Nu = 0.023\varepsilon_c Re^{0.8}Pr^{0.4}, \tag{9}$$

where $\varepsilon_c = 1 + 1.8d_e/R_c$, and R_c is the radius of curvature.

The equivalent diameter was determined from the known clear area for water passage $f = 1.85 \cdot 10^{-4}$ m^2 and the wetted perimeter $s = 8.57 \cdot 10^{-2}$ m; the values for the Prandtl number, kinematic viscosity, and thermal conductivity of the water varied only slightly in all the runs, and in the calculations we assumed $Pr^{0.4} = 2.44$, $\nu = 1.25 \cdot 10^{-6}$ m^2/sec and $\lambda = 0.575$ W/m · deg.

The reduced heat-transfer coefficient was found from the following formula incorporating the cylindrical geometry:

$$h_2 = \frac{\alpha_2 d_{2O}}{1 + \alpha_2 d_{2O} \ln (d_{2O}/d_{2i})/2\lambda_w}, \tag{10}$$

in which the thermal conductivity of the steel 45 wall was [4] $\lambda_w = 47.4$ W/m · deg.

The heat-transfer coefficient on the condensing-vapor side was determined from the formula recommended in [5]:

$$\alpha_1 = 1.01\lambda \left(\frac{g}{\nu^2}\right)^{1/3} Re^{-1/3}, \tag{11}$$

in which

$$Re = \frac{q_l L}{\pi d_{1i} r \eta}. \tag{12}$$

The heat flux appearing in (12) was calculated from

$$q_l = \pi (\Delta T)_{mea} \left[\frac{\delta}{d\lambda_m} + \frac{1}{2\lambda_w} \ln \frac{d + 2\Delta}{d - 2\Delta}\right]^{-1}, \tag{13}$$

where $(\Delta T)_{mea}$ is the temperature difference measured by the thermocouples, while $\Delta = 2$ mm is the depth of thermocouple insertion. The thermal conductivity of butyl bromide was taken from the data of [6], and for a mean column temperature of $\overline{T} \approx 330\,°K$ it was $\lambda_m = 9.72 \cdot 10^{-2}$ W/m · deg.

Tables 1 and 2 show that the heat flux varied fairly narrowly within each series, so a mean value $q_l = 5160$ W/m was used in calculations from (12) for the steam experiments, while we used $q_l = 4070$ W/m

for the ethanol vapor. A formula analogous to (10) was used for the reduced heat-transfer coefficient. The results for the steam and alcohol vapor respectively were $h_1 = 160$ W/m · deg and $h_1 = 50 \cdot 5$ W/m · deg.

The working gap δ appearing in (4)-(7) and (13) differed from the value given above because the dimensions were stated there for 20°C; the heating of the internal cylinder increased its outside diameter and reduced the gap. Then the calculated value was $\delta = 2.4 \times 10^{-4}$ m. The expansion of the internal cylinder also increased the diameter of the internal rims, which at 20°C were less than the internal diameter of the outside cylinder by $38\,\mu$m at the top and $22\,\mu$m at the bottom. Calculations show that the expansion should produce direct contact between the rims and the outside cylinder, which was confirmed by examining the rims after demounting. There were clear signs of indentation of the nickel coating from the internal surface of the outside cylinder. The calculations and observations indicated that the eccentricity due to displacement of the axes of the cylinders one relative to another was small and could be neglected, i.e., we could put $\varepsilon \approx 0$, and hence $\varphi_\varepsilon = a_\varepsilon = 0$. Then the only major factor producing temperature asymmetry in these runs was the nonuniformity in heat-transfer conditions around the perimeter; on the cold side this arose from uneven winding of the spiral insert, while on the hot side it could arise from deviation of the colum axis from vertical, which should result in variation in the thickness of the condensate film.

Parameter $\varkappa$ characterizes the parasitic convection arising from the temperature asymmetry.

It is clear from (1) and (2) that to each value of $\varkappa$ there corresponds for known y_e and b quite definite values for the degree of separation q and parameter a. The dimensionless column length y_e is defined by

$$y_e = 504 \, \frac{\alpha \eta D L}{\rho g \beta \delta^4 \overline{T}} . \tag{14}$$

Results have been given [7] on the thermal diffusion constant for bromine isotopes in butyl bromide, which indicates $\alpha = 0.030$; then $y_e \approx 0.34$ from (14). It has been shown [1] that b, which characterizes the concentration asymmetry, is

$$b = 7560 \, \frac{\eta D L \gamma}{\rho g \beta^2 \delta^4 (\Delta T)^2} , \tag{15}$$

where the coefficient for concentration expansion for isotopic mixtures is

$$\gamma = \frac{\Delta M}{M} . \tag{16}$$

Then $\gamma = 0.012$ for butyl bromide, so $b \approx 60$ in the steam heating, or $b \approx 100$ for alcohol vapor, since the temperature differences were not the same in the two cases, as Tables 1 and 2 show.

These results have been used with (1) and (2) to construct curves relating the degree of separation to a, as shown by solid lines in Fig. 2. The left-hand branch on each curve corresponds to $\varkappa > 0$, and the right-hand one to $\varkappa < 0$. The directions of the parasitic flows reverse at point C on account of the change in heat-transfer conditions. The right-hand branch is notable, for the region BCD cannot physically exist, since it corresponds to obtaining three different degrees of separation for a given temperature asymmetry. Therefore, the separation must occur along curve ABDE, and the course of the curve on part BD, shown by the broken line, is hypothetical. This indicates that it is essentially impossible to obtain the q = 1.4 implied by (14) for given process parameters. For instance, steam heating gave $q_{max} = 1.37$.

It is of interest to compare the theoretical results with experiment. We have stated above that it is unfortunately impossible to calculate h_1'/h_1'' and h_2'/h_2'', which characterize the temperature nonuniformity.

We assumed that h_1'/h_1'' and h_2'/h_2'' remained unchanged when the cooling conditions altered, and we derived the values for these giving satisfactory agreement between theory and experiment. For stem heating, $h_1'/h_1'' = 1.628$, $h_2'/h_2'' = 1.39$; while for alcohol vapor $h_1'/h_1'' = 1.26$, $h_2'/h_2'' = 1.54$, and instead of (6) and (7) we get

$$a_T = \frac{105}{8.9 + (h_1/h_2)} \left[0.628 \left(1 + \frac{40.5}{h_2} \right) - 0.485 \, \frac{h_1}{h_2} \right]$$

for stem heating and

$$a_T = \frac{140}{3.49 + (h_1/h_2)} \left[0.262 \left(1 + \frac{40.5}{h_2} \right) - 0.977 \, \frac{h_1}{h_2} \right]$$

for ethanol vapor heating.

Tables 1 and 2 give the results for σ_T, which are also shown in Fig. 2, which indicates that h_1'/h_1'' and h_2'/h_2'' closely describe the experimental results throughout the whole flow-rate range for the cooling water. The last line in the tables gives the temperature asymmetry calculated from the known σ_T. The degree of separation increases with σ_T until the effects of the temperature and concentration asymmetries balance out, as Fig. 2A shows, after which any further increase in σ_T reverses the parasitic flow, and the degree of separation begins to fall.

We see from (10) that the reduced heat-transfer coefficient at the cooled side of the column tends to the limiting value $h_2 = 310$ W/m·deg for ethanol-vapor heating with unbounded increase in the heat-transfer coefficient, and this corresponds to $\sigma_T = 5.2$ and $q = 1.23$, i.e., point B in Fig. 2B is unattainable, and no reversal in the parasitic flow will be observed.

The much lower performance of the column in ethanol-vapor heating is due in part to the smaller temperature difference at the working surfaces and to the much lower value for h_1.

The results show that h_2'/h_2'' is not the same for the two forms of heating, although the hydrodynamic conditions in the column cooler remain unchanged. Figure 3 illustrates why this is so, where I and II denote the regions for each of the two parasitic flows.

In case a, the heat transfer varies around the perimeter, but $h_2'=h_2''$; in case b, on the other hand, the region of nonuniformity is displaced, so $h_2' \neq h_2''$, although the cooling conditions remain unchanged. This displacement can be observed on replacing one heating vapor by the other, since this alters the conditions for condensate film formation, which are determined by the thickness, surface roughness, and so on.

It is clear that intermediate situations can exist, one of which is reflected in the numerical values for h_2'/h_2'' given above.

We may thus say that the heat-transfer conditions at the thermostatic surfaces can substantially influence the column performance. Figure 2A shows that a performance $q-1$ can be doubled by changing the cooling conditions. Further, even a very carefully built apparatus may not provide high performance if measures are not taken to eliminate the temperature asymmetry caused by nonuniform heat transfer around the perimeter; and finally, the optimum working conditions should be found by experiment in each particular case.

NOTATION

q	is the degree of separation;
$\varkappa$	is the sampling parameter;
y_e	is the defined by (14);
a and b	are the parameters for effects of temperature and concentration asymmetry;
σ_ε	is the parameter for temperature asymmetry due to eccentricity;
σ_T	is the parameter for temperature asymmetry due to heat transfer nonuniformity along column perimeter;

$$h_j = \alpha_j d_{ij} / \left(1 + \alpha_j d_{ij} \ln \frac{d_{io}}{d_{ii}} / 2\lambda_w \right);$$

σ_j	is the heat transfer for j-th heat transfer agent;
d_{ij}	is the diameter of i-th cylinder for j-th heat transfer agent,
d_{io}, d_{ii}	are the outside and inside diameters of i-th cylinder,
δ	is the working gap;
λ_w, λ_m	are the thermal conductivities of column walls and mixture;
d	is the diameter of separating slit;
ν, η	are the kinetic and dynamic viscosities;
r	is the heat of condensation;
ΔT	is the temperature difference between gap surfaces;
ε	is the eccentricity;
L	is the column length;
D	is the diffusion coefficient;
ρ	is the density;

β is the volume expansion coefficient;

γ is the concentration expansion coefficient;

(δT) is the temperature asymmetry.

Subscripts

1 and 2   are the hot and cold surfaces of the column.

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