

Counter-current mass transfer

V. FITT, J. R. OCKENDON and M. SHILLOR

University of Oxford, Mathematical Institute, 24-29 St Giles, Oxford OX1 3LB, U.K.

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Abstract—A simple model for a counter-current mass transfer device is analysed asymptotically in certain parameter regimes. By using some recent results about eigenfunction expansions, explicit formulae are found for the efficiency of the device in terms of numbers of theoretical plates.

1. INTRODUCTION

IN MANY industrial situations it is necessary to purify a contaminated liquid or gas and this is often achieved by mass transfer across the interface between the contaminated fluid and a different, purer fluid. Examples of the desorption of a liquid by a gas are the bubbling of a pure gas through the liquid in an agitated vessel, the spraying of liquid drops into a pure gas stream in a tower or spray chamber, and various condensing devices in which the liquid runs in thin layers down rigid plates or in pores, with its mean velocity in the opposite direction to that of a purifying gas flow. The latter counter-current devices are the motivation for this paper; our aim is to discuss the mathematical properties of a very simple model for such a device and to show how an asymptotic analysis can give an indication of the efficiency of the device.

Before describing the model, we recall a chemical engineering yardstick for the efficiency of counter-current devices. Without specifying the way in which the device operates, we suppose (Fig. 1) that a liquid stream with volume flow rate V_L enters at one end and a counter-current gas stream with flow rate V_G enters at the other. We assume the volume flow rates are constant and given throughout the device, so that if $\{C_L, C_G\}$ and $\{C'_L, C'_G\}$ are the average impurity concentrations at each end of the device, a mass balance for the impurity gives

$$\bar{C}_L V_L + \bar{C}_G V_G = \bar{C}'_G V_G + \bar{C}'_L V_L. \quad (1)$$

Now we suppose that as well as V_G and V_L we are given a desired liquid purification \bar{C}'_L and a prescribed gas inflow concentration \bar{C}'_G . Then \bar{C}_L and \bar{C}_G are linearly related by (1), and this is called the operating line in the (\bar{C}_G, \bar{C}_L) plane (Fig. 2). This line is prescribed but \bar{C}_L and

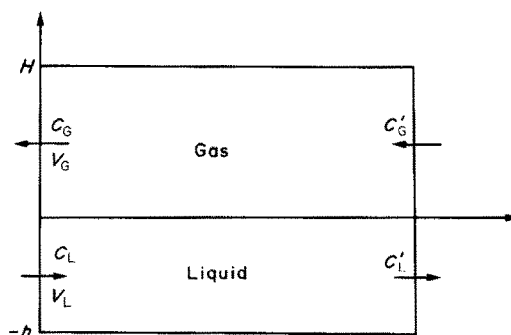


FIG. 1. Counter-current device.

so

$$\bar{C}_G = \lambda \bar{C}_L, \quad (2)$$

which is called the equilibrium line in Fig. 2. The device is called a *theoretical plate* or *tray* if the inlet concentration \bar{C}_L is given by the point A in Fig. 2, i.e. if the outlet liquid and gas concentrations are in equilibrium (see, e.g. [1], p. 217). A device which produces a value of C_L at B would be more powerful than one theoretical plate and by drawing repeated triangles, an obvious calibration is possible in terms of numbers of theoretical plates. Indeed, it is easy to show that in terms of the inlet liquid concentration \bar{C}_L , the number of theoretical plates N to which a device is equivalent is given by

$$\bar{C}_L = \frac{\{1 - (\lambda V_G/V_L)^{N+1}\} \bar{C}'_L - V_G/V_L \left\{1 - \left(\lambda \frac{V_G}{V_L}\right)^N\right\} \bar{C}'_G}{1 - \lambda V_G/V_L} \quad (3a)$$

i.e.

$$N = \begin{cases} \log \left\{ 1 + \left(\frac{V_L}{\lambda V_G} - 1 \right) \left(\frac{\bar{C}_L - \bar{C}'_L}{\bar{C}'_L - \bar{C}'_G/\lambda} \right) \right\} / \log \frac{\lambda V_G}{V_L}, & \frac{\lambda V_G}{V_L} \neq 1 \\ (\bar{C}_L - \bar{C}'_L) / (\bar{C}'_L - \bar{C}'_G/\lambda), & \frac{\lambda V_G}{V_L} = 1. \end{cases} \quad (3b)$$

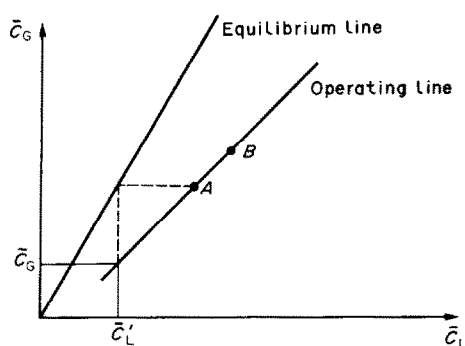
\bar{C}_G can only be found from a theoretical or experimental analysis.

We next assume that chemical equilibrium between the liquid and gas occurs when Henry's Law is obeyed,

Our aim in this paper is to assess the power, in terms of the number of theoretical plates, of the very simple two-dimensional device modelled in Fig. 1. The contaminated liquid enters at the left with uniform

NOMENCLATURE

a	a constant, (41)	x	dimensionless length variable
A	carrying capacity, $hV_L/HV_G\lambda$	y	dimensionless width variable
$A_0(x), A_1(x)$	coefficient functions, (21)	Y^\pm	infinite vectors with entries $Y_n^\pm(y)$
B	normalization coefficients, (14)	$Y_n^\pm(y)$	eigenfunctions.
b	parameter ($k_G = 1/be$)		
C	concentration of pollutant	Greek symbols	
C'	concentration of pollutant at $x = 1$	α	expansion coefficient, (36)
\bar{C}	averaged (over y) concentration of pollutant	α^*	a constant
D	diffusion coefficient	α_0	expansion coefficient, (18)
d	nondimensional channel width, $\lambda hD_G/HD_L$	α^\pm	vectors (infinite) with entries α_n^\pm
$E^\pm(x)$	infinite matrices, (42)	α_n^\pm	expansion coefficients, (18)
\bar{E}	$[E^+(1)]^{-1} \cdot E^-(1)$	β	expansion coefficient, (36)
$f(y)$	an arbitrary function of y	β_0	expansion coefficient, (18)
$g_*(y) = \begin{cases} -\frac{1}{2}y^2 + y & 0 < y < 1 \\ \frac{1}{2}y^2 + y & -1 < y < 0 \end{cases}$		β_*	vector (infinite)
$g(y)$	$w(y)g_*(y)$	γ_n	expansion coefficients
$h(x)$	interface concentration, (47)	δ_n	expansion coefficients
h	liquid channel width	δ_{mn}	Kronecker delta
H	gas channel width	ε	small parameter
I	the integral, (49)	θ	$\tan \theta = 2\sqrt{k_L k_G}/(k_G - k_L)$
k_L	nondimensional diffusivity, $D_G^2 L_* \lambda^2 / D_L H^2 V_L$	λ	Henry's constant
k_G	nondimensional diffusivity, $D_G L_* / H^2 V_G$	λ_n^\pm	eigenvalues
L_*	channel length	μ	parameter, (34)
N	number of theoretical plates	ξ	boundary layer coordinate
T, \hat{T}	matrices, (43) and (44)	η	boundary layer coordinate, (23).
V	averaged velocity		
V, V^\pm	function spaces	Subscripts	
$w(y) = \begin{cases} -1/k_G & 0 < y < 1 \\ 1/k_L & -d < y < 0 \end{cases}$	(12)	n	index for eigenfunctions and eigenvalues, 1, 2, 3, ...
		L	liquid
		G	gas.
		Superscript	
		t	transpose (of a matrix).

FIG. 2. (\bar{C}_G, \bar{C}_L) plane.

concentration \bar{C}_L of pollutant and the pure gas enters at the right.

We denote the thickness, velocity and molecular diffusion coefficients of the liquid and gas by h, H, V_L, V_G, D_L and D_G respectively. We assume uniform inviscid flow between impermeable walls so that the interface is $y = 0, 0 < x < 1$ and the walls are $y = 1$ and $y = -d = -\lambda hD_G/HD_L$ in suitable dimensionless variables. We also assume that mass transfer occurs in the liquid and

gas through convection in the x direction and diffusion in the y direction only. With a suitable normalisation of the difference between the concentrations and \bar{C}_L , the fluid equations are

$$k_L \frac{\partial^2 C_L}{\partial y^2} = \frac{\partial C_L}{\partial x}, \quad -d < y < 0, \quad k_L = \frac{D_G^2 L_* \lambda^2}{D_L H^2 V_L} \quad (4a)$$

$$k_G \frac{\partial^2 C_G}{\partial y^2} = -\frac{\partial C_G}{\partial x}, \quad 0 < y < 1, \quad k_G = \frac{D_G L_*}{H^2 V_G} \quad (4b)$$

with

$$C_L = 1 \quad \text{at} \quad x = 0, \quad -d < y < 0 \quad (5a)$$

$$C_G = 0 \quad \text{at} \quad x = 1, \quad 0 < y < 1 \quad (5b)$$

and

$$\frac{\partial C_L}{\partial y} = 0 \quad \text{at} \quad y = -d, \quad \frac{\partial C_G}{\partial y} = 0 \quad \text{at} \quad y = 1. \quad (6)$$

Continuity of mass flow at the interface gives

$$\frac{\partial C_L}{\partial y} = \frac{\partial C_G}{\partial y} \quad \text{at} \quad y = 0 \quad (7)$$

and if we finally assume that mass transfer takes place so slowly that there is chemical equilibrium at $y = 0$, our normalisation gives

$$C_L = C_G \quad \text{at } y = 0. \quad (8)$$

If the solution of (4)–(8) is known, the power of the device may be measured in terms of theoretical plates from (3b). By integrating (4) with respect to y , the operating line is

$$A(\bar{C}_L - \bar{C}_{L0}) = \bar{C}_G \quad (9)$$

where \bar{C}_{L0} is the average liquid concentration at $x = 1$ and $A = dk_G/k_L$. Hence

$$N = \begin{cases} \log \{ \bar{C}_{L0} / (1 + A(\bar{C}_{L0} - 1)) \} / \log A, & A \neq 1 \\ (1 - \bar{C}_{L0}) / \bar{C}_{L0}, & A = 1. \end{cases} \quad (10a) \quad (10b)$$

We expect the parameter A to be important; it is $hV_L/HV_G\lambda$, the ratio of the amount of impurity the liquid can carry compared to that of the gas when they are both in equilibrium everywhere, and not just at the interface $y = 0$. We will refer to A as the *carrying capacity* of the device.

Although this is a very simple model for counter-current mass transfer, it is possible to estimate values of the three parameters d , k_G and k_L which might be appropriate if (4)–(8) was regarded as a lumped parameter model for some of the practical devices mentioned earlier. From data kindly supplied by Dr. I. Parker, I.C.I., Runcorn, it is possible for k_G and k_L to be both large or small, but d is in general of $O(1)$. Even with this restriction on d , a catalogue of the possible different types of behaviour is quite complicated, but we do not expect the mathematical difficulties we will encounter to be avoided in more realistic models.

Before we begin our analysis, we note that the field equations (4) are parabolic but that in the liquid x is time-like and in the gas $-x$ is time-like. Such forward/backward diffusion equations occur in many other situations and they have been classified in [2], where their well-posedness is also discussed. One aspect of the well-posedness of our model is a uniqueness result; if the differences between two solutions which have derivatives which are square integrable over $(0 < x < 1, -d < y < 0)$ and $(0 < x < 1, 0 < y < 1)$ are denoted by C_G , $y > 0$ and C_L , $y < 0$ then, by integration,

$$\begin{aligned} & \frac{1}{2} \int_0^1 C_G^2|_{x=0} dy + \frac{1}{2} \int_{-d}^0 C_L^2|_{x=1} + k_G \\ & \times \int_0^1 \int_0^1 \left(\frac{\partial C_G}{\partial y} \right)^2 dx dy + k_L \int_{-d}^0 \int_0^1 \left(\frac{\partial C_L}{\partial y} \right)^2 dx dy = 0 \end{aligned} \quad (11)$$

so C_G and C_L are both constants, which must be zero.

2. PRINCIPAL PARAMETER REGIMES

We now consider (4)–(10) in a number of regimes for k_G, k_L . In each regime we will describe the qualitative behaviour of the solution and give an estimate for the

number of theoretical plates where analytically possible. We assume $d = O(1)$ throughout and, for convenience, we use

$$C(x, y) = \begin{cases} C_G(x, y), \\ C_L(x, y), \end{cases} \quad w(y) = \begin{cases} -1/k_G, & 0 < y < 1 \\ 1/k_L, & -d < y < 0. \end{cases} \quad (12)$$

(i) $k_G, k_L = O(1)$

Separation of variables in the form $C = Y(y) \exp(\lambda x)$ leads to the so-called indefinite Sturm–Liouville problem ([3], [4] and references therein)

$$Y'' = \lambda w(y)Y \quad -d < y < 1, \quad y \neq 0 \quad (13a)$$

with

$$Y' = 0 \quad \text{at } y = -d, 1, \quad (13b)$$

where $' = d/dy$ and Y is continuously differentiable in $(-d, 1)$. Hence

$$Y(y) = \begin{cases} B_G \cos(\sqrt{\lambda}(1-y)/k_G), & 0 < y < 1 \\ B_L \cosh(\sqrt{\lambda}(y+d)/k_L), & -d < y < 0 \end{cases} \quad (14a) \quad (14b)$$

where

$$\sqrt{k_G} \tanh(d\sqrt{\lambda/k_L}) = \sqrt{k_L} \tan(\sqrt{\lambda/k_G}). \quad (14c)$$

It is easy to show that (14c) only permits a doubly infinite set of real eigenvalues $\{\lambda_n^-, 0, \lambda_n^+\}$ where $\lambda_n^- \leq 0$, $\lambda_n^+ \geq 0$ with corresponding eigenfunctions Y_n^\pm given by (14). Clearly

$$\frac{(n-1)\pi\sqrt{k_L}}{d} < \sqrt{-\lambda_n^-} < \frac{(2n-1)\pi\sqrt{k_L}}{2d}, \quad n = 2, 3, \dots \quad (15)$$

and, if $A < 1$, $0 < \sqrt{-\lambda_1^-} < \pi\sqrt{k_G}/2$; if $A = 1$, $\lambda_1^- = 0$ and $A > 1$, λ_1^- does not exist. Similarly

$$(n-1)\pi\sqrt{k_G} < \sqrt{\lambda_n^+} < \frac{1}{2}(2n-1)\pi\sqrt{k_G}, \quad n = 2, 3, \dots \quad (16)$$

and λ_1^+ does not exist if $A < 1$ and $\lambda_1^+ = 0$ if $A = 1$. It is easy to check that in the whole interval $(-d, 1)$ the functions Y_n^\pm satisfy the ‘orthogonality’ relations

$$\int_{-d}^1 w(y) Y_m^\pm(y) Y_n^\pm(y) dy = d_{mn} \quad (17)$$

where $d_{mn} \neq 0$ only if $m = n$ and both Y ’s have the same superscript sign. With this proviso we can choose the coefficients B_G and B_L in (14) so that $d_{mn} = \pm \delta_{mn}$; where δ_{mn} is the Kronecker delta so that the left-hand side of (17) is an indefinite bilinear form.

We can formally write

$$C(x, y) = \alpha_0 + \beta_0[x + g(y)] + \sum_1^\infty \alpha_n^- e^{\lambda_n^- x} Y_n^-(y) + \sum_1^\infty \alpha_n^+ e^{\lambda_n^+ x} Y_n^+(y) \quad (18)$$

where $g(y) = w(y)(\frac{1}{2}y^2 - y)$ for $0 < y < 1$ and $g(y) = w(y)(\frac{1}{2}y^2 + dy)$ for $-d < y < 0$ and the function $x +$

$g(y)$ has been called the linear, or diffusion solution (see, e.g., [5]), and it exists only when $A = 1$, i.e. $\lambda_1^+ = \lambda_1^- = 0$. Thus we may take

$$\begin{aligned} \alpha_1 &= \beta_0 = 0 & \text{if } A > 1 \\ \alpha_1^+ &= \beta_0 = 0 & \text{if } A < 1 \\ \alpha_1^\pm &= 0 & \text{if } A = 1. \end{aligned} \quad (19)$$

It seems impossible to make further explicit analytical progress. We note that a degenerate system of four infinite matrix equations for α_n^\pm can be written down by integrating (18) with respect to y over $(-d, 1)$ at $x = 0$ and $x = 1$. This was done implicitly in [6], where numerical results are presented for a truncated version of a system of two, out of the four, matrix equations. The truncation is complicated since the forward/backward structure of the problem couples the data at $x = 0, 1$. The coefficients α_n^\pm cannot be determined independently of each other, as with conventional Sturm–Liouville problems.

The expansion (18) can be justified by a method similar to that of [7] and a similar result was proved using compact operators in [3]†. Moreover (18) converges to the unique solution of (4)–(8). Although, without computations, we can say nothing about the number of theoretical plates to which the device is equivalent in this regime, we expect the purification of the liquid to increase as the carrying capacity A decreases.

(ii) $k_G \sim k_L \gg O(1)$

With ε a small positive parameter, let $k_L = 1/\varepsilon$, $k_G = 1/b\varepsilon$ so that $A = d/b$, and we first consider the case $A < 1$. We anticipate the existence of boundary layers near $x = 0$ or $x = 1$ because it is easy to show that in general a regular perturbation in powers of ε cannot satisfy the boundary conditions (5) at both ends of the device. Indeed, writing

$$C_G \sim C_{G0} + \varepsilon C_{G1} + \varepsilon^2 C_{G2} + \cdots \quad (20a)$$

$$C_L \sim C_{L0} + \varepsilon C_{L1} + \varepsilon^2 C_{L2} + \cdots \quad (20b)$$

we find

$$C_{L0} = C_{G0} = A_0(x) \quad (21)$$

and

$$C_{L1} = A'_0(x) \left(\frac{1}{2} y^2 + dy \right) + A_1(x) \quad (22a)$$

$$C_{G1} = A'_0(x) \left(-\frac{1}{2} b y^2 + b y \right) + A_1(x) \quad (22b)$$

where (7) implies $(b-d)A'_0(x) = 0$.

This can only be satisfied to lowest order when $b = d$, but even then we need a more refined analysis to find $C_L(1, y)$.

We will return to the case $b = d$ later, but let us first

consider the case $A < 1$. Then we cannot even satisfy (5) to lowest order and boundary layers are needed near $x = 0$ or $x = 1$ or at both ends.

A boundary layer near $x = 1$ would be described by writing $x = 1 + \varepsilon\eta$ so that, to lowest order,

$$-b \frac{\partial C_G}{\partial \eta} = \frac{\partial^2 C_G}{\partial \eta^2}, \quad 0 < y < 1, \quad -\infty < \eta < 0 \quad (23a)$$

$$\frac{\partial C_L}{\partial \eta} = \frac{\partial^2 C_L}{\partial \eta^2}, \quad -d < y < 0, \quad -\infty < \eta < 0 \quad (23b)$$

with

$$C_G \sim C_L \sim A_0 \quad \text{as } \eta \rightarrow -\infty \quad (24)$$

and

$$C_G = 0 \quad \eta = 0, \quad 0 < y < 1. \quad (25)$$

and (6), (7). A trivial integration over $-d < y < 1$, $-\infty < \eta < 0$ gives

$$A_0(d-b) = \int_{-d}^0 C_L(0, y) dy$$

so that a boundary layer of this type is impossible when $d < b$, i.e. $A < 1$. We thus satisfy (5b) to the lowest order by taking $A_0 = 0$ and seek a boundary layer near $x = 0$ by writing $x = \varepsilon\xi$, which gives a system similar to (23)–(25), with $C_G \sim C_L \sim A_0 = 0$ as $\xi \rightarrow \infty$. The uniqueness of the solution to this system can be proved as in (11). A separation of variables solution $C = Y(y) \exp(\lambda\xi)$ yields eigenfunctions of the form (14a,b) where λ satisfies (14c) with $k_L = 1$, $k_G = 1/b$. With this change of notation, we may seek C in the form

$$C = \sum_{n=1}^{\infty} \gamma_n e^{\lambda_n \varepsilon \xi} Y_n^-(y), \quad -d < y < 1 \quad (26)$$

where

$$1 = \sum_{n=1}^{\infty} \gamma_n Y_n^-(y), \quad -d < y < 0. \quad (27)$$

The justification for such a representation depends on the completeness of $\{Y_n^-(y)\}_{n=1}^{\infty}$ in $(-d, 0)$ and this question of 'partial' completeness is covered by Theorem 1 of the Appendix. The fact that the Y_n^- 's are not orthogonal on $(-d, 0)$ means that again we cannot calculate the γ_n 's precisely without solving an infinite matrix equation; with $A = \frac{1}{2}$, $\lambda_1^- \simeq -1.464$, and a simple truncation gives, with $Y_1^-(y) = \cos(\sqrt{-\lambda_1^-}(y+d))$, $-d < y < 0$, that $\gamma_1 = 1.068$. It is now a simple matter to show that the leading approximation to C for $x \sim O(1)$ is

$$C \sim \gamma_1 \exp(\lambda_1^- x(\varepsilon)) Y_1^-(y) \quad (28)$$

where

$$Y_1^-(y) = \begin{cases} B \cos(\sqrt{-\lambda_1^-} d) \cosh(\sqrt{-\lambda_1^-}(y-1)), & 0 < y < 1 \\ B \cosh(\sqrt{-\lambda_1^-}) \cos(\sqrt{-\lambda_1^-}(y+d)), & -d < y < 0 \end{cases} \quad (29a)$$

$$(29b)$$

† We are grateful to Professor W. N. Everitt for this reference.

with the normalisation constant B given by (17).

Although no boundary layer such as (23) now exists near $x = 1$, a much weaker boundary layer occurs in which C_G changes from (28)–(29) to zero. After writing $x = 1 + \varepsilon\eta$ and

$$C = \exp(\lambda_1^-/\varepsilon)\tilde{C}(\eta, y)$$

$$A_1(x) = \begin{cases} \beta x + \alpha & \mu = 0 \\ \frac{2\beta}{3\mu}(\exp(-3\mu x/2) - 1) + (\alpha + \frac{1}{4}) - \frac{3\mu}{8}(1 - \exp(3\mu/2))^{-1} e^{3\mu x/2}, & \mu \neq 0, \end{cases} \quad (36)$$

we find that to lower order, \tilde{C} satisfies a system similar to (23) but with $\tilde{C} \sim \gamma_1 \exp(\lambda_1^- \eta) Y_1^-(y)$ as $\eta \rightarrow -\infty$.

Thus, as in (26), we seek

$$\tilde{C} = \gamma_1 e^{\lambda_1^- \eta} Y_1^-(y) + \sum_0^\infty \delta_n e^{\lambda_n^+ \eta} Y_n^+(y), \quad -d < y < 0. \quad (30)$$

Then

$$-\gamma_1 Y_1^-(y) = \sum_0^\infty \delta_n Y_n^+(y) \quad 0 < y < 1$$

defines the δ_n 's uniquely in terms of γ_1 , by the partial completeness result given in Theorem 2 of the Appendix. Hence

$$C_L(1, y) = e^{\lambda_1^-/\varepsilon} \left\{ \gamma_1 Y_1^-(y) + \sum_0^\infty \delta_n Y_n^+(y) \right\}, \quad -d < y < 0, \quad (31)$$

and the number of theoretical plates to which the device is equivalent is

$$N \sim \frac{-\lambda_1^-}{\varepsilon(\log d/b)} + O(1) \quad (32)$$

as $\varepsilon \rightarrow 0$. As expected the purification is good, but it is achieved over only a short length of the device, $x = O(\varepsilon)$.

A similar analysis applies when $A > 1$ with a 'strong' boundary layer near $x = 1$. In this case \bar{C}_{L0} is the average value of $1 - \sum_1^\infty \gamma_n' Y_n^+(y)$ over $(-d, 0)$ where

$$1 = \sum_1^\infty \gamma_n' Y_n^+(y), \quad 0 < y < 1.$$

Thus for $(A-1) = O(1)$, N is of $O(1)$, with $N < 1$, and the purification of the liquid is poor.

Finally when $A = 1 + \mu\varepsilon$, $\mu = O(1)$, we need to consider (20) further. We also need Theorem 3 from the Appendix. Consideration of C_{G2} and C_{L2} gives that, when $\mu < 0$,

$$\frac{1}{3}A_0''(x) - \mu A_0'(x) = 0 \quad (33)$$

so that, from (5)

$$A_0(x) = \begin{cases} 1 + (1 - e^{3\mu x/2})(e^{3\mu/2} - 1)^{-1} & \mu \neq 0 \\ 1 - x & \mu = 0. \end{cases} \quad (34)$$

No strong boundary layer of the form (23) exists near $x = 0$ or $x = 1$ but we need more terms in the expansion (20) before we can find $C_L(1, y)$. By considering C_{G2} and C_{L2} we find

$$C \sim A_0(x) + \varepsilon(A_0'(x)g_*(y) + A_1(x)) + O(\varepsilon^2) \quad (35)$$

where $g_*(y) = \frac{1}{2}y^2 + y$ for $-1 < y < 0$ and $g_*(y) = -\frac{1}{2}y^2 + y$ for $0 < y < 1$, and

and where α and β are constants. These coefficients can only be found by considering weak boundary layers at both $x = 0$ and $x = 1$. Near $x = 1$ we write $C \sim \varepsilon\tilde{C}_1(\eta, y) + \dots$ \tilde{C} satisfies a system similar to (23)–(25) but with

$$\tilde{C}_1 \sim \frac{3}{2}\mu\eta \left[\exp\left(\frac{-3\mu}{2}\right) - 1 \right]^{-1} + A_0'(1)g_*(y) + A_1(1) \quad (37)$$

as $\eta \rightarrow \infty$ for $\mu \neq 0$. Hence

$$C = \eta - A_0'(1)g_*(y) + A_1(1) + \sum_2^\infty \bar{\alpha}_n Y_n^+(y) \exp(\lambda_n^+ \eta) \quad (38)$$

where

$$A_0''(1)(\frac{1}{2}y^2 - y) + A_1(1) = \sum_2^\infty \bar{\alpha}_n Y_n^+, \quad 0 < y < 1 \quad (39)$$

defines $\bar{\alpha}_n$ and $A_1(1)$ using the completeness referred to before (33). A simple truncation gives

$$A_1(1) = 0.379 \quad \text{when} \quad \mu = 0. \quad (40)$$

So at $x = 1$

$$\bar{C}_{L0} \simeq \varepsilon[A_1(1) + \frac{1}{3}A_0'(1)] = \varepsilon\{A_1(1) + \frac{1}{2}\mu \times [\exp(-3\mu/2) - 1]^{-1}\} \equiv \varepsilon a.$$

Therefore the number of theoretical plates is

$$N = \frac{1}{\varepsilon\mu} \log \left(\frac{a}{(1 + \varepsilon\mu)a - \mu} \right). \quad (41)$$

If we let $\mu \rightarrow \infty$ in (41) together with the asymptotic expansion for λ_1^- in (14c) we find that (41) reduces to (32), to the leading order, as it should be since this is the asymptotic limit for $b \neq d$. The number of theoretical plates is of the same order as for $b \neq d$ but a more uniform mass transfer now takes place over the entire length of the device.

(iii) $k_G \sim k_L \ll 1$

In this other extreme situation, we anticipate diffusion boundary layers of thickness $O(\sqrt{k_L})$ near $y = 0$. With y rescaled to be of this thickness, we retrieve the model (4)–(8), with the exception of (6), which is a problem which has been solved when $k_L = k_G$ in [9].

Denoting $C(x, 0)$ by $h(x)$ it is easy to show that

$$\frac{\partial C_L}{\partial y} \Big|_{y=0-} = (\pi k_L)^{-1/2} \int_0^x h'(t)(x-t)^{-1/2} dt \quad (42)$$

and hence, from (2)

$$\int_0^x h'(t)(x-t)^{-1/2} dt = (k_L/k_G)^{1/2} \times \int_x^1 h'(t)(t-x)^{-1/2} dt. \quad (43)$$

When $k_L = k_G$ the formula

$$h(x) = \int_x^1 [t(1-t)]^{-3/4} dt \Big/ \int_0^1 [t(1-t)]^{-3/4} dt$$

can be obtained for the solution of (43), which is bounded at $x = 0$ and $x = 1$, by considering the integral

$$\int (t-x)^{-1/2} [t(1-t)]^{-3/4} dt$$

around a contour in the complex t plane enclosing the branch cut from $t = 0$ to $t = 1$. When $k_L \neq k_G$, we may use Carleman's method, see, e.g. [10], to show that

$$h(x) = \int_x^1 (t-x)^{-1/2} (1-t)^{-1/2} \left(\frac{1-t}{t} \right)^{\theta/2\pi} dt \\ + \int_0^1 [t(1-t)]^{-1/2} \left(\frac{1-t}{t} \right)^{\theta/2\pi} dt \quad (44)$$

where $\tan \theta = 2\sqrt{k_G k_L}/(k_G - k_L)$, $0 < \theta < \pi$, is a solution to (43). The details are given in [11], where (43) is also generalised to allow the convective velocities to be powers of $|y|$.

In order to compute the number of theoretical plates we integrate the equation over $0 < x < 1$, $-1 < y < 0$ and use (42) and find that

$$\bar{C}_{L0} = 1 + \sqrt{k_L} I \quad (45)$$

where

$$I = \frac{1}{\sqrt{\pi}} \int_0^1 \int_0^x h'(t)(x-t)^{-1/2} dt dx. \quad (46)$$

The substitution of (44) in (46), integration by parts and estimation of the resulting beta and psi functions show that if $\alpha^* < \theta < \pi - \alpha^*$, where $\alpha^* > 0$ is chosen so that $\Gamma(\alpha^*/2\pi) = O(1)$, then $I = O(1)$. Therefore it follows from (10) that

$$N = (\log k_L/k_G)^{-1} \log \frac{1 + \sqrt{k_L} I}{1 + k_G I/k_L} \\ \approx -\sqrt{k_L} I \left(\frac{k_G - k_L}{k_L \log(k_G/k_L)} \right). \quad (47)$$

In the case $k_G = k_L$ (i.e. $\theta = \pi/2$) we find that $I \simeq -0.231$ and so

$$N = 0.231 \sqrt{k_L} \quad (\theta = \pi/2).$$

As expected the small number, $N = O(\sqrt{k_L})$, of theoretical plates to which the device is equivalent reflects the poor purification.

3. DISCUSSION

We have presented approximate results for the calibration of a simple counter-current device in different parameter ranges for k_L, k_G . Our results show that the number of theoretical plates falls to zero for small k_L, k_G more slowly than the way it tends to infinity for large k_L, k_G . We have not presented an exhaustive list of regimes and in particular the case where just one of them say k_L is large is discussed in [11], where the result $N \sim O(k_L/\log k_L)$ is obtained. This large number of theoretical plates indicates good liquid purification, but a similarly large N is also obtained when $k_G \gg k_L = O(1)$, even though the liquid purification is poor. This is because the operating lines has large slope and there is good mass transfer into the gas. Thus the calibration of devices in terms of theoretical plates must be assessed in terms of whether the liquid is being purified or the gas polluted.

There is one obvious generalisation we have not mentioned here and that is to include a y -dependent convective velocity. This is relatively easy to do when the velocity is a power of $|y|$ in the case of small k_G, k_L and the details are also given in [11], but the order of magnitude of N remains as in (36).

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REFERENCES

1. R. E. Treybal, *Mass Transfer Operations*. McGraw-Hill, New York (1955).
2. M. Gevrey, Sur les équations dérivées partielles du type parabolique, *J. Math. Ser* **6**, 9 (1913) and 10, 105–137 (1914).
3. R. Beals, Partial-range completeness and existence of solutions to two-way diffusion, *J. Math. Phys.* **22**, 954–960 (1981).
4. H. G. Kaper, M. K. Kwong, C. G. Lekkerkerker and A. Zettl, Full- and half-range theory of indefinite Sturm–Liouville problems, preprint.
5. M. J. Fisch and M. D. Kruskal, Separating variables in two-way diffusion equations, *J. Math. Phys.* **21**, 740–750 (1980).
6. R. J. Nunge and W. N. Gill, Analysis of heat or mass transfer in some countercurrent flows, *Int. J. Heat Mass Transfer* **8**, 873–886 (1965).
7. G. S. S. Ludford and R. A. Robertson, Fully diffused regions, *SIAM J. Appl. Math.* **25**, 693–703 (1973).
8. R. Beals, Indefinite Sturm–Liouville problems and half-range completeness, preprint.
9. G. S. S. Ludford and S. S. Wilson, Subcharacteristic reversal, *SIAM J. Appl. Math.* **27**, 430–440 (1979).
10. G. R. Carrier, M. Krook and C. E. Pearson, *Functions of a Complex Variable*. McGraw-Hill, New York (1966).
11. V. Fitt, Mass transfer from spreading liquid films, Ph.D. thesis, Oxford University, U.K. (1984).

APPENDIX: COMPLETENESS AND PARTIAL COMPLETENESS

In [7] it is proved, by taking a Fourier transform in $-\infty < x < \infty$ and considering a suitable inversion contour, that the series (18) for arbitrary data on $x = 0$ can be written as a convolution of that data with the inverse transform of a certain Green's function. By proving that this latter inverse transform is a delta function, the systems $\{1, Y_1^-, Y_n^{\pm}\}_{n=2}^{\infty}$ and $\{1, Y_1^+, Y_n^{\pm}\}_{n=2}^{\infty}$ and $\{1, x + g(y), Y_n^{\pm}\}_{n=2}^{\infty}$ can be proved independent and complete in V , the space of all continuously differentiable functions $f(y)$, for $-d \leq y \leq 1$, such that $f'(-d) = f'(1) = 0$ and f'' is square integrable in $-d < y < 1$, for $A < 1$, $A > 1$ and $A = 1$ respectively.

Although the set $\{1, Y_1^-, Y_n^{\pm}\}_{n=2}^{\infty}$ is complete in V , the "partial completeness" needed for (27) was first conjectured in [5] and subsequently proved in [4] and [8]. Indeed, the following theorems are proved there:

1. $\{Y_n^-\}_{n=1}^{\infty}$ is complete in V^- , the space of all continuously differentiable functions $f(y)$ on $-d \leq y \leq 0$, such that $f''(y)$ is square integrable and $f'(-d) = 0$, and the set $\{1, Y_n^+\}_{n=2}^{\infty}$ is complete in V^+ [as V^- but on $0 \leq y \leq 1$ and with $f'(1) = 0$].

2. When $A > 1$, $\{Y_n^+\}_{n=1}^{\infty}$ is complete in V^+ and $\{1, Y_n^-\}_{n=2}^{\infty}$ in V^- .

3. When $A = 1$, both $\{1, Y_n^-\}_{n=1}^{\infty}$ and $\{y^2/2 + y, Y_n^-\}_{n=2}^{\infty}$ are complete in V^- and both $\{1, Y_n^+\}_{n=2}^{\infty}$, $\{-y^2/2 + y, Y_n^+\}_{n=1}^{\infty}$ are complete in V^+ .

The idea of partial completeness which is crucial to justify the eigenfunction expansion (26) also has implications for the numerical solutions discussed after (19). Indeed consider the expansion (18) in the case $A < 1$, say, so that $\alpha_1^+ = \beta_0 = 0$, (19).

For convenience we write Y_1^+ for 1 , λ_1^+ for $\lambda_0 = 0$ and denote the coefficient α_0 by α_1^+ . Let $Y^{\pm}(y)$ denote the (infinite) column vectors with entries $Y_n^{\pm}(y)$, $n = 1, 2, 3, \dots$, let α^{\pm} denote the column vectors with entries α_n^{\pm} , then we can write (18) in the form

$$C = \alpha^- \cdot E^-(x) \cdot Y^- + \alpha^+ \cdot E^+(x) \cdot Y^+$$

where $E^{\pm}(x)$ are the diagonal matrices with $\exp(\lambda_n^{\pm} x)$ in the diagonal. The set $\{Y_n^-\}_{n=1}^{\infty}$ is complete on $-d < y < 0$; therefore there exists a matrix T with constant entries such that $Y^+ = T^t \cdot Y^-$ in $-d < y < 0$, where t denotes transpose. Also there exists a vector β_* such that $1 = \beta_* \cdot Y^-$. Hence, since $C = 1$ at $x = 0$,

$$1 = \beta_* \cdot Y^- = (\alpha^- + T \cdot \alpha^+) \cdot Y^-.$$

However, the Y 's are linearly independent so

$$\alpha^- = \beta_* - T \cdot \alpha^+. \quad (A1)$$

Similarly, since the set $\{Y_n^+\}_{n=1}^{\infty}$ is complete in $0 < y < 1$, there exists a matrix \hat{T} such that $Y^- = \hat{T}^t \cdot Y^+$ in $0 < y < 1$. But $C = 0$ for $x = 0$ and $0 < y < 1$, so

$$0 = \alpha^- \cdot E^-(1) \cdot \hat{T}^t \cdot Y^+ + \alpha^+ \cdot E^+(1) \cdot Y^+.$$

Again, since the Y_n^+ are linearly independent, if we denote $\bar{E} = [E^+(1)]^{-1} \cdot E^-(1)$, then

$$\alpha^+ = -\bar{E} \cdot \hat{T}^t \cdot \alpha^-. \quad (A2)$$

Thus we have a system, (42) and (43), of two equations for the two unknowns α^{\pm} which contains all the information that we possess.

TRANSFERT MASSIQUE A CONTRE-COURANT

Résumé—Un modèle simple pour le transfert massique à contre-courant est analysé asymptotiquement dans certains régimes paramétriques. En utilisant quelques résultats récents sur les développements en fonctions propres, des formules explicites sont trouvées pour l'efficacité de l'équipement en fonction du nombre de plateaux théoriques.

STOFFAUSTAUSCH IM GEGENSTROM

Zusammenfassung—Ein einfaches Modell eines Gegenstrom-Stoffübertragungs-Apparates wird für gewisse Parameterbereiche untersucht. Unter Verwendung einiger jüngerer Ergebnisse bezüglich der Entwicklung von Eigenfunktionen ergeben sich explizite Beziehungen für den Wirkungsgrad des Apparates.

ПРОТИВОТОЧНЫЙ МАССОПЕРЕНОС

Аннотация—Асимптотическими методами исследована простая модель противоточного устройства для определенных режимных параметров. Благодаря использованию некоторых последних результатов разложения по собственным функциям, найдены в явном виде выражения для эффективности устройства через число пластин.