PSEUDO-SEDIMENTATION DIALYSIS: AN ELLIPTIC TRANSMISSION PROBLEM

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Abstract. In this paper we propose and analyze a novel membrane-based fractionation process that combines conventional countercurrent dialysis with a phenomenon of “pseudo-sedimentation”. An elliptic transmission problem is formulated to describe the steady-state concentration field of a given chemical species. Matching of spectral expansions at the transmission boundary yields an infinite system of linear algebraic equations for the eigenfunction expansion coefficients of the solution. Existence and uniqueness of the solution, framed in terms of a Fredholm alternative, are proven within a subset of the parameter space, along with regularity at the transmission boundary. The paper also develops a perturbation solution in order to interpret the theory in physical terms. Finally, numerically generated concentration profiles indicate the degree of selectivity that could be achieved with a separation apparatus based upon our physical concept.

1. Introduction.

Motivation. Countercurrent dialysis is characterized primarily by diffusive transfer of certain chemical species from a flowing solution to be purified into the dialysate, which flows in the opposite direction. The crucial element is a semipermeable membrane interposed between the flow streams, which serves two purposes: (i) to separate the flow streams (i.e., prevent cross-stream permeation of solvent) by providing a large hydraulic resistance; and (ii) to allow selective removal of small molecules from the original solution while retaining the larger species, which are unable to diffuse through the membrane [13, 15, 17, 25, 28].

It is well known [17, 25] that large differences in membrane permeability between different species may yield only modest separation efficiencies. This fact does not hamper the widespread clinical use of hemodialysis, because the toxic metabolites to be removed (e.g., urea) are vastly smaller than other constituents of blood (cells,

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FIG. 1. Schematic illustration of pseudo-sedimentation dialyzer.

proteins, etc.) But the enhancement of selectivity must be addressed in order for widespread industrial applications of dialysis to be possible. For this reason, Noda and Gryte [25] considered multistage processes, in which many dialyzers are combined with one or more concentrators into a cascade that yields higher selectivity than is possible with a single dialyzer. They noted that staging of dialyzers alone does not enhance selectivity; there must be at least one concentrator in the cascade.

Here we consider a new kind of device in which *all* solutes undergo a process analogous to sedimentation—in addition to the *species-selective* membrane diffusion that characterizes conventional dialysis. The proposed process, referred to here as “pseudo-sedimentation dialysis” (PSD), can be staged to enhance selectivity *without additional solvent-removal units*. In mathematical terms, our physical picture translates into a pair of elliptic boundary-value problems (one for each flow stream) which are coupled at the semipermeable membrane by transmission boundary conditions.

“*Pseudo-sedimentation*” and “*antisedimentation*”. Consider the system depicted in Fig. 1, in which two opposite flow streams are separated by a semipermeable membrane. The new element beyond conventional dialysis is the set of ultrafiltration membranes at the beginning and end of each stream, which serve as barriers to *all* dissolved species, while allowing the fluid to flow through—albeit with greater pressure required to overcome the additional hydraulic resistance. Here we assume that the flow velocity is uniform throughout each channel; this could be achieved in practice by filling the two channels with a coarse packing,¹ whereby the flow fields would appear uniform on the macroscopic (Darcy) scale. If the ducts contain no packing, then our assumption represents a very simple approximation (Hermans [13]) of the actual velocity profiles and could be advanced in the same spirit as the “slow-zone” model used by Chikwendu and Ojiakor [4] to analyze longitudinal dispersion in turbulent open-channel flow.

To illustrate our problem in physical terms, suppose the semipermeable membrane $A - B$ is replaced with an impenetrable surface (Fig. 2a). Then the solute concentration fields on either side will be entirely independent, and the flow streams will simply push dissolved matter toward the barrier membrane at each outlet. This

¹The permeability of the packing should be much greater than that of the semipermeable membrane.

FIG. 2. Effect upon concentration distribution of solute diffusion across boundary $A - B$: (a) Impenetrable wall; (b) Permeable membrane.

process is entirely analogous to sedimentation² in the presence of Brownian motion (Chandrasekhar [3])—hence the term “pseudo-sedimentation”. At steady state, the concentration fields on either side will be oppositely biased Boltzmann distributions, representing the balance of convective flux and diffusive back-flux necessary to give zero net transport through any cross section.

Now consider how the situation changes when solute can move across the surface $A - B$ (Fig. 2b). We assume that the flux J_y of any solute across the semipermeable membrane is proportional to the local concentration jump (Hermans [13], Fletcher [10]),³

$$J_y(x) = -\Omega[f^+(x, 0) - f^-(x, 0)], \quad (1)$$

with Ω the corresponding membrane transport coefficient. Diffusion across the membrane must therefore act to diminish spatial nonuniformities in concentration, i.e., it will counteract the “piling-up” due to the flow field—especially within a thin layer close to the membrane $A - B$. If the flow channels are narrower than this characteristic layer thickness, then the concentration profiles will be much more uniform throughout. The crucial point is that this “antisedimentation” effect is *directly related to the ease with which the solute can diffuse across the semipermeable membrane*. Thus, differences between the membrane permeabilities of various solutes, Eq. (1), will affect their concentration profiles and thereby yield a means of fractionation.

In this paper we make the preceding physical argument quantitative, by formulating and solving the boundary-value problem that governs the steady-state concentration profiles.

The physicomathematical model. Under the assumption of negligible interactions between solute particles, we formulate the mathematical description for only one chemical species. The steady-state concentration fields $f^+(x, y)$ and $f^-(x, y)$

²The cross flows employed in field-flow fractionation and SPLITT fractionation (Giddings [11]) serve the same purpose; but they occur parallel to the *short* sides of the cross section—as opposed to the case here (Fig. 1).

³Flux is counted positive in the $+y$ direction.

on either side of the semipermeable membrane are governed by standard elliptic convective-diffusion (Smoluchowski) equations together with no-flux conditions at the solid walls and barrier membranes (Chandrasekhar [3]). The two boundary value problems—denoted by $\mathcal{P}^+$ and $\mathcal{P}^-$, respectively, according to the sign of y —are coupled by transmission boundary conditions imposed along the semipermeable membrane. For simplicity we assume that the opposing streams on either side have the same speed U . Thus, we have the following dimensionless formulation:

$$\frac{\partial^2 f^\pm}{\partial x^2} + \Gamma \frac{\partial^2 f^\pm}{\partial y^2} \mp P \frac{\partial f^\pm}{\partial x} = 0, \quad 0 < x < 1, \quad 0 < \pm y < h; \quad (2)$$

$$\frac{\partial f^\pm}{\partial x}(0, y) \mp P f^\pm(0, y) = 0, \quad 0 < \pm y < h; \quad (3)$$

$$\frac{\partial f^\pm}{\partial x}(1, y) \mp P f^\pm(1, y) = 0, \quad 0 < \pm y < h; \quad (4)$$

$$\frac{\partial f^\pm}{\partial y}(x, \pm h) = 0, \quad 0 < x < 1; \quad (5)$$

$$\frac{\partial f^+}{\partial y}(x, 0) = \frac{\partial f^-}{\partial y}(x, 0) = R^{-1}[f^+(x, 0) - f^-(x, 0)], \quad 0 < x < 1. \quad (6)$$

In addition to the pointwise balance equations, an overall mass balance on any rectangle $0 \leq x \leq 1$, $y_0 \leq y \leq h$ ($-h \leq y_0 < h$) leads to the normalization condition

$$\int_0^1 f^\pm(x, y) dx = \text{const}, \quad 0 \leq \pm y \leq h. \quad (7)$$

Without loss of generality, we take the constant to be unity.

The nondimensional parameters appearing in these equations are the Péclet number P , dimensionless width (or aspect ratio) h , and dimensionless membrane transport resistance R , respectively defined as

$$P = \frac{LU}{D}, \quad h = \frac{H}{L}, \quad R = \frac{D\Gamma}{L\Omega}, \quad (8)$$

with L and H the length and width of each flow channel, respectively. Here D represents the longitudinal diffusion coefficient and Γ is the ratio of transverse to longitudinal diffusivities.⁴

REMARK 1. We note that the substitution

$$f^\pm(x, y) = e^{\pm Px/2} g^\pm(x, \eta), \quad y = \eta\sqrt{\Gamma}$$

changes the convective-diffusion equation (2) into the Helmholtz equation

$$\frac{\partial^2 g^\pm}{\partial x^2} + \frac{\partial^2 g^\pm}{\partial \eta^2} - \frac{P^2}{4} g^\pm = 0,$$

while leaving the boundary conditions (3)–(5) qualitatively the same. The constant-coefficient transmission conditions (6), however, are then transformed into the more

⁴In flow through packed beds, the longitudinal and transverse diffusivities are not generally equal; see, e.g., Koch and Brady [19].

complicated variable-coefficient form

$$e^{Px/2} \frac{\partial g^+}{\partial \eta}(x, 0) = e^{-Px/2} \frac{\partial g^-}{\partial \eta}(x, 0) = R^{-1} \sqrt{\Gamma} [e^{Px/2} g^+(x, 0) - e^{-Px/2} g^-(x, 0)].$$

The transmission conditions (6) express the fact that the diffusive flux toward (or away from) the semipermeable membrane in each channel must equal the flux across it—the latter being proportional to the transmembrane concentration difference. Although the solution itself suffers a jump along the transmission boundary (see Fig. 3 in Sec. 5), the normal derivatives on either side must match.

In the literature of elliptic equations, transmission problems arise, perhaps most frequently, in the context of acoustic or electromagnetic scattering (Helmholtz equation) in various geometries; see, e.g., Colton and Kress [6, 5], Costabel and Stephan [7, 8], Kirsch [16], Kleinman and Martin [18], Kress and Roach [20], Roach and Zhang [27], von Petersdorff [29], Wilde [30]. In the above works, separate transmission conditions are given for the solution itself and its normal derivative. Similar transmission conditions appear in connection with other elliptic operators (e.g., Carriero [2], Lishang and Jin [22]).

Mixed transmission conditions (e.g., Ladyzhenskaya and Ural'tseva [21], Meister and Speck [23]) typically relate the jump in the normal derivative to the function value along the boundary—the opposite of the case here. Conditions such as (6) occur in applications that involve membranes of finite permeability. These include dialysis (Hermans [13]) and microcirculation (Fletcher [10]). If one neglects the membrane transport resistance (the case of effectively infinite permeability), the solution must be continuous across the boundary, and the transmission conditions then appear like those mentioned above for scattering problems. In this connection we mention the paper [12] of Großmann.

Regarding the no-flux conditions (3) and (4), we note that $\partial f^\pm / \partial x$ refers to the *inward* normal derivative at $x = 0$ but to the *outward* normal derivative at $x = 1$.

2. Solution via matched spectral expansions. The solution of the pair of coupled elliptic boundary-value problems $\mathcal{P}^+$ and $\mathcal{P}^-$, Eqs. (2)–(7), is formulated here by matching eigenfunction expansions above and below the x -axis. This general approach is useful for boundary-value problems posed on irregular, rectangularly decomposable domains (Phillips [26]) as well as for situations where the differential operator and boundary conditions change across a surface—which is the case in the present analysis. In the context of a parabolic sedimentation-diffusion problem, Nitsche et al. [24] employ an analogous method for matching series solutions from adjoining time intervals.

The constant-coefficient subproblems $\mathcal{P}^+$ and $\mathcal{P}^-$ can be solved by separation of variables, yielding a common (discrete) spectrum of eigenvalues

$$\lambda_0 = 0, \quad \lambda_n = \frac{P^2}{4} + n^2 \pi^2 \quad \text{for } n = 1, 2, 3, \dots, \quad (9)$$

but different (complete) sets of eigenfunctions

$$\phi_0^\pm(x) = \left(\frac{\pm P}{e^{\pm P} - 1} \right)^{1/2} e^{\pm Px}, \quad (10)$$

$$\phi_n^\pm(x) = \left(\frac{2}{\lambda_n} \right)^{1/2} \left[n\pi \cos(n\pi x) \pm \frac{P}{2} \sin(n\pi x) \right] e^{\pm Px/2}, \quad n \geq 1, \quad (11)$$

which satisfy the orthogonality relations

$$\int_0^1 \phi_m^\pm(x) \phi_n^\pm(x) e^{\mp Px} dx = \delta_{mn}, \quad m, n \geq 0. \quad (12)$$

Subsequent arguments will utilize two kinds of relations between $\{\phi_m^+\}$ and $\{\phi_m^-\}$. Firstly, we have the equation

$$\phi_n^-(x) = (-1)^n e^{-P/2} \phi_n^+(1-x), \quad n \geq 0. \quad (13)$$

Secondly, we note the cross expansion formulas

$$\phi_n^-(x) = \sum_{m=0}^{\infty} A_{mn} \phi_m^+(x), \quad A_{mn} = \int_0^1 \phi_n^-(x) \phi_m^+(x) e^{-Px} dx, \quad (14)$$

$$\phi_n^+(x) = \sum_{m=0}^{\infty} B_{mn} \phi_m^-(x), \quad B_{mn} = \int_0^1 \phi_n^+(x) \phi_m^-(x) e^{Px} dx \quad (15)$$

($n \geq 0$). Direct computations then yield [24]

$$A_{mn} = \frac{4\pi^2 P [1 - (-1)^{m+n} e^{-P}] (\lambda_m/\lambda_n)^{1/2} mn}{[P^2 + (m+n)^2 \pi^2][P^2 + (m-n)^2 \pi^2]}, \quad m, n \geq 1, \\ A_{m0} = \left[\frac{2P}{\lambda_m (1 - e^{-P})} \right]^{1/2} [1 - (-1)^m e^{-3P/2}] \frac{8\pi m P}{9P^2 + 4m^2 \pi^2}, \quad m \geq 1, \\ A_{0n} = e^{-P/2} \delta_{0n}, \quad n \geq 0, \quad (16)$$

as well as

$$B_{mn} = (-1)^{m+n} e^P A_{mn}. \quad (17)$$

Having established these results, we can proceed to find the steady-state concentration distribution.

The respective solutions $f^+(x, y)$ and $f^-(x, y)$ of the boundary-value problems $\mathcal{P}^+$ and $\mathcal{P}^-$, satisfying Eqs. (2)–(5), are represented by the series

$$f^+(x, y) = \sum_{m=0}^{\infty} \frac{a_m}{\cosh(\gamma_m h)} \phi_m^+(x) \cosh[\gamma_m (y - h)], \quad (18)$$

$$f^-(x, y) = \sum_{m=0}^{\infty} \frac{b_m}{\cosh(\gamma_m h)} \phi_m^-(x) \cosh[\gamma_m (y + h)], \quad (19)$$

with $\gamma_m = \sqrt{\lambda_m/\Gamma}$. Here, the respective leading terms $a_0 \phi_0^+(x)$ and $b_0 \phi_0^-(x)$ represent the oppositely biased Boltzmann concentration profiles which would be observed if the semipermeable membrane were replaced with an impermeable wall (Fig. 2a);

the higher-order terms ($m = 1, 2, 3, \dots$) represent the influence of transport between the flow channels. The unknown coefficients a_m , b_m are determined from the transmission conditions (6) and the normalization condition (7). Note that

$$f^+(x, 0) = \sum_{m=0}^{\infty} a_m \phi_m^+(x), \quad (20)$$

$$f^-(x, 0) = \sum_{n=0}^{\infty} b_n \phi_n^-(x) \quad (21)$$

and

$$\frac{\partial f^+}{\partial y}(x, 0) = - \sum_{m=0}^{\infty} a_m \gamma_m \beta_m \phi_m^+(x), \quad (22)$$

$$\frac{\partial f^-}{\partial y}(x, 0) = \sum_{m=0}^{\infty} b_m \gamma_m \beta_m \phi_m^-(x), \quad (23)$$

with $\beta_m = \tanh(\gamma_m h)$. A series representation for $f^-(x, 0)$ in terms of $\{\phi_m^+(x)\}$ is obtained by combining Eqs. (14) and (21):

$$f^-(x, 0) = \sum_{m=0}^{\infty} \left[\sum_{n=0}^{\infty} A_{mn} b_n \right] \phi_m^+(x).$$

Similarly, Eqs. (15) and (20) imply that

$$f^+(x, 0) = \sum_{m=0}^{\infty} \left[\sum_{n=0}^{\infty} B_{mn} a_n \right] \phi_m^-(x).$$

These expansion formulas enable us to write the following two representations for the diffusional driving force across the semipermeable membrane (x -axis):

$$f^+(x, 0) - f^-(x, 0) = \sum_{m=0}^{\infty} \left[a_m - \sum_{n=0}^{\infty} A_{mn} b_n \right] \phi_m^+(x), \quad (24)$$

$$= \sum_{m=0}^{\infty} \left[-b_m + \sum_{n=0}^{\infty} B_{mn} a_n \right] \phi_m^-(x). \quad (25)$$

The transmission condition (6) is imposed in a weak (i.e., L^2) sense, by substituting Eqs. (22) and (24) into (6). (It should be noted that, initially, Eqs. (14), (15), (18)–(25) are interpreted in a similar manner. *Pointwise* validity is verified a posteriori from bounds on $|A_{mn}|$ and $|B_{mn}|$ and from the second part of Theorem 2, below.) This procedure leads to the following expression for each coefficient a_m in terms of the b_k :

$$a_m = \sum_{k=0}^{\infty} \frac{A_{mk}}{1 + R\gamma_m \beta_m} b_k, \quad m \geq 0. \quad (26)$$

Similarly, Eqs. (23), (25), and (6) give

$$b_k = \sum_{n=0}^{\infty} \frac{B_{kn}}{1 + R\gamma_k \beta_k} a_n, \quad k \geq 0. \quad (27)$$

These two equations can be combined to yield the following infinite system of linear algebraic equations for the eigenfunction expansion coefficients a_m :

$$a_m = \sum_{n=0}^{\infty} \left[\sum_{k=0}^{\infty} \frac{A_{mk} B_{kn}}{[1 + R\gamma_m \beta_m][1 + R\gamma_k \beta_k]} \right] a_n, \quad m \geq 0. \quad (28)$$

Because $A_{0k} B_{kn} = \delta_{0k} \delta_{0n}$ and $\gamma_0 = \beta_0 = 0$, Eq. (28) reduces to the trivial identity $a_0 = a_0$ when $m = 0$. The system (28) therefore needs to be considered only for $m \geq 1$; it can be rewritten in the form

$$a_m = \sum_{n=1}^{\infty} C_{mn} a_n + R_m a_0, \quad m \geq 1, \quad (29)$$

with

$$C_{mn} = \sum_{k=1}^{\infty} \frac{A_{mk} B_{kn}}{[1 + R\gamma_m \beta_m][1 + R\gamma_k \beta_k]}, \quad (30)$$

$$R_m = \frac{A_{m0} B_{00}}{1 + R\gamma_m \beta_m} + \sum_{k=1}^{\infty} \frac{A_{mk} B_{k0}}{[1 + R\gamma_m \beta_m][1 + R\gamma_k \beta_k]}.$$

The leading coefficient a_0 is specified independently by the normalization condition (7). It follows from

$$\int_0^1 \phi_m^+(x) dx = \left(\frac{e^P - 1}{P} \right)^{1/2} \delta_{0m}$$

that

$$a_0 = \left(\frac{P}{e^P - 1} \right)^{1/2}. \quad (31)$$

Similarly, we find

$$b_0 = \left(\frac{P}{1 - e^{-P}} \right)^{1/2}. \quad (32)$$

For future reference, we indicate our convention for defining eigenvalues λ of the linear operator $C = [C_{mn}]$. These are given by

$$C\psi = \lambda\psi \quad (33)$$

with $\psi = [\psi_m]$ the eigenvector corresponding to λ .

3. Existence, uniqueness, regularity, and symmetry. If one defines

$$E_{mn} = \frac{(-1)^n e^{P/2} A_{mn}}{1 + R\gamma_m \beta_m}, \quad S_m = \frac{e^{P/2} A_{m0}}{1 + R\gamma_m \beta_m} \quad (34)$$

then Eqs. (17) and (30) imply the relations

$$C_{mn} = \sum_{k=1}^{\infty} E_{mk} E_{kn}, \quad R_m = S_m + \sum_{k=1}^{\infty} E_{mk} S_k. \quad (35)$$

LEMMA 1. The infinite matrix operator $E = [E_{mn}]$ and vector $S = [S_m]$ are square summable, i.e.,

$$\|E\|_2^2 = \sum_{m=1}^{\infty} \sum_{n=1}^{\infty} E_{mn}^2 < 9 \left[\frac{\pi^4}{P^4} + \frac{1}{48} \right] \mathcal{E}_E^2, \quad (36)$$

$$\|S\|_2^2 = \sum_{m=1}^{\infty} S_m^2 < \frac{6}{5} \mathcal{E}_S^2 \quad (37)$$

with

$$\begin{aligned} \mathcal{E}_E(P, h, \Gamma, R) &= \frac{8P\sqrt{\Gamma} \cosh(P/2)}{\pi^3 \beta_1 R} \sqrt{1 + \frac{P^2}{4\pi^2}}, \\ \mathcal{E}_S(P, h, \Gamma, R) &= \frac{4P^{3/2} \sqrt{\Gamma} \cosh(3P/4)}{\pi^3 \beta_1 R \sqrt{\sinh(P/2)}}. \end{aligned} \quad (38)$$

Here, $\beta_1 = \tanh[h(\pi^2 + P^2/4)^{1/2} \Gamma^{-1/2}]$.

Proof. The result for $\|S\|_2$ follows directly from the fact that, by Eqs. (9), (16), and (34),

$$|S_m| \leq \mathcal{E}_S m^{-3}, \quad m \geq 1.$$

As for the proof of Eq. (36), we note the following bounds for E :

$$|E_{mn}| \leq \frac{\mathcal{E}_E m}{(m+n)^2 [(m-n)^2 + (P/\pi)^2]} \quad (39a)$$

$$\leq \begin{cases} \mathcal{E}_E 4m^{-1}(m+n)^{-2}, & |m-n| \geq m/2, \\ \mathcal{E}_E (m+n)^{-2}, & |m-n| \geq \sqrt{m}, \end{cases} \quad (39b)$$

$$\leq \begin{cases} \mathcal{E}_E m^{1/3}(m+n)^{-2}, & |m-n| \geq m^{1/3}, \\ \mathcal{E}_E (\pi/P)^2 m^{-1}, & \forall m, n \geq 1. \end{cases} \quad (39c)$$

$$\leq \begin{cases} \mathcal{E}_E 4m^{-1}(m+n)^{-2}, & |m-n| \geq m/2, \\ \mathcal{E}_E (m+n)^{-2}, & |m-n| \geq \sqrt{m}, \\ \mathcal{E}_E m^{1/3}(m+n)^{-2}, & |m-n| \geq m^{1/3}, \\ \mathcal{E}_E (\pi/P)^2 m^{-1}, & \forall m, n \geq 1. \end{cases} \quad (39d)$$

Next we split the inner sum in (36) into three parts:

$$\sum_{m=1}^{\infty} \sum_{n=1}^{\infty} = \sum_{m=1}^{\infty} \left[\sum_{n=1}^{m-[\sqrt{m}]-1} + \sum_{n=m-[\sqrt{m}]}^{m+[\sqrt{m}]} + \sum_{n=m+[\sqrt{m}]+1}^{\infty} \right]$$

utilizing estimate (39b) in the first and third sums and estimate (39d) in the second sum. Thereby we obtain

$$\begin{aligned} \|E\|_2^2 &\leq \mathcal{E}_E^2 \left\{ \sum_{m=1}^{\infty} \sum_{n=1}^{m-[\sqrt{m}]-1} (m+n)^{-4} + \sum_{m=1}^{\infty} \sum_{n=m+[\sqrt{m}]+1}^{\infty} (m+n)^{-4} \right. \\ &\quad \left. + \frac{\pi^4}{P^4} \sum_{m=1}^{\infty} \sum_{n=m-[\sqrt{m}]}^{m+[\sqrt{m}]} m^{-2} \right\} \\ &\leq \mathcal{E}_E^2 \left\{ \sum_{m=1}^{\infty} \sum_{n=1}^{\infty} (m+n)^{-4} + 3 \frac{\pi^4}{P^4} \sum_{m=1}^{\infty} m^{-3/2} \right\}. \end{aligned}$$

Inequality (36) is obtained by majorizing these series with the appropriate definite integrals. $\square$

REMARK 2. From Eq. (35) it follows as an immediate corollary that $C = [C_{mn}]$ and $R = [R_m]$ are also square summable, because

$$\|C\|_2 = \left[\sum_{m=1}^{\infty} \sum_{n=1}^{\infty} C_{mn}^2 \right]^{1/2} \leq \|E\|_2^2,$$

$$\|R\|_2 = \left[\sum_{m=1}^{\infty} R_m^2 \right]^{1/2} \leq \|S\|_2 + \|E\|_2 \|S\|_2.$$

THEOREM 2. Provided that unity is not an eigenvalue of the operator $C = [C_{mn}]$, Eq. (33):

(i) The linear system (29) has a unique square summable solution; this solution is also uniquely determined by the simpler system

$$a_m = \sum_{n=1}^{\infty} E_{mn} a_n + S_m a_0. \quad (40)$$

(ii) The expansion coefficients a_m decay according to the law

$$|a_m| \leq \mathcal{C} m^{-8/3}. \quad (41)$$

As a consequence the concentration profiles $f^+(x, 0)$ and $f^-(x, 0)$ —given by Eqs. (20) and (21), respectively—belong to the regularity class $C^{1,2/3}[0, 1]$, with $C^{m,\alpha}$ the usual Hölder spaces (Bary [1]).

(iii) The corresponding solution $\{f^+(x, y), f^-(x, y)\}$ of the boundary-value problem (2)–(7) possesses the symmetry property

$$f^-(1-x, -y) = f^+(x, y), \quad 0 < x < 1, \quad 0 < y < h. \quad (42)$$

Proof. (i) Because the respective linear operators and inhomogeneous terms are square summable, analogs of the standard Fredholm theorems for integral equations of the second kind apply to both infinite linear systems (29) and (40); see Hilbert [14], Courant and Hilbert [9]. In particular, there exists a unique square-summable solution if unity is not an eigenvalue of the infinite matrix operator. Note that square-summability of $[a_m]$ is equivalent to square-integrability of $f^+(x, 0)$ over the interval $0 < x < 1$; cf. Eq. (20).

Suppose that unity is not an eigenvalue of C . Then unity must also not be an eigenvalue of E owing to Eq. (35). Thus, each of the linear systems (29) and (40) possesses a unique solution. The two (unique) solutions are the same, since any solution of (40) also satisfies (29).

(ii) We first show that

$$|a_m| \leq \mathcal{C} m^{-\alpha} \Rightarrow |a_m| \leq \mathcal{C}' m^{-\alpha-1/2}, \quad \alpha = 0, \frac{1}{2}, 1, \frac{3}{2}, \quad (43)$$

for some constant $\mathcal{C}'$. To this end we split the summation in the inequality

$$|a_m| \leq \sum_{n=1}^{\infty} |E_{mn}| |a_n| + |S_m| |a_0| \quad (44)$$

in the same manner as was used to prove Lemma 1:

$$\begin{aligned}
 |a_m| &\leq \mathcal{C}_E \mathcal{C} \left\{ \left[\sum_{n=1}^{m-[\sqrt{m}]-1} + \sum_{n=m+[\sqrt{m}]+1}^{\infty} \right] \frac{1}{(m+n)^2 n^\alpha} + \frac{\pi^2}{P^2} \sum_{n=m-[\sqrt{m}]}^{m+[\sqrt{m}]} \frac{1}{m n^\alpha} \right\} + \frac{\mathcal{C}_S a_0}{m^3} \\
 &\leq \mathcal{C}_E \mathcal{C} \left\{ \sum_{n=1}^{\infty} \frac{1}{(m+n)^2 n^\alpha} + \frac{\pi^2}{P^2} \sum_{n=m-[\sqrt{m}]}^{m+[\sqrt{m}]} \frac{1}{m} \left(\frac{2}{m} \right)^\alpha \right\} + \frac{\mathcal{C}_S a_0}{m^3} \\
 &\leq \mathcal{C}_E \mathcal{C} \left\{ \frac{1}{m^{\alpha+1/2}} \sum_{n=1}^{\infty} \frac{1}{n^{3/2}} + 2^\alpha \frac{\pi^2}{P^2} \frac{3\sqrt{m}}{m^{\alpha+1}} \right\} + \frac{\mathcal{C}_S a_0}{m^3} \\
 &\leq \mathcal{C}_E \mathcal{C} \left\{ \sum_{n=1}^{\infty} \frac{1}{n^{3/2}} + \frac{3\pi^2}{P^2} 2^\alpha \right\} \frac{1}{m^{\alpha+1/2}} + \frac{\mathcal{C}_S a_0}{m^3} \leq \frac{\mathcal{C}'}{m^{\alpha+1/2}},
 \end{aligned}$$

where we have used the fact that $n \geq m - [\sqrt{m}] \geq m/2$ ($m \geq 2$) to write the second summation on the second line. Successive applications of the result (43) bring us from boundedness of the a_m to the statement

$$|a_m| \leq \mathcal{C} m^{-2}, \quad (45)$$

where $\mathcal{C} = \mathcal{C}(P, h, \Gamma, R)$ denotes a suitable bound.

Using inequality (45) as a starting point, it is possible to improve the exponent of m to the value $\frac{8}{3}$, as in Eq. (41). For this purpose we split the sum in Eq. (44) into five parts, each part suitable for application of the relevant bound (39a, b, d) on $|E_{mn}|$:

$$\begin{aligned}
 |a_m| &\leq \mathcal{C}_E \mathcal{C} \left\{ \sum_{n=1}^{[(m/2)^{1/3}]} \frac{4}{m(m+n)^2 n^2} + \sum_{n=[(m/2)^{1/3}]+1}^{[m/2]} \frac{4}{m(m+n)^2 n^2} \right. \\
 &\quad + \sum_{n=[m/2]+1}^{m-[m^{1/3}]-1} \frac{m^{1/3}}{(m+n)^2 n^2} \\
 &\quad \left. + \sum_{n=m-[m^{1/3}]}^{m+[m^{1/3}]} \frac{\pi^2/P^2}{m n^2} + \sum_{n=m+[m^{1/3}]+1}^{\infty} \frac{m^{1/3}}{(m+n)^2 n^2} \right\} + \frac{\mathcal{C}_S a_0}{m^3} \\
 &\leq \mathcal{C}_1 \sum_{n=1}^{[(m/2)^{1/3}]} \frac{1}{m^3} + \mathcal{C}_2 \sum_{n=[(m/2)^{1/3}]+1}^{[m/2]} \frac{1}{m^{11/3}} + \mathcal{C}_3 \sum_{n=[m/2]+1}^{m-[m^{1/3}]-1} \frac{1}{m^{11/3}} \\
 &\quad + \mathcal{C}_4 \sum_{n=m-[m^{1/3}]}^{m+[m^{1/3}]} \frac{1}{m^3} + \mathcal{C}_5 \sum_{n=m+[m^{1/3}]+1}^{\infty} \frac{1}{m^{5/3} n^2} + \frac{\mathcal{C}_6}{m^3} \\
 &\leq \mathcal{C}_1 \frac{(m/2)^{1/3}}{m^3} + \mathcal{C}_2 \frac{(m/2)}{m^{11/3}} + \mathcal{C}_3 \frac{(m/2)}{m^{11/3}} + \mathcal{C}_4 \frac{3m^{1/3}}{m^3} \\
 &\quad + \mathcal{C}_5 \frac{1}{m^{5/3} m} + \frac{\mathcal{C}_6}{m^3} \leq \frac{\mathcal{C}_7}{m^{8/3}}.
 \end{aligned}$$

(iii) To derive the symmetry property (42), we first use Eqs. (16), (17), (27), (34) to show that

$$b_0 = e^{P/2} a_0, \quad b_k = (-1)^k e^{P/2} \left[\sum_{n=1}^{\infty} E_{kn} a_n + S_k a_0 \right] \quad \text{for } k \geq 1.$$

From Eq. (40) there follows

$$b_k = (-1)^k e^{P/2} a_k$$

for all $k \geq 0$. Together with Eq. (13), this relation means that

$$b_k \phi_k^-(x) = a_k \phi_k^+(1-x),$$

from which the result (42) is demonstrated by comparison with Eqs. (18) and (19). $\square$

REMARK 3. Owing to the factors $[\cosh(\gamma_m h)]^{-1}$ in the series (18) and (19), it follows from standard arguments (see, e.g., Bary [1]) that the unique solution is an *analytic function* away from the x -axis.

REMARK 4. The symmetry property (42) conforms to physical intuition, given equal but opposite convective velocities on either side of the semipermeable membrane. Indeed, it was this physical expectation that motivated the author to look for a decomposition of the form (35).

Theorem 2 motivates us to identify regions in the (P, h, Γ, R) parameter space in which it is impossible for unity to be an eigenvalue of C . For example, when

$$R > \frac{24P\sqrt{\Gamma} \cosh(P/2)}{\pi^3 \beta_1} \left(\frac{\pi^4}{P^4} + \frac{1}{48} \right)^{1/2} \left(1 + \frac{P^2}{4\pi^2} \right)^{1/2} \quad (46)$$

we know from Eqs. (36) and (38) that $\|C\|_2 \leq \|E\|_2^2 < 1$. Therefore, no eigenvalue of C can possibly equal unity, and Theorem 2 holds. Given arbitrary values of the Péclet number P , aspect ratio h , and diffusivity ratio Γ , a sufficiently large membrane transport resistance R insures existence, uniqueness, regularity, and symmetry of the solution.

REMARK 5. Whenever $\|E\| < 1$ —as would be the case for parameter values satisfying Eq. (46)—the solution $[a_m]$ can be obtained by the process of iteration, starting from any initial vector $[a_m^{(1)}]$:

$$a_m^{(j)} = \sum_{n=1}^{\infty} E_{mn} a_n^{(j-1)} + S_m a_0 \quad (j = 2, 3, 4, \dots). \quad (47)$$

4. A perturbation formulation. On physical grounds it can be argued that—in the absence of solute transport across the x -axis (the limit as $R \rightarrow \infty$)—the concentration profiles $f^\pm(x, y)$ will simply be given by the leading terms in the expansion formulas (18) and (19),

$$f^+(x, y) = a_0 \phi_0^+(x), \quad f^-(x, y) = b_0 \phi_0^-(x), \quad (48)$$

i.e., $a_m = 0$, $b_m = 0$ for all $m \geq 1$. Thus, we are motivated to consider a regular perturbation expansion in powers of R^{-1} , namely,

$$f^\pm(x, y; P, h, \Gamma, R) = f^{[0]\pm}(x, y; P, h, \Gamma) + R^{-1} f^{[1]\pm}(x, y; P, h, \Gamma) + \dots \quad (49)$$

with $f^{[0]\pm}(x, y)$ given by Eq. (48). At each subsequent order ($i = 1, 2, \dots$), the functions $f^{[i]\pm}(x, y)$ satisfy Eqs. (2)–(5). However, the transmission condition (6) now relates the normal flux at the boundary of each channel to the diffusional driving force at the *previous* order:

$$\frac{\partial f^{[i] +}}{\partial y}(x, 0) = \frac{\partial f^{[i] -}}{\partial y}(x, 0) = f^{[i-1] +}(x, 0) - f^{[i-1] -}(x, 0), \quad i = 1, 2, 3, \dots \quad (50)$$

Finally, the normalization condition (7) implies that

$$\int_0^1 f^{[i]\pm}(x, y) dx = 0 \quad (0 \leq \pm y \leq h) \quad \text{for } i = 1, 2, 3, \dots \quad (51)$$

Given the resulting perturbation expansion of each eigenfunction expansion coefficient,⁵

$$a_m(P, h, \Gamma, R) = R^{-1} a_m^{[1]}(P, h, \Gamma) + R^{-2} a_m^{[2]}(P, h, \Gamma) + \dots, \quad (52)$$

$$b_m(P, h, \Gamma, R) = R^{-1} b_m^{[1]}(P, h, \Gamma) + R^{-2} b_m^{[2]}(P, h, \Gamma) + \dots \quad (53)$$

($m \geq 1$), we find from Eqs. (22)–(25) and (50) the recursion relations

$$a_m^{[i]} = \frac{1}{\gamma_m \beta_m} \left\{ -a_m^{[i-1]} + \sum_{n=1}^{\infty} A_{mn} b_n^{[i-1]} \right\}, \quad (54)$$

$$b_m^{[i]} = \frac{1}{\gamma_m \beta_m} \left\{ -b_m^{[i-1]} + \sum_{n=1}^{\infty} B_{mn} a_n^{[i-1]} \right\} \quad (55)$$

($i \geq 2$), where

$$a_m^{[1]} = b_0 A_{m0} / (\gamma_m \beta_m), \quad b_m^{[1]} = a_0 B_{m0} / (\gamma_m \beta_m), \quad m \geq 1. \quad (56)$$

Using Eqs. (17), (54), (55), and (56) it can be shown by mathematical induction that

$$b_m^{[i]} = (-1)^m e^{P/2} a_m^{[i]} \quad \text{for all } i = 1, 2, 3, \dots, \quad (57)$$

whereby the recursion relations for the $a_m^{[i]}$ and $b_m^{[i]}$ can be decoupled:

$$a_m^{[i]} = \frac{1}{\gamma_m \beta_m} \left\{ -a_m^{[i-1]} + \sum_{n=1}^{\infty} [(-1)^n e^{P/2} A_{mn}] a_n^{[i-1]} \right\} \quad (58)$$

($i \geq 2$), with a similar formula for the $b_m^{[i]}$.

It follows a fortiori from Eq. (56) that $a_m^{[1]} = O(m^{-3})$. Furthermore, if one assumes that $|a_m^{[i-1]}| \leq \mathcal{E} m^{-8/3}$ then arguments analogous to the proof of regularity in Theorem 2 can be applied to the recursion relation (58) to show that

$$|a_m^{[i]}| \leq \mathcal{E} \mathcal{E}' m^{-8/3} \quad (59)$$

⁵Subsequently we will omit explicit mention of the parameters P , h , and Γ .

with

$$\mathcal{E}' = \mathcal{E}'(P, h, \Gamma) \stackrel{\text{def}}{=} \frac{\sqrt{\Gamma}}{\pi \beta_1} \left\{ 1 + \left[1 + 2^{2/9} + \frac{13}{5} \frac{\pi^2}{P^2} \right] 2^{5/3} \mathcal{E}_A(P) \right\},$$

where $\mathcal{E}_A(P) = (8P/\pi^2) \cosh(P/2) \sqrt{1 + P^2/(4\pi^2)}$. (60)

Thus, we obtain the following result.

THEOREM 3. If $R > \mathcal{E}'(P, h, \Gamma)$, Eq. (60), then the perturbation series (52) is absolutely convergent for all $m \geq 1$, and $a_m = O(m^{-8/3})$.

REMARK 6. The iteration scheme (47) described in Remark 5 is *not* directly equivalent to the perturbative recursion formula (58). However, the infinite linear system (40) can be rewritten so as to give a new iteration scheme:

$$a_m^{(j)} = \frac{1}{R\gamma_m\beta_m} \left\{ -a_m^{(j-1)} + \sum_{n=1}^{\infty} [(-1)^n e^{P/2} A_{mn}] a_n^{(j-1)} \right\} + \frac{e^{P/2} A_{m0}}{R\gamma_m\beta_m} a_0,$$

which does agree, at each order, with the perturbation solution—provided that one starts with the initial vector $a_m^{(1)} = \exp(P/2) A_{m0} a_0 / (R\gamma_m\beta_m)$. This can be verified by summing both sides of Eqs. (56) and (58) over $i = 1, 2, \dots, j$, from which it follows by mathematical induction that

$$a_m^{(j)} = \sum_{i=1}^j a_m^{[i]} R^{-i} \quad \text{for all } j = 1, 2, 3, \dots$$

5. Numerical solution, applications. We consider the sensitivity of numerically generated concentration profiles to the dimensionless membrane transport resistance R . Such considerations illustrate how our mechanism of pseudo-sedimentation dialysis (PSD) could be applied to the fractionation of fine particles or macrosolutes on the basis of membrane permeability.

Concentration profiles. Numerical solutions were obtained by solving finite subsections ($M \times M$) of the infinite linear system (40) for specific parameter values. Spot checks on accuracy indicated that it was sufficient to use $M = 200, 300, 400$ for the respective cases $P = 5, 10, 20$ ($R \geq 0.05$) in order to obtain the first hundred eigenfunction expansion coefficients within an uncertainty of two in the fifth digit. These coefficients were used in a truncated version of the series representation (18). In the computations reported here, the parameter sets were selected as examples to illustrate physically interesting features. Many of these values do not satisfy the sufficient condition (46) for the applicability of Theorem 2.

Figure 3 depicts three concentration profiles $f^+(x, y)$ for $P = 10$, $h = 0.1$, and $\Gamma = 1$; these correspond to $R = 10.0, 1.0, 0.1$, respectively. The solute concentration field (a) is sharply skewed in the direction of the flow stream because transport across the semipermeable membrane $A - B$ is inhibited (Fig. 2a). But when the membrane transport resistance R is small, the distribution (c) of solute is much more uniform, approximating a situation in which the solute is effectively “antisedimented” against the pull of the flow (Fig. 2b). Figure 4 on p. 98 shows that the antisedimentation effect is confined to a thin layer near the semipermeable membrane and that it is weak even within this layer if the channel is too wide; cf. Fig. 3(c).

FIG. 3(a). $R = 10.0$. Numerically generated concentration distribution $f^+(x, y)$ for $P = 10$, $h = 0.1$, and $\Gamma = 1$.

FIG. 3(b). $R = 1.0$. Numerically generated concentration distribution $f^+(x, y)$ for $P = 10$, $h = 0.1$, and $\Gamma = 1$.

FIG. 3(c). $R = 0.1$. Numerically generated concentration distribution $f^+(x, y)$ for $P = 10$, $h = 0.1$, and $\Gamma = 1$.

FIG. 4. Concentration distribution $f^+(x, y)$ for $P = 10$, $h = 0.5$, $R = 0.1$, and $\Gamma = 1$.

Good selectivity for membrane separations (i.e., appreciable sensitivity of the solution to the membrane transport resistance R) is obtained at large Péclet number, with $h = O(P^{-1})$. We note that in practice the Péclet number could be made arbitrarily large simply by increasing the flow speed U ; cf. Eq. (8).

Selectivity for separations. In an actual experiment, the system depicted in Fig. 1 would be allowed to reach steady state, whereupon each channel (“+”, “−”) would be divided into two samples, collected separately. Owing to symmetry, Eq. (42), we need only consider the “+” channel explicitly; the “−” channel is treated in an analogous fashion. Thus, we consider the two subdomains

$$\begin{aligned}\mathcal{G}_1[\phi] &= \{(x, y): 0 < x < \phi, 0 < y < h\}, \\ \mathcal{G}_2[\phi] &= \{(x, y): \phi < x < 1, 0 < y < h\}.\end{aligned}\quad (61)$$

For the average concentration of solute in each sample we then find

$$\begin{aligned}\bar{f}_1[\phi] &\stackrel{\text{def}}{=} \frac{1}{h\phi} \int_0^\phi \int_0^h f^+(x, y) dy dx \\ &= \frac{1}{\phi} \frac{e^{P\phi} - 1}{e^P - 1} + \frac{e^{P\phi/2}}{h\phi} \sum_{m=1}^{\infty} \left[\sqrt{\frac{2}{\lambda_m}} \frac{\beta_m}{\gamma_m} \sin(m\pi\phi) \right] a_m, \\ \bar{f}_2[\phi] &\stackrel{\text{def}}{=} \frac{1}{h(1-\phi)} \int_\phi^1 \int_0^h f^+(x, y) dy dx \\ &= (1 - \phi \bar{f}_1) / (1 - \phi).\end{aligned}\quad (62)$$

These average concentrations indicate how the solute is distributed between the two samples $\mathcal{G}_1$ and $\mathcal{G}_2$.

REMARK 7. The perturbation scheme from Sec. 4 yields the asymptotic formula

$$\bar{f}_1[\phi] = \frac{1}{\phi} \frac{e^{P\phi} - 1}{e^P - 1} + [\Gamma h^{-1} \mathcal{F}(P, \phi)] R^{-1} + O(R^{-2}), \quad (63)$$

with

$$\mathcal{F}(P, \phi) = \frac{8\pi P^2 \exp[P(1+\phi)/2]}{\phi \sinh(P/2)} \sum_{m=1}^{\infty} \frac{m[1 - (-1)^m \exp(-3P/2)]}{\lambda_m^2 [9P^2 + 4m^2\pi^2]} \sin(m\pi\phi). \quad (64)$$

In order to compare the selectivity of pseudo-sedimentation dialysis with that of conventional dialysis, Fig. 5 on p. 100 shows how $\bar{f}_1$ varies with R at fixed Péclet number ($P = 5, 20$) when the original domain is divided into equal parts ($\phi = 0.5$); see also the accompanying table. A perfectly uniform concentration distribution would give $\bar{f}_1 = 1$, whereas one would find $\bar{f}_1 = 0$ if all of the solute accumulated at the downstream barrier membrane. Thus, the average concentration $\bar{f}_1$ represents the degree of antis sedimentation and serves as a measure of selectivity. The analogous quantity in the context of countercurrent dialysis is the *fractional extraction* E , which is related to the relevant membrane transport resistance R' by the equation (Noda and Gryte [25]) $E = (1 + R')^{-1}$. This relation is also plotted in Fig. 5. Thus, we see that pseudo-sedimentation dialysis can give selectivity essentially comparable to that of countercurrent dialysis.

FIG. 5. Selectivity of pseudo-sedimentation dialysis with respect to R compared with that of conventional countercurrent dialysis.

Stagewise operation. In the above discussion of pseudo-sedimentation dialysis, we have envisioned a batch separation process whereby the two fractions $\mathcal{G}_1$ and $\mathcal{G}_2$ are collected after the concentration profiles of all species have reached steady state. The process could also be carried out on a continuous basis by employing a secondary flow field perpendicular to the page in Fig. 1. This is practically feasible; for, the same approach is used in a related (but different) context in SPLITT fractionation (Giddings [11]).

A standard paradigm in the field of separations is to link many fractionation units together into a countercurrent cascade, thereby achieving much greater selectivity than is possible with a single unit. Pseudo-sedimentation dialysis is directly applicable to such a procedure, because it has the effect of splitting one incoming stream into two outgoing streams, with a fixed relationship (here, a direct proportionality) between the exit concentrations $\bar{f}_1$ and $\bar{f}_2$; see Fig. 6 and Table 1. This is not true of conventional dialysis, which works by bringing a *separate flow stream* into contact with the solution to be purified. Noda and Gryte [25] show that cascading of dialyzers alone does *not* change the selectivity. Thus, they consider how concentrators can be incorporated into the cascade to enhance separations.

Pseudo-sedimentation dialysis can be regarded as combining the operations of conventional dialysis and solvent removal in a single unit; furthermore, it embodies solute-solute selectivity with respect to *two* physicochemical parameters: Péclet number P and membrane transport resistance R . Therein lies its potential for useful applications.

FIG. 6. Single pseudo-sedimentation dialysis stage in a countercurrent cascade.

TABLE 1. Average concentration (or degree of antis sedimentation) $\bar{f}_1$ and concentration ratio $\bar{f}_2/\bar{f}_1$ as functions of membrane transport resistance R for $\phi = 0.5$ and $\Gamma = 1$.

R	$(P = 5, h = 0.2)$		$(P = 20, h = 0.05)$	
	$\bar{f}_1$	$\bar{f}_2/\bar{f}_1$	$\bar{f}_1$	$\bar{f}_2/\bar{f}_1$
0.05	0.839	1.38	0.921	1.17
0.1	0.804	1.49	0.879	1.28
0.2	0.743	1.69	0.805	1.48
0.5	0.612	2.27	0.642	2.12
1.0	0.488	3.10	0.480	3.17
2.0	0.370	4.41	0.319	5.27
5.0	0.258	6.76	0.159	11.6
10.0	0.209	8.57	0.087	22.1
20.0	0.181	10.0	0.045	43.1

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