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Moment Analysis of the Time-Dependent Transmission of a Step-Function Input of a Radioactive Gas through an Adsorber Bed

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Abstract

The time-dependent concentration of a radioactive gas at the outlet of an adsorber bed for a step change in the input concentration is analyzed by the method of moments. This moment analysis yields analytical expressions for calculating the kinetic parameters of a gas adsorbed on a porous solid in terms of observables from a time-dependent transmission curve. Transmission is the ratio of the adsorbate outlet concentration to that at the inlet. The three nonequilibrium parameters are the longitudinal diffusion coefficient, the solid-phase diffusion coefficient, and the interfacial mass-transfer coefficient. Three quantities that can be extracted in principle from an experimental transmission curve are the equilibrium transmission, the average residence (or propagation) time, and the first-moment relative to the propagation time. The propagation time for a radioactive gas is given by the time integral of one minus the transmission (expressed as a fraction of the steady-state transmission). The steady-state transmission, the propagation time, and the first-order moment are functions of the three kinetic parameters and the equilibrium adsorption capacity. The equilibrium adsorption capacity is extracted from an experimental transmission curve for a stable gaseous isotope. The three kinetic parameters can be obtained by solving the three analytical expressions simultaneously. No empirical correlations are required.

INTRODUCTION

Huang et al. (*1*) developed a mathematical model for a linear chromatographic reactor with a first-order chemical reaction. This model includes three nonequilibrium processes: 1) longitudinal diffusion, 2)

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solid-phase diffusion, and 3) gas–solid interfacial mass transfer. The decay of a radioactive isotope is a first-order reaction. For a radioactive gas, the reaction rate constant is the known radioactive decay constant λ, which is the same in both the gas and solid phases. This system of a linear chromatographic reactor with a radioactive gas is characterized by four parameters: 1) the equilibrium adsorption capacity K, 2) the resistance R_F of the film at the interface between the gas phase and the solid phase, 3) the longitudinal diffusion coefficient D_L, and 4) the solid-phase intraparticle diffusion coefficient D_S. The objective of this paper is to show how the three nonequilibrium parameters R_F, D_L, and D_s can be obtained from measurements of the time-dependent transmission of a step-function input concentration C_0 of a radioactive gas flowing through a linear chromatographic reactor. Transmission is the ratio of the adsorbate concentration $C(L,t)$ at the outlet to the concentration C_0 at the inlet of the chromatographic reactor (or adsorber bed) of length L.

The equilibrium adsorption capacity K for a stable gas can be obtained from a time-dependent transmission curve in the manner described by Huang et al. (*2*). The value for K for a radioactive isotope is assumed to be the same as that for a stable isotope. To extract the three nonequilibrium parameters (viz., R_F, D_L, and D_s), we apply the method of moments to derive expressions for the propagation time t_p^*, the first-order moment m_1, and the steady-state transmission T_{ss} as functions of the three nonequilibrium parameters. The method of moments has the advantage that an analytical expression for the transmission is not needed in order to extract the kinetic parameters. Since the three quantities (viz., t_p^*, m_1, and T_{ss}) can be obtained from an experimental time-dependent transmission curve, the three nonequilibrium parameters then can be determined by solving the three equations simultaneously.

MOMENT ANALYSIS

A typical time-dependent transmission curve for a step-change in the input concentration of a radioactive gas flowing through an adsorber bed is shown in Panel (a) of Fig. 1. The transmission function is an S-shaped curve which rises slowly from zero and reaches a steady-state value T_{ss}. Let t_p^* denote the mean residence time (or the propagation time) for the adsorbate to saturate the adsorber bed. In order for the moments of the transmission function to be finite, we define a new function $f(t)$:

$$f(t) = g(t) - T(t) \tag{1}$$

where the step-function $g(t)$ is

$$g(t) = \begin{cases} 0, & \text{for } 0 < t < t_p^* \\ T_{ss}, & \text{for } t \geqslant t_p^* \end{cases} \tag{2}$$

Graphical representations of the functions $g(t)$ and $f(t)$ are shown in Panels (b) and (c), respectively, of Fig. 1. The mean residence time for the step-function (i.e., plug-flow) transmission function $g(t)$, shown in Panel (b), is the same as that for the actual transmission function $T(t)$, shown in Panel (a). The propagation time t_p^* is the time that yields areas of equal magnitude for the two cross-hatched regions in Panel (c); that is,

$$\int_0^{t_p^*} f(t)dt = -\int_{t_p^*}^{\infty} f(t)dt \tag{3}$$

According to Eq. (3), the zeroth-order moment m_0 of the function $f(t)$ vanishes; that is,

$$m_0 \equiv \int_0^{\infty} f(t)dt = 0 \tag{4}$$

Substituting Eqs. (1) and (2) into Eq. (3), and adding $t_p^* T_{ss}$ to both sides of the resultant equation, we find that the propagation time t_p^* is related to the transmission function:

$$t_p^* = \int_0^{\infty} \left[1 - \frac{T(t)}{T_{ss}}\right] dt \tag{5}$$

For stable gases, $T_{ss} = 1$; then Eq. (5) reduces to

$$t_p = \int_0^{\infty} [1 - T(t)]dt \tag{6}$$

Equation (6) is identical to the expression for the propagation time derived previously by Huang et al. (*2*) for a stable adsorbate.

The *n*th-order moment of the function $f(t)$ with respect to the propagation time t_p^* is

$$m_n = \int_0^{\infty} (t - t_p^*)^n f(t)dt \tag{7}$$

Unlike the moments of the transmission function $T(t)$, all of the moments of the function $f(t)$ with respect to the propagation time t_p^* are finite. Now, the first-order moment of the function $f(t)$ with respect to t_p^* is

$$m_1 = \int_0^{\infty} (t - t_p^*) f(t)dt \tag{8}$$

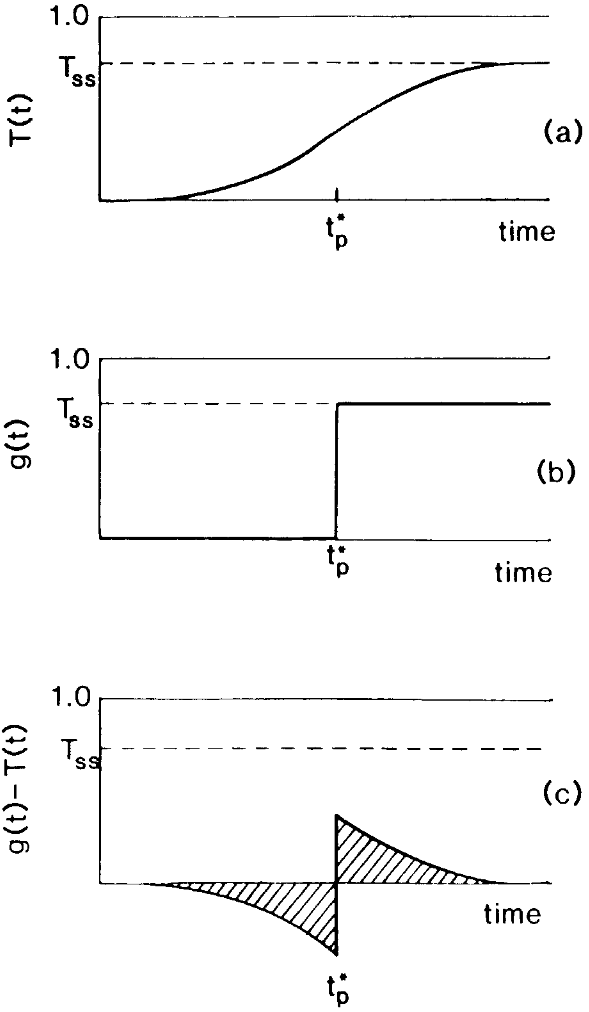

FIG. 1. The time-dependent transmission function $T(t)$ and the step-function $g(t)$ at the propagation time t_p^* for a radioactive adsorbate. Panel (c) is the difference $g(t) - T(t)$.

The vanishing of the zeroth-order moment means that the first-order moment reduces to

$$m_1 = \int_0^\infty tf(t)dt \tag{9}$$

From the theorem for differentiation of a Laplace transform [Hall et al., (*3*)], we may express the first moment:

$$m_1 = \int_0^\infty tf(t)dt = -\lim_{p\to 0} \frac{dF(p)}{dp} = -\lim_{p\to 0} \frac{d}{dp}[G(p) - T(p)] \tag{10}$$

Here p is the Laplace operator and $F(\mathrm{p})$ is the Laplace transform of the function $f(t)$. Similarly, $G(p)$ and $T(p)$ denote the Laplace transforms of the functions $g(t)$ and $T(t)$, respectively, in Eq. (2).

Now, the Laplace transform of the step function $g(t)$ (viz., Eq. 1) is

$$G(p) \equiv \mathscr{L}[g(t)] = \frac{1}{p}[T_{ss} \exp(-t_p^* p)] \tag{11}$$

The Laplace transform of the time-dependent transmission $T(t)$ is attainable from Eq. (17) of Huang et al. (*1*); that is,

$$T(p) \equiv \mathscr{L}[T(t)] = \frac{1}{p} \exp\left[\frac{uL}{2D_L} - L\sqrt{Y_A(p)}\right] \tag{12}$$

Here the function $Y_A(p)$ is

$$Y_A(p) = \frac{u^2}{4D_L^2} + \frac{p}{D_L} + \frac{\alpha Y_T(p)}{D_L} + \frac{\lambda_g}{D_L} \tag{13}$$

where

$$Y_T(p) = \frac{Y_D(p)}{1 + R_F Y_D(p)} \tag{14}$$

$$Y_D(p) = \frac{3KD_s}{R^2}(\sigma \coth \sigma - 1) \tag{15}$$

$$\sigma = R\sqrt{\frac{p + \lambda_s}{D_s}} \tag{16}$$

$$R_F = R/3k_f \tag{17}$$

$$\alpha = \frac{1-\varepsilon}{\varepsilon} \tag{18}$$

Here u is the interstitial flow velocity, R is the average radius of the spherical particles, k_f is the mass-transfer coefficient, λ_s and λ_g are the solid- and gas-phase radioactive decay constants, and ε is the void fraction of the adsorber bed.

Note also that the equilibrium transmission T_{ss}, which is the value of the transmission after a time long enough to establish equilibrium, is obtainable from Eqs. (19) and (21a) of Huang et al. (*1*); that is,

$$T_{ss} = \frac{R_{\text{out}}}{R_{\text{in}}} \equiv \frac{\int_0^\infty C(L,t)dt}{\int_0^\infty C_0 dt} \tag{19}$$

Here R_{in} (R_{out}) denotes the total amount of reactant at the column inlet (outlet). In the right-hand member of Eq. (19), the constant volumetric flow rate Q does not appear because it multiplies both the numerator and the denominator. The result of evaluating the integrals in Eq. (19) is

$$T_{ss} = \exp\left\{-\frac{uL}{2D_L}\left[\sqrt{1+\frac{4D_L}{u^2}\left(\lambda_g + \frac{\frac{3\alpha KD_s}{R^2}(\sigma_0\coth\sigma_0 - 1)}{1+\frac{3KD_sR_F}{R^2}(\sigma_0\coth\sigma_0 - 1)}\right)}-1\right]\right\}$$

$$\equiv \exp(-\gamma L) \tag{20}$$

with

$$\gamma \equiv \frac{u}{2D_L}\left[\sqrt{1+\frac{4D_L}{u^2}\left(\lambda_g + \frac{\frac{3\alpha KD_s}{R^2}(\sigma_0\coth\sigma_0 - 1)}{1+\frac{3KD_sR_F}{R^2}(\sigma_0\coth\sigma_0 - 1)}\right)}-1\right] \tag{21}$$

and

$$\sigma_0 = R(\lambda_s/D_s)^{1/2} \tag{22}$$

When Eqs. (11) and (12) are combined to evaluate m_1 in Eq. (10), the derivative can be split into two parts, H_1 and H_2:

$$m_1 = H_1 + H_2 \tag{23}$$

where H_1 includes all terms proportional to $1/p$, and H_2 includes all terms proportional to $1/p^2$. The resulting expressions for H_1 and H_2 are

$$H_1 = \lim_{p\to 0} \frac{1}{p} \left\{ \left[\exp\left(\frac{uL}{2D_L} - L\sqrt{Y_A(p)} \right) \frac{d}{dp} (-L\sqrt{Y_A(p)} \right] + [T_{ss} t_p^* \exp(-t_p^* p)] \right\} \tag{24}$$

and

$$H_2 = \lim_{p\to 0} \frac{-1}{p^2} \left\{ \left[\exp\left(\frac{uL}{2D_L} - L\sqrt{Y_A(p)} \right) \right] - [T_{ss} \exp(-t_p^* p)] \right\} \tag{25}$$

The quantity H_2 is indeterminate because each term in the numerator reduces to T_{ss} as $p \to 0$, as can be seen by rewriting the first term in the curly bracket of Eq. (25):

$$\lim_{p\to 0} \exp\left[\frac{uL}{2D_L} - L\sqrt{Y_A(p)} \right] = \lim_{p\to 0} \exp\left\{ \frac{uL}{2D_L} \left[1 - \frac{2D_L}{u} \sqrt{ \frac{u^2}{4D_L^2} + \frac{p}{D_L} + \frac{\alpha Y_T(p)}{D_L} + \frac{\lambda_g}{D_L} } \right] \right\} = T_{ss} \tag{26}$$

Applying L'Hôspital's rule to Eq. (25), we obtain

$$H_2 = \lim_{p\to 0} \frac{-1}{2p} \left\{ \left[\exp\left(\frac{uL}{2D_L} - L\sqrt{Y_A(p)} \right) \frac{d}{dp} \left(-L\sqrt{Y_A(p)} \right) \right] + T_{ss} t_p^* \exp(-t_p^* p) \right\} \equiv -\frac{H_1}{2} \tag{27}$$

Thus Eq. (23) becomes

$$m_1 = H_1/2 \tag{28}$$

Since the first-order moment m_1 is finite, the numerator of H_1 in Eq. (23) must vanish as the operator p approaches zero. By setting the numerator of Eq. (23) equal to zero, we obtain the following expression for the propagation time:

$$t_p^* = \frac{L}{u} \frac{\left\{1 + \dfrac{\dfrac{3\alpha K}{2}\left(\dfrac{\coth \sigma_0}{\sigma_0} - \operatorname{csch}^2 \sigma_0\right)}{\left[1 + \dfrac{3KD_sR_F}{R^2}(\sigma_0 \coth \sigma_0 - 1)\right]^2}\right\}}{\sqrt{1 + \dfrac{4D_L}{u^2}\left[\lambda_g + \dfrac{\dfrac{3\alpha KD_s}{R^2}(\sigma_0 \coth \sigma_0 - 1)}{1 + \dfrac{3KD_sR_F}{R^2}(\sigma_0 \coth \sigma_0 - 1)}\right]}} \tag{29}$$

Now applying L'Hôspital's rule to Eq. (24), we obtain the following expression for the first-order moment m_1:

$$m_1 = \left(\frac{LT_{ss}}{u}\right) \left\{ \frac{\left(\dfrac{\dfrac{9\alpha R_F K^2}{4}\left[\dfrac{\coth \sigma_0}{\sigma_0} - \operatorname{csch}^2 \sigma_0\right]^2}{\left[1 + \dfrac{3KD_sR_F}{R^2}(\sigma_0 \coth \sigma_0 - 1)\right]^3} - \dfrac{3\alpha KR^2}{8D_s}\left[\dfrac{2 \operatorname{csch}^2 \sigma_0 \coth \sigma_0}{\sigma_0} - \dfrac{\operatorname{csch}^2 \sigma}{\sigma_0^2} - \dfrac{\coth \sigma_0}{\sigma_0^3}\right]\left[1 + \dfrac{3KD_sR_F}{R^2}(\sigma_0 \coth \sigma_0 - 1)\right]\right)}{\left[1 + \dfrac{4D_L}{u^2}\left(\lambda_g + \dfrac{\dfrac{3\alpha KD_s}{R^2}(\sigma_0 \coth \sigma_0 - 1)}{1 + \dfrac{3KD_sR_F}{R^2}(\sigma_0 \coth \sigma_0 - 1)}\right)\right]^{1/2}} \right.$$

$$\left. + \frac{D_L}{u^2} \frac{\left[1 + \dfrac{3\alpha K}{2}\left(\dfrac{\coth \sigma_0}{\sigma_0} - \operatorname{csch}^2 \sigma_0\right)\right]^2}{\left\{1 + \dfrac{4D_L}{u^2}\left[\lambda_g + \dfrac{\dfrac{3\alpha KD_s}{R^2}(\sigma_0 \coth \sigma_0 - 1)}{1 + \dfrac{3KD_sR_F}{R^2}(\sigma_0 \coth \sigma_0 - 1)}\right]\right\}^{3/2}} \right\} \tag{30}$$

Equations (20), (29), and (30) are expressions for the equilibrium transmission T_{ss}, the propagation time t_p^*, and the first-order moment m_1 of the time-dependent transmission of a step-function input of a radioactive gas through an adsorber bed. These three quantities depend on D_s, D_L, R_F, and K. Numerical values of each can be obtained from a transmission curve. The equilibrium transmission is observed directly. The propagation time is calculated from the integral expression in Eq. (5), and the first-order moment from the integral expression in Eq. (8). For a radioactive gas the reaction rate constant is the known radioactive decay constant λ, which is same in both of the gas and solid phases; that is,

$$\lambda = \lambda_g = \lambda_s \tag{31}$$

where the subscripts g and s denote the gas and solid phases, respectively. The value of the equilibrium adsorption capacity K for a radioactive isotope is considered to be the same as that for a stable isotope. As shown in Huang et al. (*2*), the equilibrium adsorption capacity K can be obtained from an appropriate integral of the time-dependent transmission for a stable isotope. After the equilibrium adsorption capacity K is determined, then it is possible to extract the three nonequilibrium parameters (viz., D_L, D_s, and R_F) by solving Eqs. (20), (29), and (30) simultaneously.

DISCUSSION

Madey et al. (*4*) investigated the transmission of a gaseous radioactive isotope through an adsorber bed. They considered the gas-solid interfacial resistance to be negligible (i.e., $R_F = 0$) and that equilibrium in the solid phase is established instantaneously (i.e., $D_s \rightarrow \infty$). A time-dependent solution for the transmission of radioactive gases with a single effective diffusion coefficient for a step-function input was obtained in Eq. (21) of Madey et al. (*4*). Substituting this transient solution into the integral expression for t_p^* in Eq. (5) of this paper, and performing the integrations, we obtain:

$$t_p^* = \frac{\frac{L}{u}(1 + \alpha K)}{\sqrt{1 + \frac{4D_L\lambda}{u^2}(1 + \alpha K)}} \tag{32}$$

Under the same conditions (viz., $R_F = 0$ and $D_s \to \infty$), Eq. (29) reduces to Eq. (32) also.

In a study of the transmission of a stable gas through an adsorber bed, Huang et al. (*2*) derived a time-dependent expression for the transmission of a stable gas with a step-function input. Substitution of this time-dependent solution [viz., Eq. (4) of Huang et al. (*2*)] into the integral expression Eq. (6) yields an expression independent of the diffusion coefficient:

$$t_p = \frac{L}{u}(1 + \alpha K) \tag{33}$$

For the case of a stable gas where the reaction rate constants λ_s and λ_g are zero, Eq. (29) and Eq. (32) reduce to Eq. (33).

The results shown above indicate that (a) Eq. (5) is an appropriate expression for the propagation time t_p^* for a radioactive gas in terms of an integral of the transmission curve, and (b) Eq. (29) is an appropriate expression for the propagation time t_p^* of a radioactive gas in terms of three nonequilibrium parameters (viz., D_L, D_s, R_F) and the equilibrium adsorption capacity K.

As shown in Appendix A, for stable gases (i.e., $\lambda_s = \lambda_g = 0$) with negligible film resistance (i.e., $R_F = 0$), the first-order moment m_1 in Eq. (30) reduces to

$$m_1 = \frac{\alpha LKR^2}{15D_s u} + \frac{LD_L}{u^3}(1 + \alpha K)^2 \tag{34}$$

Equation (34) is identical to Eq. (13) reported by Huang et al. (*5*) in a moment analysis of breakthrough curves for stable gases. This result shows that the expression of the first-order moment (viz., Eq. 30) reduces to the proper expression for a stable gas.

APPENDIX A

For the special case of no chemical reaction ($\lambda_s = \lambda_g = 0$) and no mass-transfer resistance ($R_F = 0$), Eq. (34) for the first-order moment m_1 can be obtained from Eq. (30) by expanding the hyperbolic functions as power series in σ_0 ($= R\sqrt{\lambda_s/D_s}$) and taking the limit as λ_s and λ_g approach zero. The expansions of the hyperbolic functions are

$$\coth \sigma_0 = \frac{1}{\sigma_0} + \frac{\sigma_0}{3} - \frac{{\sigma_0}^3}{45} + \frac{2}{945}{\sigma_0}^5 - \frac{1}{4725}{\sigma_0}^7 + \cdots \tag{A.1}$$

$$\operatorname{csch} \sigma_0 = \frac{1}{\sigma_0} - \frac{\sigma_0}{6} + \frac{7}{360}{\sigma_0}^3 - \frac{31}{15120}{\sigma_0}^5 + \frac{127}{604800}{\sigma_0}^7 + \cdots \tag{A.2}$$

Retaining terms of $0(\sigma_0^4)$ or lower, we have

$$\frac{2 \operatorname{csch}^2 \sigma_0 \coth \sigma_0}{\sigma_0} - \frac{\operatorname{csch}^2 \sigma_0}{{\sigma_0}^2} - \frac{\coth \sigma_0}{{\sigma_0}^3} = \frac{-8}{45} - \frac{4}{135}{\sigma_0}^2 + \frac{8}{2025}\sigma_0^4 + \cdots \tag{A.3}$$

$$\frac{\coth \sigma_0}{\sigma_0} - \operatorname{csch}^2 \sigma_0 = \frac{2}{3} - \frac{\sigma_0^2}{45} + \frac{2}{189}{\sigma_0}^4 + \cdots \tag{A.4}$$

Now Eq. (30) for the first-order moment can be rewritten:

$$\frac{um_1}{L} = \lim_{\lambda_s \to 0} \frac{3\alpha K R^2}{8D_s}\left[\frac{8}{45} + 0({\sigma_0}^2)\right] + \frac{D_L}{u^2}\left[1 + \frac{3\alpha K}{2}\left(\frac{2}{3} - 0({\sigma_0}^2)\right)\right]^2 \tag{A.5}$$

In the limit, Eq. (A.5) reduces to Eq. (34):

$$m_1 = \frac{\alpha L K R^2}{15 D_s u} + \frac{L D_L}{u^3}(1 + \alpha K)^2 \tag{34}$$

SYMBOLS

C	gas-phase concentration (mol/cm^3)
C_0	inlet concentration (mol/cm^3)
D_L	longitudinal diffusion coefficient (cm^2/s)
D_s	solid-phase diffusion coefficient (cm^2/s)
K	adsorption capacity (dimensionless)
k_f	mass transfer coefficient (cm/s)
L	length of chromatographic column (cm)
m_1	first-order moment (s^2)

p	Laplace transform operator (s^{-1})
q	solid-phase concentration (mol/cm^3)
R	radius of particles (cm)
R_F	$R/3k_f$; film resistance (s)
t	time (s)
t_p	propagation time (s) for a stable gas
t_p^*	propagation time (s) for a radioactive gas
T	$C(z,t)/C_0$; transmission
T_{ss}	steady-state transmission
u	Interstitial flow velocity (cm/s)
Y_A	defined in Eq. (13)
Y_D	defined in Eq. (15)
Y_T	defined in Eq. (14)
α	$(1 - \varepsilon)/\varepsilon$
ε	void fraction of adsorber bed
λ_g	gas-phase reaction-rate constant (s^{-1})
λ_s	solid-phase reaction-rate constant (s^{-1})
λ	decay constant for a radioactive gas
σ	$R\sqrt{(p + \lambda_s)/D_s}$
σ_0	$R\sqrt{\lambda_s/D_s}$

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