A Unified Approach for Moments in Chromatography

Wen-Chien Lee, Stanley H. Huang,
George T. Tsao
Laboratory of Renewable Resources
Engineering
Purdue University
West Lafayette, IN 47907

Derivative of analytical expressions for moments in chromatography is a tedious work involving successive differentiations of the Laplace transformed solution of a specified model. Several expedient methods have been proposed to simplify the derivations (Lees, 1969; Suzuki, 1973). However, these methods can be applied only to axial chromatography. With the enhanced applications of radial chromatography in bioseparations (Huang et al., 1988a,b), a simple and generalized method is described in this work for moments in chromatography with any geometric configurations and kinetic models.

Mathematical Models of Chromatography

The governing equations of chromatography can be derived from material balance. For a simple system containing one single solute, the equations are given in Eqs. 1 and 2 for axial and radial chromatography respectively.

$$\epsilon \frac{\partial C}{\partial t} = -\frac{Q}{A_c}\frac{\partial C}{\partial z} + \epsilon D \frac{\partial^2 C}{\partial z^2} + R \qquad (1)$$

$$\epsilon \frac{\partial C}{\partial t} = \pm \frac{Q}{2\pi h z}\frac{\partial C}{\partial z} + \frac{\epsilon}{z}\frac{\partial}{\partial z}\left(Dz\frac{\partial C}{\partial z}\right) + R \qquad (2)$$

The "±" in Eq. 2 represents the flow directions with "+" for the inward flow in radial chromatography. Several assumptions are made in the derivation of above equations. The packing is homogeneous, the flow is plug type, and the dispersion coefficient is constant. The rate of adsorption R can be further expanded according to the kinetic models. Five models of common use are studied in this work and are listed in Table 1. With the exception of Model I, all others assume a linear isotherm with an equilibrium constant K.

After Laplace transformation and some rearrangements, Eqs. 1 and 2 can be rewritten into Eqs. 3 and 4, respectively.

$$\frac{d^2\overline{C}}{dz^2} - \frac{2\nu}{L}\frac{d\overline{C}}{dz} = f(s)\overline{C} \quad \text{where } \nu = \frac{QL}{2A_c\epsilon D} \qquad (3)$$

$$\frac{1}{z}\frac{d}{dz}\left(z\frac{d\overline{C}}{dz}\right) \pm 2\nu\frac{1}{z}\frac{d\overline{C}}{dz} = f(s)\overline{C} \quad \text{where } \nu = \frac{Q}{4\pi h\epsilon D} \qquad (4)$$

The two equations can be further reduced to a generalized operator $\overline{T}$ as shown in Eq. 5.

$$\overline{T}\overline{C} = f(s)\overline{C} \qquad (5)$$

The operator $\overline{T}$ represents the left hand side of either Eq. 3 or 4 and is determined by the configuration of the packed bed alone. $f(s)\overline{C}$ in Eq. 5 is dependent on the kinetic models and is the Laplace transformation of two terms containing time derivatives, $(1/D)\,\partial C/\partial t$ and $-R/\epsilon D$. The latter has its dependence on time derivatives expressed explicitly in Table 1. One natural constraint on $f(s)$ is that it should vanish as s approaches zero, which represents infinite time scale and the system reaches steady state. The constraint can be expressed mathematically by

$$f(s) \rightarrow 0 \quad \text{as} \quad s \rightarrow 0 \qquad (6)$$

Since $f(s)$ is finite and is continuous in the neighborhood of $s = 0$ for a reasonable physical model, it can be expanded by Taylor series as shown in Eq. 7

$$f(s) = f_1 s + f_2\frac{s^2}{2!} + f_3\frac{s^3}{3!} + \cdots + f_n\frac{s^n}{n!} + \cdots \qquad (7)$$

where f_n is the Taylor expansion coefficient. The constant term f_0 in Eq. 7 does not appear because of the constraint expressed in Eq. 6. Table 1 lists $f(s)$ and the first three coefficients of the Taylor series for each model.

Solution Methods

The definitions of the moment and central moment of n-th order are given in Eqs. 9 and 10, respectively (Abramowitz and Stegun, 1964).

$$\mu_n = \frac{\int_0^\infty t^n C\,dt}{\int_0^\infty C\,dt} = (-1)^n \lim_{s\to 0} \frac{d^n \overline{C}}{ds^n} \tag{8}$$

$$\mu'_n = \frac{\int_0^\infty (t-\mu_1)^n C\,dt}{\int_0^\infty C\,dt} = \sum_{j=0}^{n} (-1)^{n-j} \binom{n}{j} \mu_j \mu_1^{n-j} \tag{9}$$

Deriving moments in the transformed domain involves differentiations in the neighborhood of $s = 0$.

The solution of Eq. 5 can be expressed as a function of z and $f(s)$ and Eq. 10 shows the expansion of $\overline{C}$ with respect to $f(s)$.

$$\overline{C} = C_0 - C_1 f + C_2 \frac{f^2}{2!} + \cdots + (-1)^n C_n \frac{f^n}{n!} + \cdots \tag{10}$$

where $C_n(z)$ is the Taylor expansion coefficient. C_0 represents the time-independent solution and can be easily shown to be 1. In this work, the coefficients C_n in Eq. 10 are obtained through power expansion of $\overline{C}$ after the analytical solution for Eq. 5 is obtained. No tedious differentiation is involved.

Substituting Eqs. 7 and 10 into Eqs. 8 and 9 and after some rearrangements, we have the following relations

$$\mu_1 = C_1 f_1 \tag{11}$$

$$\mu'_2 = -C_1 f_2 + (C_2 - C_1^2) f_1^2 \tag{12}$$

$$\mu'_3 = C_1 f_3 - 3(C_2 - C_1^2) f_1 f_2 + (C_3 - 3C_1C_2 + 2C_1^3) f_1^3 \tag{13}$$

These relations are used for the derivation of moments. Note that f_n and C_n are mutually independent. The former is determined by the kinetic models, whereas the latter by the geometric

Table 1. Kinetic Models in Chromatography

Adsorption Models	$f(s) = \frac{s}{1!} f_1 + \frac{s^2}{2!} f_2 + \frac{s^3}{3!} f_3 + \cdots$
Model I Trace Analysis* $-R = 0$	$f(s) = \frac{s}{D}$ $f_1 = \frac{1}{D}$ $f_2 = f_3 = 0$
Model II Linearized Rate Expression** $-R = \rho_B \frac{\partial q}{\partial t}$ $\frac{\partial q}{\partial t} = \overline{k_f a}\left(C - \frac{q}{K}\right)$	$f(s) = \frac{s}{D}\left[1 + \frac{K\rho_B/\epsilon}{1 + \frac{Ks}{\overline{k_f a}}}\right]$ $f_1 = \frac{1}{D}\left(1 + \frac{\rho_B K}{\epsilon}\right)$ $-f_2 = \frac{2}{D} \frac{1}{\overline{k_f a}} \frac{\rho_B k^2}{\epsilon}$ $f_3 = \frac{6}{D} \frac{1}{(\overline{k_f a})^2} \frac{\rho_B k^3}{\epsilon}$
Model III Pore Diffusion with External Film Resistance (Spherical Particles)***† $-R = \frac{6(1-\epsilon)}{d_p} D_i \frac{\partial c}{\partial r}\Big\vert_{r=d_p/2}$ $D_i\left(\frac{\partial^2 c}{\partial r^2} + \frac{2}{r}\frac{\partial c}{\partial r}\right) - \epsilon_p \frac{\partial c}{\partial t} - \rho_p \frac{\partial q}{\partial t} = 0$ $k_f(C - c\vert_{r=d_p/2}) = D_i \frac{\partial c}{\partial r}\Big\vert_{r=d_p/2}$ $\frac{\partial q}{\partial t} = k_a\left(C - \frac{q}{K}\right)$ $\frac{\partial c}{\partial r}\Big\vert_{r=0} = 0$	$f(s) = \frac{s}{D}\left[1 + \frac{6k_f D_i}{s d_p}\left(\frac{1-\epsilon}{\epsilon}\right) \frac{d_p b(s) - 2\tanh\left(\frac{d_p b(s)}{2}\right)}{D_i d_p b(s) + (d_p k_f - 2D_i)\tanh\left(\frac{d_p b(s)}{2}\right)}\right]$ where $b(s) = \sqrt{\frac{s}{D_i}\left(\epsilon_p + \frac{\rho_p k_a}{s + \frac{k_a}{K}}\right)}$ $f_1 = \frac{1}{D}\left[1 + \left(\frac{1-\epsilon}{\epsilon}\right)(\epsilon_p + \rho_p K)\right]$ $-f_2 = \frac{2}{D}\left(\frac{1-\epsilon}{\epsilon}\right)\left[\frac{\rho_p K^2}{k_a} + \left(\frac{1}{6}\frac{d_p}{k_f} + \frac{1}{60}\frac{d_p^2}{D_i}\right)(\epsilon_p + \rho_p K)^2\right]$ $f_3 = \frac{6}{D}\left(\frac{1-\epsilon}{\epsilon}\right)\left[\frac{\rho_p K^3}{k_a^2} + \frac{2\rho_p K^2}{k_a}(\epsilon_p + \rho_p K)\left(\frac{1}{6}\frac{d_p}{k_f} + \frac{1}{60}\frac{d_p^2}{D_i}\right)\right.$ $\left. + \frac{3}{8}\frac{d_p^4}{D_i^2}(\epsilon_p + \rho_p K)^3\left(\frac{1}{945} + \frac{2}{135}\frac{D_i}{d_p k_f} + \frac{2}{27}\frac{D_i^2}{d_p^2 k_f^2}\right)\right]$

Table 1. (Continued)

Adsorption Models	$f(s) = \frac{s}{1!}f_1 + \frac{s^2}{2!}f_2 + \frac{s^3}{3!}f_3 + \cdots$
Model IV Intraparticle Diffusion with External Film Resistance (Spherical Particles)**	$f(s) = \frac{s}{D}\left[1 + \frac{36k_f D_{ip}\rho_B}{s\epsilon d_p^2} \frac{d_p b(s) - 2\tanh(d_p b(s)/2)}{D_{ip} d_p b(s) + \left(\frac{6k_f}{K} - 2D_{ip}\right)\tanh(d_p b(s)/2)}\right]$
$-R = \rho_B \frac{\partial \bar{q}}{\partial t}$	where $b(s) = \sqrt{\frac{s}{D_{ip}}}$
$\frac{\partial q}{\partial t} = \frac{1}{r^2}\frac{\partial}{\partial r}\left(D_{ip} r^2 \frac{\partial q}{\partial r}\right)$	$f_1 = \frac{1}{D}\left(1 + \frac{\rho_B K}{\epsilon}\right)$
$\bar{q} = \frac{24}{d_p^3}\int_0^{d_p/2} q r^2\, dr$	$-f_2 = \frac{2\rho_B K d_p^2}{D\epsilon D_{ip}}\left(\frac{1}{60} + \frac{D_{ip}K}{36k_f}\right)$
$D_{ip}\frac{\partial q}{\partial r}\Big\vert_{d_p/2} = \frac{6}{d_p}k_f\left(C - \frac{q}{K}\Big\vert_{r=d_p/2}\right)$	$f_3 = \frac{6\rho_B d_p^4 K}{16\, D\epsilon D_{ip}^2}\left(\frac{1}{315} + \frac{2}{135}\frac{KD_{ip}}{k_f} + \frac{1}{81}\frac{D_{ip}^2 K^2}{k_f^2}\right)$
$\frac{\partial q}{\partial r}\Big\vert_{r=0} = 0$	
Model V Dual Pore (Macropore-Micropore) Diffusion with External Film Resistance (Spherical Particles)**‡	$f(s) = \frac{s}{D}\left[1 + \frac{6k_f D_i \epsilon_p}{s d_p}\frac{1-\epsilon}{\epsilon}\frac{d_p b(s) - 2\tanh\left(\frac{d_p b(s)}{2}\right)}{D_i \epsilon_p d_p b(s) + (d_p k_f - 2D_i\epsilon_p)\tanh\left(\frac{d_p b(s)}{2}\right)}\right]$
$-R = (1-\epsilon)\left[(1-\epsilon_p)\frac{\partial \bar{\bar{q}}}{\partial t} + \epsilon_p \frac{\partial \bar{c}}{\partial t}\right]$	where $b(s) = \sqrt{\frac{s}{D_i\epsilon_p}\left[\epsilon_p + 3K(1-\epsilon_p)\left(\frac{\coth(a(s))}{a(s)} - \frac{1}{a^2(s)}\right)\right]}$
$= \frac{6(1-\epsilon)k_f}{d_p}(C - c\vert_{r=d_p/2})$	and $a(s) = R_c\sqrt{\frac{s}{D_i}}$
$\left(1-\epsilon_p\right)\frac{\partial \bar{q}}{\partial t} + \epsilon_p\frac{\partial c}{\partial t} = \frac{1}{r^2}\frac{\partial}{\partial r}\left(\epsilon_p D_i r^2 \frac{\partial c}{\partial r}\right)$	$f_1 = \frac{1}{D}\left[1 + \left(\frac{1-\epsilon}{\epsilon}\right)(\epsilon_p + (1-\epsilon_p)K)\right]$
$\frac{\partial q}{\partial t} = \frac{1}{\rho}\frac{\partial}{\partial \rho}\left(\rho^2 D_c \frac{\partial q}{\partial \rho}\right)$	$-f_2 = \frac{2}{D}\left(\frac{1-\epsilon}{\epsilon}\right)\epsilon_p\left[\frac{1}{15}\frac{K R_c^2(1-\epsilon_p)}{D_c\epsilon_p}\right.$
$\bar{q} = \frac{3}{R_c^3}\int_0^{R_c}\rho^2 q\, dp$	$\left. + \left(\frac{1}{6}\frac{d_p\epsilon_p}{k_f} + \frac{1}{60}\frac{d_p^2}{D_i}\right)\left(1 + \frac{1-\epsilon_p}{\epsilon_p}K\right)^2\right]$
$\bar{c} = \frac{24}{d_p^3}\int_0^{d_p/2} r^2 \bar{c}\, dr$	$f_3 = \frac{6}{D}\left(\frac{1-\epsilon}{\epsilon}\right)\left[\frac{2}{315}\frac{(1-\epsilon_p)KR_c^4}{D_c^2}\right.$
$\bar{\bar{q}} = \frac{24}{3}\int_0^{d_p/2} r^2 \bar{q}\, dr$	$+ \frac{2}{15}(1-\epsilon_p)\frac{KR_c^2}{D_c}\left(\frac{1}{6}\frac{d_p\epsilon_p}{k_f} + \frac{1}{60}\frac{d_p^2}{D_i}\right)\left(1 + \frac{1-\epsilon_p}{\epsilon_p}K\right)$
$\frac{\partial q}{\partial \rho} = 0, \quad \rho = 0$	$\left. + \frac{3}{8}\frac{d_p^4\epsilon_p}{D_i^2}\left(1 + \frac{1-\epsilon_p}{\epsilon_p}K\right)^3\left(\frac{1}{945} + \frac{2}{135}\frac{D_i\epsilon_p}{d_p k_f} + \frac{2}{27}\frac{D_i^2\epsilon_p^2}{k_f^2 d_p^2}\right)\right]$
$q = Kc, \quad \rho = R_c$	
$\frac{\partial c}{\partial r} = 0, \quad r = 0$	

*Wen and Fan (1975)
**Ruthven (1984)
***Arnold et al. (1985)
†McCoy (1986)
‡Weber (1984)

configuration of the packed bed and boundary conditions specified.

Examples

Three types of boundary conditions are studied in this work for axial chromatography. The first one is the semiinfinite boundary conditions, which specify inlet concentrations and impose a constraint of finite concentration as the packed bed is extended to infinity. The second is the Danckwerts-type (Danckwerts, 1953), and the third is the Kucera-type (Kucera, 1965) which specifies the sample injection through the initial conditions rather than through the boundary conditions. The solutions for geometric contributions and the literature available for comparison are listed in Table 2. All the coefficients C_n are evaluated at the outlet of the packing. In all the cases studied, the results of this work are in agreement with those in literature.

Table 2. Solutions for Geometric Contributions in Chromatography

	Axial Chromatography	Radical Chromatography (Outward Flow)	Radial Chromatography (Inward Flow)
Danckwerts Boundary Conditions	$\bar{C}(z=L) = \dfrac{\left(1-\frac{\lambda_-}{\lambda_+}\right)e^{\lambda_- L}}{1-\frac{\lambda_-}{\lambda_+}e^{(\lambda_- -\lambda_+)L}-\frac{L\lambda_-}{2\nu}[1-e^{(\lambda_- -\lambda_+)L}]}$ where $\lambda_\pm = \frac{\nu}{L} \pm \sqrt{\frac{\nu^2}{L^2}+f(s)}$	$\bar{C}(z=R_1) = \left(\frac{R_1}{R_0}\right)^\nu \left[K_\nu(\sqrt{f}R_1) + \frac{K_{\nu-1}(\sqrt{f}R_1)}{I_{\nu-1}(\sqrt{f}R_1)} I_\nu(\sqrt{f}R_1)\right] \Big/ \left\{K_\nu(\sqrt{f}R_0) + \frac{K_{\nu-1}(\sqrt{f}R_1)}{I_{\nu-1}(\sqrt{f}R_1)} I_\nu(\sqrt{f}R_0) - \frac{\sqrt{f}R_0}{2\nu}\left[\frac{I_{\nu-1}(\sqrt{f}R_0)}{I_{\nu-1}(\sqrt{f}R_1)}K_{\nu-1}(\sqrt{f}R_1) - K_{\nu-1}(\sqrt{f}R_0)\right]\right\}$	$\bar{C}(z=R_0) = \left(\frac{R_1}{R_0}\right)^\nu \left[K_\nu(\sqrt{f}R_0) + \frac{K_{\nu+1}(\sqrt{f}R_0)}{I_{\nu+1}(\sqrt{f}R_0)} I_\nu(\sqrt{f}R_0)\right] \Big/ \left\{K_\nu(\sqrt{f}R_1) + \frac{I_\nu(\sqrt{f}R_1)}{I_{\nu+1}(\sqrt{f}R_0)} K_{\nu+1}(\sqrt{f}R_0) + \frac{\sqrt{f}R_1}{2\nu}\left[\frac{I_{\nu+1}(\sqrt{f}R_1)}{I_{\nu+1}(\sqrt{f}R_0)}K_{\nu+1}(\sqrt{f}R_0) - K_{\nu+1}(\sqrt{f}R_1)\right]\right\}$
C_1	$\frac{L^2}{2\nu}$	$\frac{R_1^2-R_0^2}{4\nu}$	$\frac{R_1^2-R_0^2}{4\nu}$
$C_2 - C_1^2$	$\frac{L^4}{4\nu^3} - \frac{L^4}{8\nu^4} + \frac{L^4}{8\nu^4}e^{-2\nu}$	$\frac{R_1^4}{16\nu^2(\nu+1)} - \frac{R_0^4}{16\nu^2(\nu-1)}$	$\frac{R_1^4}{16\nu^2(\nu+1)} - \frac{R_0^4}{16\nu^2(\nu-1)}$
$C_3 - 3C_1C_2 + 2C_1^3$	$\frac{3L^6}{8\nu^5} + \frac{3L^6}{8\nu^5}e^{-2\nu} - \frac{3L^6}{8\nu^6} + \frac{3L^6}{8\nu^6}e^{-2\nu}$	$\frac{R_1^6}{16\nu^3(\nu+1)(\nu+2)} - \frac{R_0^6}{16\nu^3(\nu-1)(\nu-2)}$	$\frac{R_1^6}{16\nu^3(\nu+1)(\nu+2)} - \frac{R_0^6}{16\nu^3(\nu-1)(\nu-2)}$
Semiinfinite Boundary Conditions	$\bar{C}(z=L) = e^{\lambda_- L}$	$\bar{C}(z=R_1) = \left(\frac{R_1}{R_0}\right)^\nu \frac{K_\nu(\sqrt{f}R_1)}{K_\nu(\sqrt{f}R_0)}$	$\bar{C}(z=R_0) = \left(\frac{R_1}{R_0}\right)^\nu \frac{I_\nu(\sqrt{f}R_0)}{I_\nu(\sqrt{f}R_1)}$
C_1	$\frac{L^2}{2\nu}$	$\frac{R_1^2-R_0^2}{4(\nu-1)}$	$\frac{R_1^2-R_0^2}{4(\nu+1)}$
$C_2 - C_1^2$	$\frac{L^4}{4\nu^3}$	$\frac{R_1^4-R_0^4}{16(\nu-1)^2(\nu-2)}$	$\frac{R_1^4-R_0^4}{16(\nu+1)^2(\nu+2)}$
$C_3 - 3C_1C_2 + 2C_1^3$	$\frac{3L^6}{8\nu^5}$	$\frac{R_1^6-R_0^6}{16(\nu-1)^3(\nu-2)(\nu-3)}$	$\frac{R_1^6-R_0^6}{16(\nu+1)^3(\nu+2)(\nu+3)}$
Kucera Type	$\bar{C}(z=L) = \frac{\nu}{L}\dfrac{e^{\lambda_- L}}{\sqrt{\frac{\nu^2}{L^2}+f(s)}}$		
C_1	$\frac{L^2}{2\nu} + \frac{L^2}{2\nu^2}$		
$C_2 - C_1^2$	$\frac{L^4}{4\nu^3} + \frac{L^4}{2\nu^4}$		
$C_3 - 3C_1C_2 + 2C_1^3$	$\frac{3L^6}{8\nu^5} + \frac{L^6}{\nu^6}$		

Literature Available for Comparison

Axial Chromatography: Arnold et al. (1985), Furusawa et al. (1976), Goto and Goto (1976), Goto et al. (1983), Grubner (1984), Grubner et al. (1967), Kucera (1965), van der Laan (1958), Wen and Fan (1975)

Radial Chromatography: Huang et al. (1988a)

The method is applied to radial chromatography and the results are also listed in Table 2. Only semi-infinite and Danckwerts boundary conditions are studied. Notice the difficulties one may encounter if one tries to apply Eq. 8 to derive the moments. By the method described in this work, the complexity of derivation has been reduced to a manageable degree.

Notation

A_c = column cross section area, m^2
c = concentration of solute in pore, kg/m^3
C = concentration of solute in bulk flow phase, kg/m^3
$\bar{C}$ = Laplace transformation of C/Q_0
d_p = diameter of spherical particle, m
D = dispersion coefficient, m^2/s
D_c = pore diffusivity of solute in micropore, m^2/s
D_i = pore diffusivity of solute in macropore, m^2/s
D_{ip} = effective intraparticle diffusivity of solute, m^2/s
K = equilibrium constant, m^3/kg
k_a = adsorption rate constant, m^3/kg · s
k_f = fluid film mass transfer coefficient of solute, m/s
$\overline{k_f a}$ = average mass transfer coefficient of solute, m^3/kg · s
L = length of the axial chromatography, m

q = sorbate concentration, kg/kg particle
Q = volumetric flow rate, m^3/s
Q_0 = amount of solute injected, kg
r = radial distance in particle, m
R = adsorption rate in the packing, kg/s
R_c = radius of micropore, m
R_0 = radius of the inner channel in radial chromatography, m
R_1 = radius of the outer channel in radial chromatography, m
s = Laplace transformation variable
z = spatial coordinate of chromatography, m

Greek letters

ϵ = void fraction of the packed bed
ϵ_p = porosity of particle
ν = dimensionless reciprocal dispersion, defined in Eqs. 3 and 4
ρ = radial distance in micropore, m
ρ_B = particle bulk density = $(1 - \epsilon)\rho_p$, kg/m^3
ρ_p = particle density, kg/m^3

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