CONCLUSIONS

A direct and accurate method for the prediction of the critical temperatures of multicomponent hydrocarbon systems has been developed. From a knowledge of the normal boiling points and critical temperatures of the pure components the critical temperature of a mixture containing an unlimited number of components may be estimated with an expected error of approximately 1%. The components may be normal paraffins, isoparaffins, olefins, acetylenes, naphthenes, and aromatics and are limited only by the prescribed boiling-point ratio ranges. This approach may be extended to systems containing nonhydrocarbon components once a reasonable quantity of data on such systems becomes available.

NOTATION

A_{ij} = temperature coefficient for binary system consisting of i and j components
n = number of components
T_{b_i} = normal boiling point of ith component, °R.
T_c^* = corrected critical temperature for aromatic component, °R.
T_{c_i} = critical temperature of ith component, °R.
T_{c_m} = critical temperature of mixture, °R.
x_i = mole fraction of ith component
x_j = mole fraction of jth component
r = normal boiling-point ratio, T_{b_2}/T_{b_1}

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COMMUNICATIONS TO THE EDITOR

A Note on the Method of Moments

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Increasing attention has been devoted to methods of unsteady state analysis in the investigation of various systems of chemical engineering interest. Response analysis techniques are standard methods in feedback control theory, and any system which is rate controlled is subject to such analysis. Analytical solution of the equations of many systems, even though linear, is often formidable, and various alternatives have been used. Characterization of a response function in terms of moments can be useful in some cases and has been discussed by several workers (*1, 2, 3, 5, 7, 9, 10*) after the original suggestions of Van der Laan (*15*). Applications to chemical engineering have been almost exclusively in various discussions of the fixed-bed, axial dispersion problem. Although there are certain limitations associated with use of the technique (*10*), in many cases the method is most useful and warrants particular consideration as a convenient and accurate way to determine rate parameters from experimental data.

The use of moments in system response analysis, as reported by the majority of workers, involves the comparison of experimentally determined moments with those generated by various mathematical models which can be postulated for the system. By these means the proper model is selected and the associated rate parameters (that is diffusion coefficients, rate constants) determined. In practical applications to this task the efficacy of the method is often limited by the mathematical similarity of moments generated by different models and the resulting need of extremely accurate response data in the analysis. The difficulty in distinguishing between higher moments is often the culprit in these cases; thus for most purposes the use of moments is most convenient and accurate for the interpretation of response data when only the lower (zeroth to second) moments are required.

If $\xi(x, y, t)$ represents the response function of a system such that the following operational definition of the moments of this function

$$M_n = \int_0^\infty t^n \xi(x, y, t)\, dt \qquad (1)$$

is finite, then

$$\lim_{s \to 0} \frac{d^n}{ds^n} f(s) = -M_n \qquad (2)$$

where

$$f(s) = \int_0^\infty e^{-st} \xi(x, y, t)\, dt = \mathcal{L}[\xi(x, y, t)] \qquad (3)$$

Equations (1) to (3) require the function $\xi(x, y, t)$ to be bounded within $0 \leq t \leq \infty$ and to be defined by a boundary condition at infinity for the variable with respect to which the moments are defined.

The response function may also be expressed in terms of its moments via orthogonal polynomial expansion. Laguerre polynomials, orthogonal in the semi-infinite interval with an exponential weight function, are often convenient.* Various other expansions are discussed by Szegö (*14*) and have been applied to a chemical engineering problem by Olson and Southworth (*10*).

There are a great number of systems, aside from the residence-time distribution problem considered previously, where moments analysis is easily accomplished and, since integration rather than differentiation is involved, produces more accurate results than normal interpretative methods. Application of the method to some typical cases of transient response data will serve best to illustrate these comments.

CASE I—FIXED BED ADSORPTION

Eagleton and Bliss (*6*) investigated the drying of air in a packed bed of alumina. For this system they reported an unusual nonlinear equilibrium which was resolved into an isotherm involving a combination of highly favorable and linear equilibrium over the range of concentrations involved. The solution presented for the breakthrough curves involves the Klotz equation (*8*) for the region of concentrations on the highly favorable portion of the isotherm and an equation including both fluid and solid diffusional resistance terms for the region of concentrations on the linear portion of the isotherm. Both equations are based on the constant band-width assumption of de Vault (*16*).

The following differential equations are written for the breakthrough curve:

$$\frac{dc}{c} = \frac{c_o\, KgS}{q_\infty v}\, dz;\ c < c_d \qquad (4)$$

$$\frac{dc}{c_o} = \left(1 - \frac{c}{c_o}\right) \left[\frac{(\alpha_1/q_\infty)}{1 - \dfrac{\alpha_1}{q_\infty} - \dfrac{Rc_o}{q_\infty}}\right]$$

$$\frac{c_o\, KgS}{q_\infty v}\, dz;\ c > c_d \qquad (5)$$

The moments analysis of these equations is based on their integration along the breakthrough curves. In interpreting a breakthrough curve presented as (c/c_o) vs. z, it is convenient to define an arbitrary z scale, $z^+ = 0$ at the point where $c = c_d$. On the lower portion $(c < c_d) z^+$ increases with decreasing z, and on the upper portion $(c > c_d) z^+$ increases with increasing z, z^+ defined so as to remain always positive. For $c < c_d$, then

$$c(z^+ \to \infty) = 0$$
$$c(z^+ = 0) = c_d \qquad (6)$$

The requirements of the moments definition are satisfied for the variable c, and the form of Equation (4) is unchanged with the exception of the difference in sign between z^+ and z. Transforming with respect to z^+ one obtains

$$\hat{c} = \frac{c_d}{s + \left(\dfrac{c_o\, KgS}{q_\infty v}\right)} \qquad (7)$$

The moments are obtained as

$$M_o = \lim_{s \to o} \hat{c} = \frac{c_d\, q_\infty v}{c_o\, KgS} = \int_0^\infty c\, dz^+ \qquad (8)$$

$$M_1 = \lim_{s \to o} \left(\frac{d\hat{c}}{ds}\right) = c_d \left(\frac{q_\infty v}{c_o\, KgS}\right)^2 = \int_0^\infty z^+ c\, dz^+ \qquad (9)$$

For the portion of the breakthrough where $c > c_d$, the variable c must be redefined to satisfy the moments definition:

$$c = c_o + c^+$$

Then

$$c^+ = c - c_o$$
$$c^+(z^+ = 0) = c_d - c_o \qquad (10)$$
$$c^+(z^+ \to \infty) = 0$$

Substituting these relationships into Equation (5) and transforming the result with respect to z^+ one gets

$$\hat{c}^+ = \frac{c_d - c_o}{s + \mathrm{H}} \qquad (11)$$

where

$$\mathrm{H} = \left[\frac{\alpha_1/q_\infty}{1 - \dfrac{\alpha_1}{q_\infty} - \dfrac{Rc_o}{q_\infty}}\right] \left(\frac{c_o\, KgS}{q_\infty v}\right)$$

The moments are

$$M_o = \frac{c_d - c_o}{H} = \int_0^\infty c^+ dz^+ \qquad (12)$$

$$M_1 = -\frac{(c_d - c_o)}{H^2} = \int_0^\infty z^+ c^+ dz^+ \qquad (13)$$

A typical run carried out by Eagleton and Bliss involved the following known parameters:

$c_o = 0.00282$ lb. H_2O/lb. air
$v = 0.216$ lb. air/hr.
$\pi = 766.1$ mm. Hg
$T = 80°F$.
$q_\infty = 0.0552$ lb. H_2O/lb. solid
$\alpha_1 = 0.025$ lb. H_2O/lb. solid

The problem of interpretation thus involves determination of KgS, R (and hence KsS), and c_d. For this, only the zeroth moments are required, and the first moments may be used to check the consistency of results. Equation (8) is applied to the breakthrough data for successively higher assumed values of c_d, and KgS is calculated by this procedure. Over the region of breakthrough where this equation is valid, KgS will be constant; the point where the calculated KgS fails to agree with previously calculated values serves to define the value of c_d. Equation (12) is then applied to the breakthrough for $c > c_d$ to determine the value of R. The comparison of reported values with those obtained by the moments methods for this case is

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* An example of this form:

$$\xi(x, y, t) = e^{-\lambda t} \sum_{n=0}^{\infty} \left[\lambda \sum_{m=0}^{n} \frac{(-\lambda)^m M_n}{(m!)^2 (N - m)!}\right] L_n(\lambda t)$$

Parameter	Reported value (*6*)	Moments value
$KgS, \dfrac{\text{lb. } H_2O \text{ adsorbed}}{\text{hr.} - \text{lb. solid}\left(\dfrac{\text{lb. } H_2O}{\text{lb. air}}\right)}$	117	121
c_d, lb. H_2O/lb. air	7.03×10^{-4}	7.00×10^{-4}
$KsS, \dfrac{\text{lb. } H_2O \text{ adsorbed}}{\text{hr.} - \text{lb. solid}\left(\dfrac{\text{lb. } H_2O}{\text{lb. air}}\right)}$	4.1	11.3

Comparison of the breakthrough predicted by the two sets of constants shows that a considerable improvement in the agreement with experimental data is obtained with the moments values rather than with the authors' originally reported values. This result is typical of many cases of adsorption or ion exchange. The method of moments, involving integration of breakthrough data, yields more accurate values of the parameters than the slope techniques used previously.

CASE II—AXIAL DISPERSION IN FLUIDIZED BEDS

Rohan (*13*) investigated mass dispersion in fluidized beds using a transient response technique. The axial dispersion, plug flow model of Danckwerts (*4*) was satisfactory in describing the system, and values of the axial dispersion coefficient were determined with measurements of the slope of the breakthrough curves at $C = 0.5$. The following equation describes the process:

$$\left(\frac{1}{Pe}\right)\frac{\partial^2 C}{\partial X^2} - \frac{\partial C}{\partial X} = \frac{\partial C}{\partial \theta} \qquad (14)^*$$

where the boundary conditions are

$$\begin{aligned} C(X,O) &= 1 \quad & X &\geqq 0 \\ C(O,\theta) &= 0 \quad & \theta &\geqq 0 \\ C(X,\theta) &= 1 \quad & \theta &\geqq 0 \\ X &\rightarrow \infty & & \end{aligned} \qquad (15)$$

The system is subjected to a step function decrease in concentration at $\theta = 0$; thus C satisfies the requirements of the moments definition. Equation (14) is transformed with respect to θ:

$$\check{C} = \frac{1}{s}\left[1 - \exp\left(\frac{1 - \sqrt{1 + (4s/Pe)}}{2/Pe}X\right)\right] \qquad (16)$$

and the moments may be computed from this expression with L'Hôpital's rule. In this case the normalized response data yield

$$M_o = {}_o\int^\infty C d\theta = 1 \qquad (17)$$

Thus it is convenient to define higher moments about the zeroth moment. The first moment is then

$$M_1 = {}_o\int^\infty (\theta - 1) C d\theta = \left(\frac{E}{L\bar{v}}\right) - \frac{1}{2} \qquad (18)$$

A form more suitable for determination of E is given by

$${}_o\int^\infty (2\theta - 1) C d\theta = \frac{2E}{L\bar{v}} \qquad (19)$$

For a typical run with a water-fluidized bed of 1.09-mm. steel shot in a 2.489-cm. diameter column, the following values of E were calculated:

$E = 7.86$ sq. cm./sec. by mid-point slope method

$E = 6.20$ sq. cm/sec. by method of moments.

In this run L was 112.7 cm. and $\bar{v}$ was 15.6 cm./sec.

It would again be anticipated that the moments value is more accurate than that evaluated from the slope, even if moments and slopes are determined with the same accuracy. The former is proportional to the error in evaluation of the integral of Equation (19), whereas the latter is proportional to the square of the error involved in the slope determination:

$$E = (L\bar{v})\left[-\frac{1}{2\sqrt{\pi}\left(\dfrac{dC}{d\theta}\right)_{C=1/2}}\right]^2$$

These comments are supported by the comparison of predicted and experimental results over the entire concentration range. The standard deviation between predicted and experimental concentrations on the breakthrough in this run is 0.030 for the moments value and 0.037 for the mid-point slope value. Similar results attain over a large range of experimental conditions.

The general residence-time distribution problem of which this illustration is a part has been studied by Van der Laan and subsequent authors for much more complex boundary conditions using the $\delta(+0)$ forcing function response expanded about the mean. The simplified boundary conditions used here are justified by the small and approximately equal values of Pe^{-1} at entrance and exit to the experimental system (*13*). The relationship between the moments defined for the step function and the Van der Laan development, with the notation of Equation (1), is (*11*)

$$\lim_{s \to o} \frac{d^n f_1(s)}{ds^n} = \frac{(-M_1')^n}{(N+1)}\left[1 + \sum_{m=0}^{n-2} \frac{M'_{N-m}}{M_1^{(N-m)}}\binom{n}{m}\right] \qquad (20)$$

where

$$M_n' = {}_o\int^\infty (t - M_1)^n\ \xi(t)dt$$

$$M_1 = {}_o\int^\infty t\ \xi(t)dt$$

$$f_1(s) = \frac{1}{s}[1 - f(s)]$$

$$f(s) = \mathcal{L}[\delta(+0) \text{ forcing function}]$$

In addition arbitrary input functions may be used (*3, 9*) if one simultaneously determines the system input and response.

CASE III—ANALYSIS OF KINETIC DATA

Moments analysis is quite powerful in many cases of interpretation of kinetics. The method is limited to linear systems, but many important types of reactions involve complex schemes of simultaneous and successive reactions which are first order. Owing to the cumbersome form of the integrated equations for these schemes, interpretation methods usually employ differential or initial rate data, whereas moments analysis requires use only of the normally more reliable conversion-time data.

One such system of particular interest and importance is the homogeneous reaction

$$A \underset{K_2}{\overset{K_1}{\rightleftarrows}} B \underset{K_4}{\overset{K_3}{\rightleftarrows}} C$$

Let:

A, B, C = concentrations of A, B, and C at time t

A_o = concentration of A at time zero

$B_o = C_o = 0$

$A_\infty B_\infty C_\infty$ = concentrations of A, B, and C at equilibrium

The concentration variables are rearranged easily to conform to the moments definition:

$$A = A_\infty + A^+$$

$$B = B_\infty + B^+$$

$$C = C_\infty + C^+$$

and at $t = 0$ the boundary conditions are:

$$\begin{aligned} A &= A_o; \quad & A^+ &= A_o - A_\infty \\ B &= 0; \quad & B^+ &= -B_\infty \\ C &= 0; \quad & C^+ &= -C_\infty \end{aligned}$$

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* The assumptions of this model are an average mean velocity in the axial direction, negligible radial diffusion, and mass dispersion coefficient independent of time and position.

The rate equations may be written with the aid of equilibrium and material balance expressions as

$$\frac{dA^+}{dt} = -K_1A^+ + K_2B^+ \qquad (22)$$

$$\frac{dB^+}{dt} = (K_1 - K_4)A^+ - (K_2 + K_3 + K_4)B^+ \qquad (23)$$

$$\frac{dC^+}{dt} = K_3B^+ - K_4C^+ \qquad (24)$$

The transforms of these equations with respect to t yield the following:

$$\hat{A}^+ = \frac{K_2\hat{B}^+ + (A_o - A_\infty)}{s + K_1} \qquad (25)$$

$$\hat{B}^+ = \frac{(K_1 - K_4)\hat{A}^+ - B_\infty}{(s + K_2 + K_3 + K_4)} \qquad (26)$$

$$\hat{C}^+ = \frac{K_3\hat{B}^+ - C_\infty}{(s + K_4)} \qquad (27)$$

The zeroth moments are evaluated as

$$M_{OB^+} = \frac{(K_1 - K_4)(A_o - A_\infty) - K_1B_\infty}{K_2K_4 + K_1K_3 + K_1K_4} \qquad (28)$$

$$M_{OA^+} = \left(\frac{K_2}{K_1}\right) M_{OB^+} + \left(\frac{A_o - A_\infty}{K_1}\right) \qquad (29)$$

$$M_{OC^+} = \left(\frac{K_3}{K_4}\right) M_{OB^+} - \left(\frac{C_\infty}{K_4}\right) \qquad (30)$$

Since $K_1A_\infty = K_2B_\infty$, then

$$K_1 = \frac{(A_o - A_\infty)}{M_{OA^+} - \left(\frac{B_\infty}{A_\infty}\right) M_{OB^+}}; \quad K_2 = K_1\left(\frac{A_\infty}{B_\infty}\right) \qquad (31)$$

$$K_4 = \frac{C_\infty}{\left(\frac{C_\infty}{B_\infty}\right) M_{OB^+} - M_{OC^+}}; \quad K_3 = K_4\left(\frac{C_\infty}{B_\infty}\right) \qquad (32)$$

The method requires conversion-time data to equilibrium. The use of supplementary information such as the point of maximum concentration of B (if such exists) or initial rate data is not required. These data are necessary in usual interpretation methods; they are normally not as reliable as conversion data over a wide range and certainly not as reliable as the conversion-time integral employed in the moments analysis.

The procedure is readily extended to the general case considered by Rakowske (12):

$$N_1 \underset{K_2}{\overset{K_1}{\rightleftarrows}} N_2 \underset{K_4}{\overset{K_3}{\rightleftarrows}} N_3 \underset{K_6}{\overset{K_5}{\rightleftarrows}} \ldots N_{n-1} \underset{K_{2n-2}}{\overset{K_{2n-3}}{\rightleftarrows}} N_n \underset{K_{2n}}{\overset{K_{2n-1}}{\rightleftarrows}} \ldots N_N$$

The moments equations may be written for the system as before:

$$M_{ON^+_1} = \left(\frac{K_2}{K_1}\right) M_{ON^+_2} + \frac{(N_{10} - N_{1\infty})}{K_1} \qquad (33)$$

where

N_{10} = initial concentration of N_1

$$M_{ON^+_n} = \left[\frac{K_{2n-3}}{K_{2n-2} + K_{2n-1}}\right] M_{ON^+_{n-1}} + \left[\frac{K_{2n}}{K_{2n-2} + K_{2n-1}}\right] M_{ON^+_{n+1}} - \frac{N_{n\infty}}{K_{2n-2} + K_{2n-1}} \quad \text{(general)} \qquad (34)$$

$$M_{ON^+_n} = \left[\frac{K_{2n-3}}{K_{2n-2}}\right] M_{ON^+_{n-1}} - \frac{N_{n\infty}}{K_{2n-2}} \quad \text{(final)} \qquad (35)$$

The equilibrium concentrations are again required. In the general case there are $(n - 1)$ independent moments equations corresponding to Equations (33) and (34) and $(2n - 2)$ unknown rate constants. The complete system of $(2n - 2)$ equations is established by combining $(n - 1)$ equilibrium relationships of the form

$$\frac{N_{n\infty}}{N_{(n+1)\infty}} = \frac{K_{2n}}{K_{2n-1}} \qquad (36)$$

with the $(n - 1)$ independent rate equations. On substitution into the general matrix the odd rate constants are evaluated by the simple equation

$$K_{2n-1} = \frac{N_{10} - \sum_{i=1}^{n} N_{i\infty}}{M_{ON^+_n} - M_{ON^+_{n+1}} \cdot \frac{N_{n\infty}}{N_{(n+1)\infty}}} \qquad (37)$$

The simplicity of this form results from the use of the $M_{ON^+_n}$th moment in the equation for the K_{2N-1} rate constant.

NOTATION

c = concentration, $(MM^{-1}$ or $ML^{-3})$
c_o = initial concentration
c_d = water vapor concentration of gas phase at discontinuity on adsorption isotherm (MM^{-1})
C = dimensionless concentration
E = axial dispersion coefficient, $(L^2\theta^{-1})$
K_1, K_2, K_n = reaction rate constants, (θ^{-1})
KgS = fluid film mass transfer coefficient*
KsS = solid film mass transfer coefficient*
L = length of fluidized bed, (L)
$L_n(\lambda t)$ = Laguerre polynomial = $\sum_{m=0}^{n} \frac{N!}{m!}\binom{n}{m}(-\lambda t)^m$
$\mathcal{L}[\xi(x, y, t)]$ = Laplace transform of $\xi(x, y, t) = f(s) = \hat{\xi}$
M_n = n^{th} moment with respect to origin
M_n' = n^{th} moment with respect to mean, M_1
N_1, $N_2 \ldots N_n$ = reactant-product concentrations, (ML^{-3})
$N_{n\infty}$ = equilibrium concentration, (ML^{-3})
Pe = length Peclet number, $L\bar{v}/E$
q_∞ = adsorption capacity of solid in equilibrium with gas of concentration c_o, (MM^{-1})
R = $-KgS/KsS$
s = Laplace transform variable
t = time, (θ)
v = velocity, $(M\theta^{-1})$
$\bar{v}$ = mean velocity, $(L\theta^{-1})$
x, y = dimension coordinates, (L)
X = dimensionless length
z = total flow of air up to time t, (M)

Greek Letters

α_1 = point of discontinuity on adsorption isotherm, (MM^{-1})
π = total pressure, (FL^{-2})
$\xi(x, y, t)$ = system response function
θ = dimensionless time
λ = constant in argument of Laguerre polynomial

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* Units given in Case II.