The object of this column is to enhance our readers' collections of interesting and novel problems in chemical engineering. Problems of the type that can be used to motivate the student by presenting a particular principle in class, or in a new light, or that can be assigned as a novel home problem, are requested, as well as those that are more traditional in nature and that elucidate difficult concepts. Manuscripts should not exceed ten double-spaced pages if possible and should be accompanied by the originals of any figures or photographs. Please submit them to Professor James O. Wilkes (e-mail: wilkes@engin.umich.edu), Chemical Engineering Department, University of Michigan, Ann Arbor, MI 48109-2136.

RATE MEASUREMENT WITH A LABORATORY-SCALE TUBULAR REACTOR

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Electrically heated tubular flow reactors have been commonly used for rate measurement in research laboratories. Questions often arise concerning the effect of velocity profile, axial and radial molecular diffusions, and axial temperature distribution on the measured conversion and estimated rate. Analysis of reactors of this type can be a fruitful area for homework for a reaction engineering course. Two home problems are offered in this article; both are aimed at measurement of reaction rate of nitrogen oxide and char with a flow reactor of commonly adopted size.

The first problem justifies the use of conversion data from a laminar flow reactor for rate estimation. In a laminar tubular flow system without reaction, the concentration of a gaseous species, C, can be described to a two-dimensional dispersion model, *i.e.*

$$\frac{\partial C}{\partial t} = D_M \frac{\partial^2 C}{\partial z^2} + D_M \left(\frac{\partial^2 C}{\partial r^2} + \frac{1}{r} \frac{\partial C}{\partial r} \right) - u_0 \left(1 - \frac{r^2}{R^2} \right) \frac{\partial C}{\partial z} \quad (1)$$

where D_M represents the molecular diffusivity of the species of interest, and u_0 is the maximum centerline velocity. The analyses by Taylor,[1,2] Aris,[3] and Hunt[4] demonstrated that, under a specific criterion, Eq. (1) reduces to an axial-dispersed, plug-flow model, or

$$\frac{\partial C}{\partial t} = E_z \frac{\partial^2 C}{\partial z^2} - u \frac{\partial C}{\partial z} \quad (2)$$

where u is the mean fluid velocity and E_z is the effective axial dispersion coefficient, or the sum of molecular diffusivity and the apparent diffusivity contributed by the laminar velocity profile, *i.e.*

$$E_z = D_M + \frac{R^2 u^2}{48 D_M} \quad (3)$$

Equations (2) and (3) have been derived assuming that the radial mixing is great enough compared to the longitudinal convective mixing to ensure a uniform cross-sectional concentration. Mathematically, this criterion implies a small radial Peclet number, Pe_r, or

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$$\frac{1}{Pe_r} = \frac{D_M}{uR} \gg \frac{R}{16\,L} \tag{4}$$

where L is the length of the tube.

For a laminar-flow reactor involving a first-order chemical reaction and back mixing, the full description of the reacting system involves an additional reaction term in Eq. (1). Wissler[5] demonstrated that the Taylor-Aris model remains valid, i.e., Eqs. (3) and (4) remain the basis for

$$\frac{\partial C}{\partial t} = E_z \frac{\partial^2 C}{\partial \partial z^2} - u\frac{\partial C}{\partial z} + kC \tag{5}$$

provided that the reaction rate, k, satisfies the condition

$$\frac{kR^2}{\left(3.8^2\, D_M\right)} < 1 \tag{6}$$

At steady state, Eq. (5) has been solved by Wehner and Wilhelm.[6] For small $E_z/(uL)$, their solution after dropping higher-order terms in its series expansion becomes

$$1 - X = \exp\left[-kt + (kt)^2 \frac{E_z}{uL}\right] \tag{7}$$

where X is the conversion. The second factor in the above exponential term represents the deviation of the dispersion model, Eq. (5), from a plug-flow reactor.

The second problem investigates the effect of axial temperature variation on the estimation of intrinsic rate. In a tubular reactor heated by a single heating element of uniform resistance, reacting gas travels successively through heating, isothermal, and cooling sections, and rate estimation requires analysis of conversions in all three stages. Using an experimentally measured wall temperature distribution and a heat-transfer algorithm established by Sellars, et al.,[7] it is possible to calculate the average gas temperature
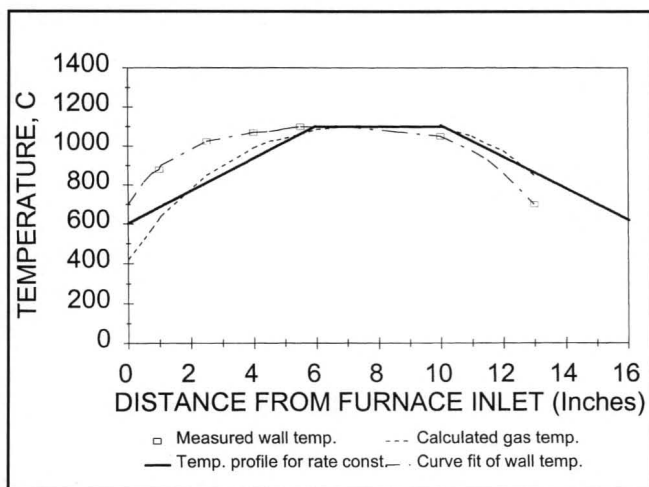

Figure 1. *Measured wall temperature, calculated gas temperature based on Sellars,* et al.,[7] *and simplified three-zone temperature profile for kinetic analysis.*

over a particular cross-sectional area of the tubular flow (see Figure 1). The predicted distribution indicates that, for a reactor equipped with a 30.5-cm-long heating element, each nonisothermal section has about the same length as the isothermal section. Since the gas travels slower and has a longer residence time in these nonisothermal sections, the rate estimation should take into consideration the conversions in these two sections. Problem 2 addresses this issue and seeks to recover the intrinsic rate of NO/char reaction and mass-transfer limitations. To simplify the calculation, the gas temperatures in the two nonisothermal sections are approximated by two constant heating-rate profiles. It is also demonstrated that, while this type of analysis traditionally requires extensive effort on trial and error, it can be conveniently solved now by contemporary software such as MathCad.

PROBLEM 1

An electrically heated tubular reactor with an I.D. of 1.91 cm and a heated length of 30.5 cm is used for measuring the rate of NO/char reaction. Products of the reaction are N_2, CO, and CO_2. The feed at 1 atm contains 1000 ppm of NO and char particles in a helium base. Using the heat transfer algorithm of Sellars, et al.,[7] and an experimentally measured temperature distribution, the axial gas temperature distribution is calculated (see Figure 1); in the central segment of the reactor the gas reaches 1100°C for about 10 cm. Well-dispersed char particles in the reacting gas are fed vertically downwards into the reactor at 25°C at a volumetric flow rate of 2000 cm³/min. Data collected from this reactor indicate that the NO disappearance rate is first order with respect to the NO concentration, and the NO conversions are always below 95% if a proper char feeding rate is chosen.

1. Using the Taylor-Aris[1-3] criterion, determine if an axial-dispersed, plug-flow model is appropriate for the approximation of gas concentration in a laminar flow system similar to that discussed above but containing no chemical reaction. If so, determine the effective diffusivity.

2. Using the Wissler[5] criterion, determine if the axial-dispersed, plug-flow model (Eq. 5) can be used for estimating the rate of the NO/char reaction.

3. Using the solution of Wehner and Wilhelm,[6] conclude if it is pertinent to use the plug-flow reactor model without dispersion for estimating the reaction rate.

Solution

1. A laminar flow system without chemical reaction can be approximated by an axial-dispersed, plug-flow model when Eq. (4) is satisfied or the radial Peclet number, $Pe_r (=uR/D_M)$, is small. At 1100°C, the flow rate and average residence

time of gas in the isothermal section of the reactor are 153.6 cm³s⁻¹ and 0.2 s, respectively. The diffusivity of NO in He can be estimated, based on molecular theory,[8] as

$$\frac{PD_{AB}}{(P_{cA}P_{cB})^{\frac{1}{3}}(T_{cA}T_{cB})^{\frac{5}{12}}\left(\frac{1}{M_A}+\frac{1}{M_B}\right)^{\frac{1}{2}}} = a\left(\frac{T}{\sqrt{T_{cA}T_{cB}}}\right)^b \quad (8)$$

where

D_{AB} cm²sec⁻¹
P   atm
T   K
a   2.745 x 10⁻⁴
b   1.823

This formula suggests D_{AB} = 13.7 cm²s⁻¹ at 1100°C. By resorting to the Chapman-Enskog kinetic theory (see Problem 2), D_{AB} = 8.1 cm²s⁻¹. Variations of diffusivity in this range do not change the conclusions to be discussed below. Based on the system parameters obtained above, the radial Peclet number at 1100°C and dimensionless term on the right-hand side of Eq. (4) are estimated as

$$\frac{1}{Pe_r} = \frac{D_M}{Ru} = 0.288 \quad \text{and} \quad \frac{R}{16L} = 0.00597 \quad (9)$$

Thus, the Taylor-Aris criterion, Eq. (4), is satisfied, and by resorting to Eq. (3), the effective axial dispersion coefficient is

$$E_z = D_M + \frac{R^2 u^2}{48 D_M} = 17.1 \frac{cm^2}{s} \quad (10)$$

2. Wissler's criterion gives the range of k for adopting the dispersion model, Eq. (5), as

$$\frac{kR^2}{(3.8)^2 D_M} < 1 \quad \text{or} \quad k < 216 \, s^{-1} \quad (11)$$

This condition corresponds to an extremely fast rate or a nearly complete conversion, specifically

$$1 - X = \frac{[NO]_{out}}{[NO]_{in}} = e^{-kt} = 10^{-19} \quad (12)$$

Since NO conversions from the reactor have always been below 95%, Wissler's criterion is satisfied.

3. At a high conversion, e.g., X=0.95, and a reaction time t=0.2s, Eq. (12) gives the rate from a plug-flow reactor without dispersion, or k=15.0 s⁻¹.The reciprocal value of the effective axial Peclet number is $1/Pe_z = E_z/uL = 3.38 \times 10^{-2}$. This small number implies only a small deviation from the assumption of zero dispersion. Specifically, resorting to the solution of Wehner and Wilhelm, or Eq. (7), we obtain the rate under the influence of dispersion, k = 16.9 s⁻¹. Thus, the assumption of zero dispersion leads to a maximum error of

about 13% at X = 0.95; this error is lower at lower conversions (4.4% at X=0.70 and 2.5% at X = 0.50).

PROBLEM 2

While Problem 1 justifies the use of a plug-flow reactor model for rate estimation, this problem seeks to study the effect of the two nonisothermal sections of the flow on measuring the intrisic rate of NO reaction with char.

1. Assuming the reaction is important only in the 10-cm isothermal region and the reaction is free from internal and external mass-transfer limitations, derive a design equation that relates reaction rate to experimentally observed NO conversion, internal surface area of char, char feeding rate, and residence time. The first-order reaction rate of NO on char surface can be expressed as

$$r_{NO} = -kAP_{NO} \quad (13)$$

where

r_{NO} rate of NO formation, moles s⁻¹ (g of char)⁻¹
A   specific, internal surface area of char, m²g⁻¹
P_{NO} partial pressure of NO, atm
k   rate constant, moles s⁻¹m⁻²atm⁻¹

2. Redo Part 1 of the problem assuming the conversions in the two nonisothermal regions of the reactor are not negligible and can be approximated by two constant heating-rate profile (see Figure 1).

3. Assuming the internal mass-transfer limitation cannot be ignored, estimate the frequency factor, activation energy, Thiele modulus, and effectiveness factor in a three-zone reactor as described in Part 2. For a reactive char derived from lignite under mild pyrolysis conditions, the following data are given:

conversion at 1100°C: 0.875
conversion at 1000°C: 0.815
mean particle radius: 0.0064 cm
char internal surface area, A: 255 m²g⁻¹
char feeding rate, W_1: 1.067 x 10⁻³ g min⁻¹
bulk density of char: 1.2 g cm⁻³
pore volume: 0.07 cm³g⁻¹

4. Estimate the external mass-transfer limitation.

Solution

1. *Conversion in the Isothermal Region* • A molar balance over a small section of tubular reactor yields[9]

$$F_{NO} + dF_{NO} = F_{NO} + (r_{NO})dW_t \quad (14)$$

where W_t is the char weight in the isothermal region and F_{NO} is the molar flow rate of NO. By the definition of conver-

sion, X, we obtain

$$dF_{NO} \equiv d\left[F_{NO,in}(1-X)\right] = -F_{NO,in}dX = +F_{NO,in}d\left(\frac{P_{NO}}{P_{NO,in}}\right) \quad (15)$$

Substituting Eqs. (13) and (14) into Eq. (15) and integrating gives

$$\ell n(1-X) = \ell n \frac{P_{NO}}{P_{NO,in}} = \frac{-kAW_t P_{NO,in}}{F_{NO,in}} \quad (16)$$

Furthermore, the molar flow rate of an ideal gas mixture can be expressed as

$$F_{NO,in} = \frac{P_{NO,in}}{P} \frac{v}{2.445 \times 10^4} \quad (17)$$

where the constant in the denominator denotes the specific volume of an ideal gas at 25°C in cm³ mole⁻¹, and v is the total inlet volumetric flow rate measured at 25°C in cm³s⁻¹. The char weight in the reactor, W_t, in Eq. (16) can be expressed in terms of char feeding rate, W_1, through the relation $W_t=W_1 t_h$, where W_1 is in g s⁻¹ and t_h the residence time in the isothermal section in s. Substituting the relation and Eq. (17) into Eq. (16), with P=1 atm and v=33.33 cm³s⁻¹, we obtain

$$\ell n(1-X) = \frac{-2.445 \times 10^4 kAW_1 t_h}{33.33} = -7.34 \times 10^2 kAW_1 t_h \quad (18)$$

This expression can be used in the estimation of reaction rate, k, if only the conversion in the isothermal region is important.

2. *Conversion in the Nonisothermal Regions* • For an ideal gas, the gas residence time t, in s, can be expressed as a function of temperature and distance traveled

$$dt = \frac{a}{G}dz = 25.62\frac{1}{T}dz \quad (19)$$

where

    a   cross-sectional area of the reactor tube, cm²
    G   gas volumetric flow rate, cm³s⁻¹
    z   longitudinal distance of gas traveled in the tubular reactor, cm
    T   gas temperature in the tube, K

During the heating and cooling periods, the estimated gas temperature is approximated by two linear temperature profiles, as shown in Figure 1. Specifically, since the gas temperature varies from 700 to 1100°C in 11.9 cm, the heating rate can be characterized as dT/dz=±33.51°C/cm. Substituting this expression into Eq. (19), we obtain

$$dt = \pm\frac{0.765}{T}dT \quad (20)$$

From a material balance, *i.e.*, Eqs. (13) through (15), we have

$$\frac{dP_{NO}}{P_{NO}} = \frac{-kAP_{NO,in}dW}{F_{NO,in}} \quad (21)$$

Substituting Eqs. (17) and (20) into Eq. (21) and integrating the resultant expression over the heating, isothermal, and cooling sections, we obtain

$$\ell n(1-X) = 733.6\left(-\int\frac{0.765}{T}W_1 Ak_0 e^{-\frac{E_a}{RT}}dT - t_h W_1 Ak_0 e^{-\frac{E_a}{RT}}\right.$$
$$\left. -\int-\frac{0.765}{T}W_1 Ak_0 e^{-\frac{E_a}{RT}}dT\right) \quad (22)$$

where k_0 and E_a are the preexponential factor and the activation energy. The two unknowns in Eq. (22), k_0 and E_a, can be estimated by implementing it twice for two sets of temperature/conversion data. Numerically, this set of two equations can be conveniently solved with MathCad. A temperature at which the reaction is slow, *e.g.*, 600°C, can be chosen as the lower integration limit. This procedure can be repeated for data collected over a wide range of temperatures so that linear regression can be conducted and the average k_0 and E_a can be obtained.

3. *Internal Mass Transfer Limitation* • To investigate the extent of internal mass transfer limitation, the observed Arrhenius rate in the last section can be considered the product of the intrinsic surface reaction rate and the effectiveness factor, η, *i.e.*, from Eq. (22)

$$\ell n(1-X) = 733.6\left(-\int\frac{0.765}{T}W_1 A\eta k_0 e^{-\frac{E_a}{RT}}dT - t_h W_1 A\eta k_0 e^{-\frac{E_a}{RT}}\right.$$
$$\left. -\int-\frac{0.765}{T}W_1 A\eta k_0 e^{-\frac{E_a}{RT}}dT\right) \quad (23)$$

The effectiveness factor is a function of the Thiele modulus, ϕ,[10] *i.e.*,

$$\eta = \frac{3}{\phi}\left(\frac{1}{\tanh\phi} - \frac{1}{\phi}\right) \quad (24)$$

where the Thiele modules is defined as

$$\phi = R\left(\frac{akA\rho_p}{D_{eff}}\right)^{0.5} \quad (25)$$

and

    R   radius of char particles, cm
    a   molar volume of an ideal gas, 2.445x10⁴, cm³ mole⁻¹
    D_{eff} effective diffusion coefficient, cm²s⁻¹
    ρ_p bulk density of char, g cm⁻³

The reciprocal of the effective diffusivity can be considered

a linear combination of the resistances contributed by the Knudsen and the bulk diffusivity[10]

$$\frac{1}{D_{eff}} = \frac{1}{D_{k,eff}} + \frac{1}{D_{12,eff}} \tag{26}$$

$$D_{k,eff} = 19400\left(\frac{\theta^2}{T_m A \rho_p}\right)\left(\frac{T}{M}\right)^{0.5} \tag{27}$$

$$D_{12,eff} = \frac{0.001858^{1.5}\left(\frac{M_1 + M_2}{M_1 M_2}\right)^{0.5}}{P\sigma_{12}^2 \Omega_D} \tag{28}$$

where

$D_{k,eff}$ Knudsen diffusion coefficient for a porous solid, cm²s⁻¹
$D_{12,eff}$ bulk diffusion coefficient of species 1 in species 2, cm²s⁻¹
θ particle void fraction
τ_m tortuosity factor based on the mean pore radius, assumed 2
M_1, M_2 molecular weights of diffusing molecules, M_{NO}=30, M_{HE}=4
M molecular weight of the gas medium
P pressure, 1 atm
Ω_D the "collision integral," a function of $k_B T / \varepsilon_{12}$, dimensionless
ε, σ force constant of the Lenard-Jones potential function, ε in g cm²s⁻², σ in Å
k_B Boltzmann constant, 1.38x10⁻¹⁶ g cm²s⁻²K⁻¹

Equations (26) through (28) allow calculation of the effective diffusivity. The pore fraction can be considered a product of the pore volume and bulk density. Thus, θ = pore volume density = 0.084. From Bird, et al.,[8] the parameters for the transport properties can be estimated as

$\varepsilon_{He}/k_B = 10.2$ K, $\sigma_{He} = 2.576$ Å
$\varepsilon_{NO}/k_B = 119$ K, $\sigma_{NO} = 3.47$ Å
$\sigma_{He-NO} = 1/2(\sigma_{He} + \sigma_{NO}) = 3.023$ Å
$\varepsilon_{He-NO} = (\varepsilon_{He}\varepsilon_{NO})^{0.5} = 34.84\ k_B g\ cm^2\ sec^{-2}$
$k_B T / \varepsilon_{He-NO} = 23.83$

$\Omega_D \approx 0.6776$

Substituting these constants into Eqs. (26) through (28), we obtain

$D_{12,eff}$ = 8.127 cm²s⁻¹
$D_{k,eff}$ = 3.98 x 10⁻⁵ cm²s⁻¹ for lignite char at 1100°C
D_{eff} = 3.98 x 10⁻⁵ cm²s⁻¹ for lignite char at 1100 °C

These results show that Knudsen diffusion controls the overall diffusion rate.

Equations (23) through (25) can be solved simultaneously by MathCad for the four unknowns η, ϕ, k_0, and E_a. Equation (23) is implemented twice for two sets of temperature/conversion data in each calculation. The results are: η=0.872, ϕ=1.53, k_0=0.07 moles s⁻¹m⁻²atm⁻¹, and E_a=3.80x10³ cal mole⁻¹. The results suggest that internal mass transfer limitation is

not negligible in the reacting system. This procedure can be repeated for data collected over a wide range of temperatures so that linear regression can be conducted and the average k_0 and E_a can be obtained.

4. *External Transfer Limitation* • The overall effectiveness factor is defined as

$$\Omega = \frac{\eta}{1+F} \tag{29}$$

where

$$F = \frac{C_g - C_s}{C_s} \tag{30}$$

is an index of external mass-transfer limitations; a large concentration gradient indicates large mass-transfer resistance. In the above expression, C_g=NO concentration in the mainstream of gas flow, in moles cm⁻³, and C_s=NO concentration at the particle surface, in moles cm⁻³. Under steady-state conditions, the mass-transfer rate of NO through the boundary layer equals the reaction rate, i.e.,

$$k_c S(C_g - C_s) = (-r)W_p \tag{31}$$

where

k_c mass transfer coefficient, cm s⁻¹
S external surface area of a single particle, cm²
$-r$ reaction rate, moles g⁻¹s⁻¹
W_p weight of a single particle, g

NO reduction on the char surface has been expressed as

$$(-r) = \eta k A P_{NO} \tag{32}$$

Assuming the gas is ideal, we have

$$P_{NO} = C_s RT \tag{33}$$

and the weight of a single char particle can be expressed as

$$W_p = \rho_p \frac{1}{6}\pi d_p^3 \tag{34}$$

Substituting Eqs. (31-34) into Eq. (30), we obtain

$$F = \frac{C_g - C_s}{C_s} = \frac{\eta k A R T \rho_p \pi d_p^3}{6 k_c S} \tag{35}$$

Assuming that the particles are entrained in the flow, the mass-transfer coefficient can be estimated based on the Frossling correlation with the Sherwood number (Sh)=2, i.e.,

$$k_c = \frac{D_{12} Sh}{d_p} = \frac{2 D_{12}}{d_p} \tag{36}$$

where D_{12}=8.127 cm²s⁻¹ (see Eq. 28). The particles have a mean diameter of 0.0128 cm; therefore, from Eq. (36),

$$k_c = 1.27 \times 10^3 \text{ cm s}^{-1} \tag{37}$$

Substituting this value and the parameters discussed in the

last section into Eq. (35), we obtain

$$F = 34.34\, \eta k \qquad (38)$$

and, from Eq. (29)

$$\Omega = \frac{\eta}{1 + 34.34\, \eta k} \qquad (39)$$

In the parameter-recovery process, Eq. (39) is added into the four-equation set discussed in Part 3, and the resultant five unknowns have been solved by MathCad (see Figure 2). In fact, results from the four-equation model can be used as initial trial values for the five-equation model to warrant convergence and save trial time. For this particular case, we obtain $\eta = 0.765$, $\phi = 2.294$, $k_0 = 0.156$ moles $s^{-1}m^{-2}atm^{-1}$, $E_a = 3.80 \times 10^3$ cal mole^{-1}, and $\Omega = 0.389$. The value of Ω is

smaller than that of η, suggesting that external mass transfer limitation cannot be ignored for the reactive char chosen in this investigation. At flame temperatures higher than 1100°C, the reaction rate will be higher and the extent of internal and external mass transfer limitations are expected to be even higher.

Figure 2 also contains an estimation of the contributions from the nonisothermal regions, which have the same order of magnitude as the conversion in the isothermal region, emphasizing the importance of including the conversions in the two nonisothermal regions for rate estimation.

ACKNOWLEDGMENTS

Figure 2. Solution of reaction parameters by MathCad. The conversions are assumed to be governed by both internal and external mass transfer limitations and reaction in a three-zone tubular reactor.

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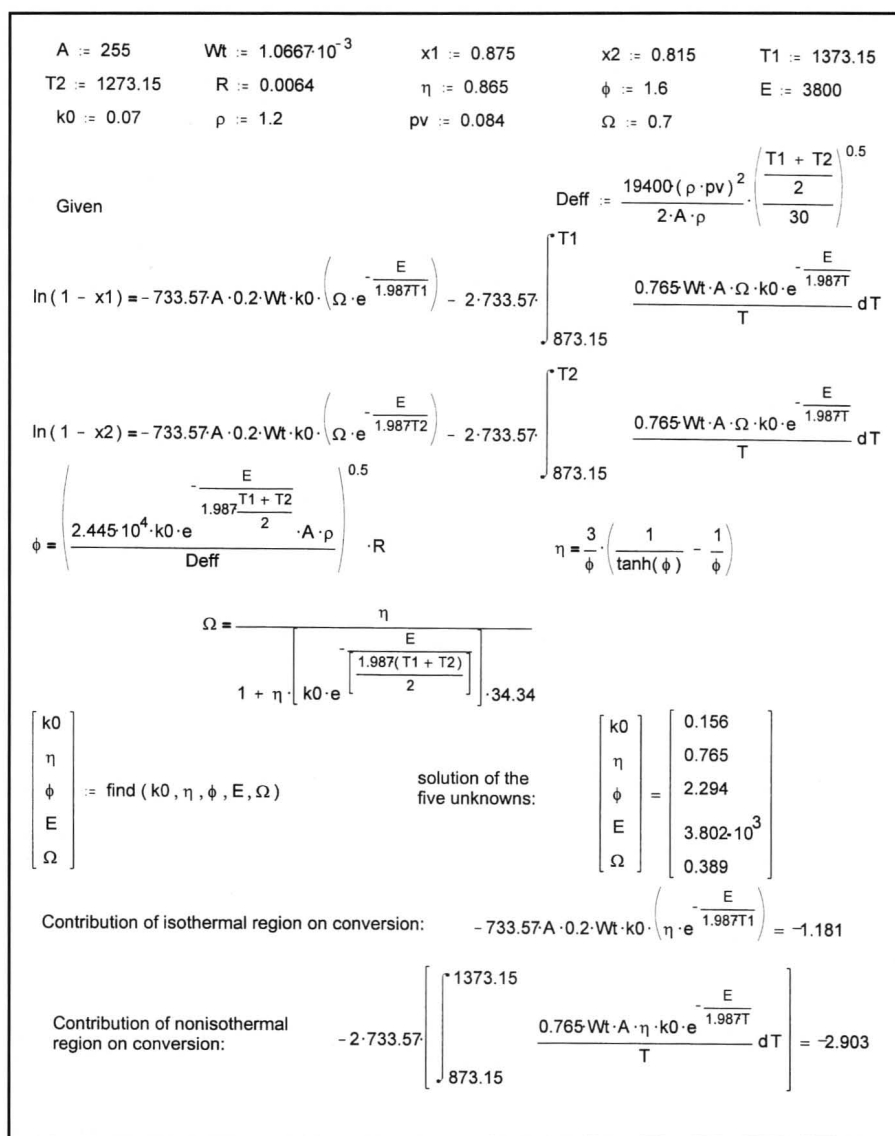10. Satterfield, C.N., *Mass Transfer in Heterogeneous Catalysis*, MIT Press, Cambridge, MA (1970) p