Design of fixed bed catalytic reactors

Raymond Charles Richardson
Iowa State University

Follow this and additional works at: http://lib.dr.iastate.edu/rtd
Part of the Chemical Engineering Commons

Recommended Citation
Richardson, Raymond Charles, "Design of fixed bed catalytic reactors " (1963). Retrospective Theses and Dissertations. 2555.
http://lib.dr.iastate.edu/rtd/2555

This Dissertation is brought to you for free and open access by Iowa State University Digital Repository. It has been accepted for inclusion in Retrospective Theses and Dissertations by an authorized administrator of Iowa State University Digital Repository. For more information, please contact digirep@iastate.edu.
This dissertation has been microfilmed exactly as received

RICHARDSON, Raymond Charles, 1929–
DESIGN OF FIXED BED CATALYTIC REACTORS.

Iowa State University of Science and Technology
Ph.D., 1963
Engineering, chemical

University Microfilms, Inc., Ann Arbor, Michigan
# Table of Contents

<table>
<thead>
<tr>
<th>Section</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>Nomenclature</td>
<td>iii</td>
</tr>
<tr>
<td>Introduction</td>
<td>1</td>
</tr>
<tr>
<td>Review of Literature</td>
<td>6</td>
</tr>
<tr>
<td>Experimental Reaction Data, Conversions, and Temperatures</td>
<td>64</td>
</tr>
<tr>
<td>Development of Reactor Design Variables</td>
<td>72</td>
</tr>
<tr>
<td>Results and Summary</td>
<td>87</td>
</tr>
<tr>
<td>Conclusions</td>
<td>148</td>
</tr>
<tr>
<td>Literature Cited</td>
<td>151</td>
</tr>
<tr>
<td>Acknowledgments</td>
<td>156</td>
</tr>
<tr>
<td>Appendix A</td>
<td>157</td>
</tr>
<tr>
<td>Appendix B</td>
<td>177</td>
</tr>
</tbody>
</table>
NOMENCLATURE

a  constant in reaction rate equation
a_p surface area of catalyst particle, sq ft
A_c cross-sectional area of reactor, ft^2
A_R wall heat transfer area per unit length of reactor, sq ft/ft
b  constant in reaction rate equation
B  constant in series solution
c  constant in reaction rate equation
C  concentration, lb moles/cu ft
C  mean concentration, lb moles/cu ft
C_p heat capacity, BTU/lb mole °F
D_p diameter of catalyst particle, ft
D_T diameter of catalytic reactor, ft
E  total diffusivity for mass transfer, sq ft/hr
E* dimensionless mass diffusivity, E/E_o
F  feed to reactor, lb moles/hr
G  mass velocity, lb/sq ft hr
G  mean mass velocity
G* dimensionless mass velocity, G/G_o
h  film heat transfer coefficient, BTU/hr sq ft °F
h_w overall heat transfer coefficient at wall, BTU/hr sq ft °F
\( h_{wf} \)  
wall film heat transfer coefficient, BTU/hr sq ft °F

\( \Delta H \)  
heat of reaction

\( k \)  
thermal conductivity, BTU/hr ft °F

\( k_e \)  
point effective thermal conductivity

\( k^* \)  
dimensionless effective thermal conductivity, \( k_e/k_e^o \)

\( L \)  
length of packed bed, ft

\( m \)  
position increment in radial direction, \( r/\Delta r \)

\( M \)  
total number of radial increments, \( R/\Delta r \)

\( M_{ave} \)  
average molecular weight

\( n \)  
summation index

\( n \)  
position increment in longitudinal direction, \( z/\delta z \)

\( N \)  
total number of longitudinal increments, \( L/\delta z \)

\( p_{E_A} \)  
partial pressure of component A in main gas stream

\( p_{E_A}^* \)  
equilibrium partial pressure of component A in main gas stream

\( p_{i-A} \)  
partial pressure of component A at interface

\( Pe_H \)  
Peclet number for heat transfer, \( (r_w C_p G/k_e)(r_w/L) \)

\( Pe_M \)  
Peclet number for mass transfer, \( (r_w v/E)(r_w/L) \)

\( Pe_H^* \)  
modified Peclet number for heat transfer, \( D_p C_p G/k_e \)

\( Pe_M^* \)  
modified Peclet number for mass transfer, \( D_p v/E \)

\( Pr \)  
Prandtl number, \( C_p \mu/k \)

\( Q \)  
heat generated, BTU/cu ft hr

\( r \)  
radial position measured from center of packed bed, ft
\( R \) radius of packed bed, ft
\( R_A \) reaction rate, gm-moles limiting reactant converted/gm catalyst hr
\( R_A^0 \) reaction rate at zero conversion
\( Re \) Reynolds number, \( D_T G/\mu \)
\( Re' \) modified Reynolds number, \( D_p G/\mu \)
\( Re_m' \) modified Reynolds number, \( \sqrt{a_p} G/\mu \)
\( s \) distance measured from wall of reactor, ft
\( t \) temperature
\( t \) time, \( z/v \)
\( T \) absolute temperature
\( U \) internal energy
\( v \) velocity, ft/hr
\( W \) mass of catalyst in reactor
\( x \) fractional conversion, \( (C_i v_i - C_v)/C_i v_i \)
\( x' \) moles reactant converted/moles reactant in feed
\( X^2 \) mean square deviation of deflected particles moving through packed bed
\( y_o \) mole fraction reactant in feed
\( z \) longitudinal distance
\( \alpha_w \) mass velocity of fluid flowing in direction of heat or mass transfer/mass velocity of fluid based on sectional area of empty tube in direction of fluid flowing
\( \beta \) packing constant for packed bed
\( \beta_\text{p} \) particle packing constant for lateral deflection
\( \gamma \) packing constant

\( \Gamma \) dimensionless concentration ratio, \( C / C_i \)

\( \delta \) finite difference operator in z-direction

\( \Delta \) finite difference operator in r-direction

\( \epsilon \) void fraction

\( \zeta \) dimensionless variable, \( (z / r_w \text{Pe}_H)(r_w / L) \)

\( \eta \) dimensionless variable, \( 5 / 3 \sqrt{\zeta} \)

\( \theta \) dimensionless temperature ratio, \( (T - T_w) / (T_o - T_w) \)

\( \lambda \) dimensionless longitudinal distance, \( z / L \)

\( \mu \) viscosity, lb/ft hr

\( \xi \) dimensionless variable, \( s / R \)

\( \rho \) density, lb/cu ft

\( \rho_B \) bulk density of catalyst, lb/cu ft

\( \sigma \) dimensionless radial variable, \( r / R \)

\( \tau \) shear stress

\( \nabla \) gradient operator

**Subscripts**

b bulk mean value
c convection
e exit conditions
f film coefficient
g gas
i inlet conditions
m  number of $\Delta r$ increments measured from center of packed bed
M  last $\Delta r$ increment, located at wall of packed bed
n  number of $\Delta z$ increments measured from reactor entrance
N  last $\Delta z$ increment, located at reactor exit
o  evaluated at the center of packed bed
r  radial coordinate
TD  turbulent conditions
w  wall conditions
x  cartesian coordinate
y  cartesian coordinate
z  cartesian coordinate

Superscripts

*  ratio of point value to that at center of reactor bed
o  conditions of no flow
INTRODUCTION

In its simplest form a catalytic reactor consists of a cylindrical tube packed with small catalyst pellets and surrounded, in the case of an exothermic reaction, by a cooling medium. Reactant gases enter the bottom of the reactor and the reaction occurs on the surface of the catalyst. Because of the presence of the cooling medium, the temperature at the center of the reactor will be higher than that at the tube wall. As a result the reaction rate will be much higher at the center than at the wall and accordingly the products of the reaction will tend to accumulate at the center while the reactants are accumulating at the wall.

Mass transfer occurs essentially by the mechanism of convection. Heat transfer can take place by a variety of mechanisms: by convection through the gas phase, by conduction through the solid, by radiation from solid to solid or from gas to solid, or by a combination of these mechanisms. In order to design such a fixed bed reactor, it is necessary to be able to predict the temperature and concentration at every point in the reactor, since present design methods are based on the numerical integration of basic differential equations which can be derived from material and energy balances over a differential element of the bed. In addition knowledge of the temperature and conversion at each
point is important in order to provide a means for estimation of local temperature conditions, which if too severe, might result in the destruction of the catalyst or in the occurrence of undesirable side reactions. Finally, a complete solution should reveal additional design information, probably in the form of dimensionless groups, in addition to the Peclet numbers for heat and mass transfer presently used, which is important in the design procedure for any fixed-bed catalytic reactor.

A bed of catalyst pellets is generally considered as if it were a continuous homogeneous media, and the physical properties of the packed bed or system are assigned values on the basis of a weighted average of each of the individual constituents making up the system. The weighting procedure is, in almost all cases, determined by a macroscopic or bulk contribution of all the components present in the system. The properties, such as porosity, mass velocity, and thermal conductivity will vary smoothly throughout the packed bed and, as a consequence, the solution of the differential equations representing the heat, mass, and momentum distributions will necessarily give rise to, correspondingly, smoothly varying values of temperature, concentration, and velocity.

Since the packed bed is composed of both solid particles and fluid, any typical volume element must be composed of a representative portion of the solid and the void space, which
will be a strong function of the spatial position in the bed. The volume chosen is designed to be small compared to the size of the bed but large enough so that the average or point value over the region chosen does represent a statistically stable average of the fluctuations. It is apparent that any given volume element will contain varying proportions of void and solid to such an extent that it is only an approximation to a point value even when the diameter of the tube is large compared to the particle size (i.e., $D_v/D_p > 10$). Even though these point values are subject to large fluctuations they do give meaning to the concept of smooth variation of properties in a packed bed. As more fundamental information of a microscopic or molecular nature becomes available, a more reliable statistical approach should give rise to a mathematical model which would be better able to predict accurately the point conditions in a packed bed.

The primary objective of this research was to determine the result of varying the transport properties, which affect the design of a fixed bed catalytic reactor. These transport properties are always determined empirically or calculated independently before they are combined in the form in which they appear in the representative differential equation. The manner in which these properties are combined has a definite, pronounced effect on the resulting temperature, concentration, and velocity distributions as predicted by the solution
of the corresponding differential equations. A second objective was to determine some of the more important parameters which would be useful in the design procedure of a fixed bed catalytic reactor. In order to accomplish these objectives it was necessary to obtain a general solution to the energy and mass transfer equations which would predict the point values of temperature and concentration as a function of radial and longitudinal position in the catalyst bed.

The use of groups of variables collected together to form dimensionless groups has been used extensively and profitably in the past, especially in the areas of heat transfer and fluid dynamics. An ultimate goal of research of this type would be to generalize the design of these reactors in terms of such groups. This method or technique is particularly important since the fundamental equations can be so arranged that the quantities enter the equations through these dimensionless combinations and the form of such equations is independent of the size of the units involved in the various terms in the equation. The use of dimensionless groups allows for interpretation of information and data where the mathematical relations are unknown or complex and in cases where two or more factors may vary in different experiments.

A computer program was developed which can provide temperature and concentration at any point in the reactor
for a given set of conditions such as:

1. inlet temperature distribution of feed gas
2. inlet composition distribution of feed gas
3. gas mass flow rate
4. reactor surface temperature
5. reactor size
6. catalyst pellet size
7. type of reaction
8. no reaction

The effect of varying the system parameters on temperature and concentration of the product can be determined by introducing these parameters, independently or in any combination, into the computer program.
Experimental data relating temperature to position in fixed bed reactors in which no reaction is occurring have shown that the factor used to measure the rate of radial heat transfer, effective thermal conductivity, varies with position. In addition, there are data which indicate that the mass velocity of the fluid through the reactor may vary with radial position. When a reaction occurs on the surface of the catalyst pellet, the heat of reaction will be adsorbed or released on the surface. This means that the mean temperature of the catalyst and the gas must be exactly the same. The size and shape of the catalyst pellets and the reactor, which is described by the porosity of the bed, effects the degree of turbulence which, in turn, will cause variations in heat and mass transfer across the diameter of the reactor. The design is based on which of the various assumptions can be made without undue loss in accuracy. Following are the types of design procedures that have been used.

**Isothermal and adiabatic operation**

In isothermal reactors, which represent the simplest case of a design problem, the rate of reaction will decrease
as the gases pass through the catalyst bed. The decrease in rate will depend upon the concentration change and pressure change in cases where the pressure drop is significant with respect to the total pressure. In practice it is difficult to operate a flow reactor under isothermal conditions because most reactions have a large heat effect.

In adiabatic operation heat transfer through the reactor wall is negligible and the temperature will change only in the longitudinal direction. In this situation the rate will vary in the direction of flow as a result of temperature changes, concentration changes, and pressure changes, if the pressure drop is significant.

For these reactors both radial temperature and concentration gradients are sufficiently small so that they may be neglected and the integration of the design equations becomes relatively simple.

Non-adiabatic, non-isothermal operation

The most difficult situation occurs when heat transfer through the wall must be taken into account. Generally the rate at which heat is transferred to or from the reactor is not sufficient to approach isothermal operation. This is usually the case for fixed bed reactors since the fluid velocities must be low enough to allow for the necessary
contact time. This results in insufficient mixing to obtain uniform concentration and temperature profiles. As a result the concentration and temperature will change in both the longitudinal and radial direction, and the integration of the design equation becomes a numerical, stepwise procedure. A general treatment of this type of reactor involves an incremental calculation across the diameter of the reactor tube for a small longitudinal increment and the repetition of this process for each successive longitudinal increment.

**Simplified method**

Heat and mass transfer within the bed are not considered. Plug flow is assumed although experimental temperature profiles appear to be parabolic. The only data necessary to establish the energy exchange with the surroundings would be the heat transfer coefficient at the wall, based on the bulk mean temperature of the reaction mixture. The relationships required are:

The basic design equation or mass balance,

\[ R_A dW = Fdx' \]

or

\[ A_c R_A' a_B dz = Fdx' = Fy_0 \sigma dx \]
The rate equation,

\[ R_A = f(\text{composition, temperature, pressure}) \]

The energy balance, taking into account the heat transfer to the reactor wall,

\[ F y_o dx(-\Delta H) - h_w A_w (t_m - t_w) dz = \sum_{n} m_n c_p \, dt_m \]

**Semi-rigorous procedure**

The variations in mass velocity, effective thermal conductivity, and mass diffusivity in the radial direction change the form of the differential equation for the energy and mass balances, but not the procedure for the solution.

The simplest procedure would involve integration of the equations in which the effective thermal conductivity, mass velocity, and mass diffusivity are assumed constant across the reactor diameter. Hall and Smith (20) investigated the reaction, \( SO_2 + \frac{1}{2}O_2 = SO_3 \) and

1. measured reaction rate data for the catalytic oxidation of \( SO_2 \) in a differential (small bed depth) reactor over the range of conditions encountered in an integral (large bed depth) reactor. A platinum catalyst on an alumina carrier was used;

2. measured the effective thermal conductivity at the
same conditions as the integral reactor;

(3) measured the radial temperature distribution in the gas and in the catalyst, and measured the conversion in the integral reactor, both the temperature and conversion were determined at catalyst bed depth of 0-, 2-, 4-, 6-, and 8-inches;

(4) predicted temperature and conversion in the integral reactor using Grossman's method (19) and the data obtained in 1 and 2 for comparison with experimental results of 3.

An overall effective thermal conductivity was determined from the experimental temperature data by graphical differentiation, but it was noted that the conductivity decreased as the tube wall is approached. These workers concluded that there was little radial mixing of the gas in the reactor despite the fact that the flow was in the turbulent range, since the radial temperature distribution in both the solid and gas phases were parabolic in nature.

The differential equation expressing the temperature distribution was integrated to give the point values of temperature for the integral reactor. The differential equation was integrated numerically with a constant effective thermal conductivity, neglecting the term representing the longitudinal or axial heat conduction. The average value of effective thermal conductivity for the packed bed was determined by adjusting the numerical value until the
calculated temperature profiles matched those obtained experimentally for the case of no reaction. The computed temperatures were in fair agreement with the experimental values near the center of the tube. As the tube wall was approached the computed values decreased much more rapidly than they should which indicated that the use of a constant thermal conductivity is not satisfactory. These workers found that the temperatures calculated with a higher value of thermal conductivity indicated that the temperatures were very sensitive to the value of the thermal conductivity.

The mean conversion as a function of catalyst bed depth based on the calculated temperatures were lower than those observed experimentally after a depth of two inches had been reached. This difference in computed and experimental values was attributed to the low calculated temperatures near the tube wall.

Irvin, Olson, and Smith (27) measured temperatures at various catalyst bed depths and radial positions in a 2-inch I.D. reactor through which $\text{SO}_2$ and air were passed. The catalyst was 1/8-inch alumina pellets coated with 0.2% platinum. Conversions were also determined at four gas mass velocities ranging from 147 to 512 lb/(hr)(ft$^2$). The Grossman (19) method was used to predict temperatures and conversions, neglecting mass transfer in the radial direction. These workers also correlated the effective thermal
diffusivity, $k_e/C_p G$, with the gas mass velocity and the packing size.

The values of $k_e/C_p G$ were evaluated from temperature versus radial position data for the case of no reaction. Point values of the effective thermal conductivity were determined by correlating them in the following manner:

$$(k_e/C_p) = a(D_p G/\mu)^{-0.51} \quad (\text{Eq. 1})$$

where $a$ is a function of the radial position and $\bar{G}$ is the overall average mass gas velocity. This equation represents the conditions fairly accurately for radial positions from the center of the bed up to 70% of the distance from the center to the wall. The values of the effective thermal diffusivity thus obtained were used in the solution of the differential equations to obtain bed temperatures and concentrations.

The Grossman method was a numerical-graphical approach based on rewriting the differential equation in finite difference form. The introduction of a variable effective thermal conductivity, through the $k_e/C_p G$ group, gave rise to non-uniform bed depth increments at increasing radial positions. The original method was modified by these workers by the adjustment to a uniform bed depth after each increment using linear interpolation. The general finite difference
equation for a cylindrical reactor is indeterminant at the center of the reactor and a special form of the equation is necessary. This special form is based on the assumption that radial symmetry exists for the temperature and concentration profiles. These workers presented three additional methods of obtaining the center temperatures:

1. Extrapolation of a curve of temperature versus radial position to the center of the reactor;
2. Modification of the finite difference equation using the assumption that the temperature versus radial position curve was parabolic when no reaction occurs. The experimental temperature data reported by both these workers and Hall and Smith (20) seem to reinforce this postulate;
3. Use of a special equation based on the assumption that a straight-line relationship existed between the temperature and the logarithm of the dimensionless radial position. The best agreement of computed and experimental temperature profiles was obtained using the third alternative method.

The maximum temperature deviation was 21°C at higher mass velocities (near 350 lb/hr ft²) while the average deviation was approximately 4°C. At the center of the bed, a maximum deviation of 60°C was observed at the lowest mass gas velocity (147 lb/hr ft²) and low bed depths. The largest contribution to this deviation was attributed to the use of the small number of radial increments used in the numerical
solution. The range of experimental mean conversion was 22.3 to 27.5% compared to a calculated value of 26.9%.

For a more complex design problem the values of the Peclet number for heat transfer may be obtained from experimental data for reactors in which no reaction is occurring or it may be obtained using the values of effective thermal conductivity predicted by the method of Argo and Smith (1). The Peclet number for mass transfer may be obtained from correlations developed by Fahien and Smith (17), Bernard and Wilhelm (6), and Baron (3). The differential equations are reduced to finite difference form and the composition and temperature versus position in the bed is determined by a step-ahead method using iteration at each point to satisfy the requirements for heat and mass transfer and the rate of reaction simultaneously.

In the design of continuous-absorption and extraction equipment the height of a transfer unit (HTU) has sometimes been used rather than a mass transfer coefficient and rate equations. This concept can be applied to the diffusion of reactants to the surface of a catalyst particle in a tubular flow type of reactor. The definition of the HTU is given by

\[
HTU = \frac{L}{\int dp_g / (p_g - p_{i_A})} = \frac{L}{NTU}
\]  

(Eq. 2)
\[ L = \text{total height of catalytic reactor}, \]

\[ p_{gA} = \text{partial pressure of component A in gas phase}, \]

\[ p_{iA} = \text{partial pressure at surface of catalyst particle} \]

The numerator in the integral is proportional to the amount of gas that must be transferred (since the number of moles is proportion to \(dp\)) and the denominator represents the driving force that causes the transfer. Hence the integral as a whole is a measure of the difficulty of the transfer job and is called the number of transfer units (NTU).

Caddell and Hurt (9) have extended this concept to gas-solid catalytic reactions by introducing two additional quantities:

1. height of a catalyst unit (HCU)

\[
\text{HCU} = \frac{L}{\int \frac{dp_{gA}}{(p_{iA} - p_{gA}^*})} \quad \text{(Eq. 3)}
\]

2. height of a reactor unit (HRU)

\[
\text{HRU} = \frac{L}{\int \frac{dp_{gA}}{(p_{g} - p_{gA}^*)}} \quad \text{(Eq. 4)}
\]

Writing Equations 2 and 3 in differential form to get the
pressure terms explicitly, it can be shown that

\[ \text{HRU} = \text{HTU} + \text{HCU}, \]  \hspace{1cm} (Eq. 5)

which means the process may be represented as two separate steps. The HTU represents the diffusional resistance and the HCU represents the surface processes.

For Equation 4 to be useful, the surface processes must be first order, or the linear driving force in Equation 2 cannot represent the kinetics of the surface steps. There is an additional problem in that the HCU includes the diffusional resistance of the products formed. This is because the equilibrium value of \( p_{\text{gA}} \) is related to the partial pressure of the products in the gas phase, not at the interface.

Due to these complications this method has not proved as useful as the procedure for a careful separation of diffusional effects from the surface resistances. However, where only an approximate kinetic treatment is desirable and a first order irreversible equation can be used for the surface processes, the HRU approach is simple to apply and gives results which are easy to visualize physically.

Gee, Linten, Maier and Raines (18) have made a process study of an industrial reactor which led to kinetic relationships too difficult to be solved mathematically by ordinary
desk methods. The system studied was a homogeneous gas-phase reaction occurring in a tubular reactor in which part of the heat of reaction was transferred to the surroundings. Although this reaction was carried out in an open tube, rather than in fixed bed reactor, many of the problems that occur in programming chemical processes on digital computers were discussed.

These workers developed a special relationship to take into account the change in heat transfer caused by fouling of the tube wall. The variation in the heat transfer coefficient with position in the tube and the on-stream time, the kinetic equation, pressure drop equations for the non-adiabatic non-isothermal conditions in the reactor lead to simultaneous non-linear partial differential equations. The Whirlwind I computer at M.I.T., an electronic digital computer, was used in the solution of these equations. The program was used for 50 typical runs designed to cover all possible combinations of initial conditions.

A companion paper by Beutler (7) presented some of the particular considerations for use of computers. The size of the Whirlwind, comparison of analog and digital types, information flow diagrams, typical machine orders, and errors resulting from finite difference approximations were discussed.

Beutler also presented a method for comparing the cost
of solution using various machines and the cost using manual methods.

An expression for cost comparison, in cost per solution is given by:

\[ C = \frac{a \cdot A}{n} + b \cdot B \]  
(Eq. 6)

\( C = \) cost per solution, dollars,
\( a = \) hours of programming, coding, and debugging time,
\( b = \) machine hours per solution,
\( A = \) programming cost, dollars per hour,
\( B = \) machine cost, dollars per hour,
\( n = \) total number of solutions required.

The first term in this formula represents the preliminary costs charged against each solution. The important factor in many cases where \( n \) is relatively small is the size of \( a \), since it basically determines the lower limit on problems that can be solved economically. In addition, \( a \) has a strong influence on the value of \( n \), at which curves cross for various computers.

For a digital computer, as a first approximation,

\[ a = \frac{100}{E} \left[ w + \left(1 + \frac{B}{A}\right)d \right] \]  
(Eq. 7)
where

\[ E = \text{programmer efficiency, } 100\% \text{ for professionals,} \]
\[ \text{programmers, perhaps } 20\% \text{ for a novice,} \]
\[ w = \text{programming and coding time, hours,} \]
\[ d = \text{debugging time, hours,} \]
\[ A, B = \text{as before.} \]

For large computers Beutler estimates \( B/A \) to be as large as 30.

A recent paper by von Rosenberg, Durrill, and Spencer (46) demonstrated the use of partial differential equations to treat temperatures and reactant concentrations in the longitudinal and radial directions of the reactor. The subsequent finite difference equations were solved for a number of different inlet concentrations on an IBM 650 computer.

Equations have been formulated to describe a flow reactor from which heat is being removed at the walls. A single, first order, irreversible reaction is considered for which the effect of temperature on the reaction rate constant is represented by an Arrhenius type relation.

The mathematical description of an exothermic reaction in packed beds has been developed to assist in the design of a pilot unit, in guiding the experimental work, and in interpretation of the data obtained from the unit.

Deans and Lapidus (15) have recently developed a unique
mathematical model for predicting the mixing characteristics of fixed beds packed with spheres for the case of no reaction. This model was based on a 2-dimensional network of perfectly stirred tanks. The results based on their method were compared with those predicted by the partial differential equation description of flow in fixed beds against the experimental axial and radial mixing characteristics. This model was able to predict the abnormally low axial Peclet numbers observed in the liquid phase system in unsteady state by the introduction of a capacitance effect.

In a further study this model was extended to include the effect of a first order, irreversible, exothermic Arrhenius-type reaction. The transient and steady state cases were developed for a reactor of constant wall temperature.

Derivation and Application of General Design Equations

In order to make a proper design for a fixed bed catalytic reactor, it is necessary to be able to predict accurately point values of temperatures and concentration for any given set of initial conditions. Therefore, it is necessary to write both an energy and mass balance for the reactor. Then these equations can be solved explicitly for the desired variables.
Equation of change for energy

The general energy equation for a fluid flowing in a packed bed can be expressed in the following form:

\[ \rho \frac{D}{Dt} (U + \frac{1}{2}v^2) = -\nabla \cdot q + \rho (v \cdot q) - \nabla \cdot (p v) \]

\[ -\nabla \cdot (r \cdot v) + R_A \rho_B \Delta H \]  
(Eq. 8)

For flow through a cylindrical packed bed, in terms of dimensionless variables, Equation 8 can be transformed to give

\[ \frac{\partial \theta}{\partial \lambda} = \frac{1}{Pe_H G* \sigma} \frac{\partial}{\partial \sigma} (k_e \sigma \frac{\partial \theta}{\partial \sigma}) - \frac{R_A \rho_B \Delta H L}{G(T_o - T_w) C_p G*} \]  
(Eq. 9)

where

\[ \theta = \frac{T - T_w}{T_o - T_w}, \quad \sigma = \frac{r}{r_w}, \quad \lambda = \frac{z}{L}, \]

\[ k_e = k_e / k_{e_o} = \text{ratio of point effective thermal conductivity to effective thermal conductivity at center of bed}, \]

\[ G* = G / G_o = \text{ratio of point mass velocity to mass velocity at center of bed}, \]
\[ P_{e}^{I} = \left( R_{w} G_{o} C_{p} / k_{o} \right) (r_{w}/L) = \text{Peclet number for heat transfer at center of bed}, \]

\[ R_{A} = \text{reaction rate, gm-moles SO}_{2} \text{ converted/(gm catalyst)(hr)}; R_{A} \text{ is a function of both temperature and conductivity}, \]

\[ \rho_{B} = \text{bulk density of catalyst}, \]

\[ \Delta H = \text{heat of reaction}, \]

\[ r_{w} = \text{radius of tube}. \]

By the method of Crank and Nicolson (14) Equation 9 can be further transformed to represent the temperature at any point \((m,n)\) in the bed as described in Figure 1 to give

\[ \theta_{n+1,m} = \theta_{n,m} + \frac{\delta t}{2P_{e}^{I} (\Delta \sigma)^{2} G_{m}^{*}} \left\{ k_{*} e_{m+\frac{1}{2}}^{*} (1 + 1/2m) \right\} \]

\[ (\theta_{n+1,m+1} + \theta_{n,m+1} - \theta_{n+1,m} - \theta_{n,m}) \]

\[ - k_{*} e_{m-\frac{1}{2}}^{*} (1 - 1/2m) (\theta_{n+1,m} + \theta_{n,m} - \theta_{n+1,m-1} - \theta_{n,m-1}) \}

\[ - \frac{R_{A} \rho_{B} \Delta H L}{G_{o} (T_{o} - T_{w}) C_{p} G_{m}^{*}} \]

\( \text{(Eq. 10)} \)

where

\[ \sigma_{m} = m \Delta \sigma; \quad 1 \leq m \leq M; \quad \Delta \sigma = 1/M \]
Figure 1. Schematic layout of reactor bed for development of finite difference equations

**Curve A** - Temperature profile

**Curve B** - Concentration profile
\[ \lambda_n = n \delta \lambda; \quad 0 \leq n \leq N; \quad \delta \lambda = 1/N \]

The notation, \( \theta_{n+1, m} \), is used to indicate the value of the dimensionless temperature ratio, \( \theta[(n + 1)\delta\lambda, m\Delta\sigma] \), at the points, \( \sigma = m\Delta\sigma \) and \( \lambda = n\delta\lambda \). Other variables are subscripted in the same manner.

At the center of the packed bed, Equation 9 has the following form:

\[
\theta_{n+1, 0} = \theta_{n, 0} + \frac{2\delta\lambda}{Pe_H (\Delta\sigma)^2 G^*_h} \left[ k_e^* \left( \theta_{n+1, 1} - \theta_{n+1, 0} + \theta_{n, 1} - \theta_{n, 0} \right) \right] \\
- \frac{Ra_{PB} \Delta HL}{G_0 (T_o - T_w) C_p G^*_h} (Eq. 11)
\]

This special form is necessary since Equation 10 becomes indeterminate at the center of the bed, \( \sigma = 0 \).

For the special case when the derivatives in Equation 9 are represented by a first order approximation to the differentials and when average bed properties are assumed \( (k_e^* = G^*_h = 1) \), Equation 9 can be transformed to give:

\[
\theta_{n+1, m} = \theta_{n, m} + \frac{\delta\lambda}{Pe_H (\Delta\sigma)^2} \left[ (1 + 1/2m)(\theta_{n, m+1} - \theta_{n, m}) \right]
\]
At the center of the bed, \( m = 0 \), Equation 12 reduces to the form:

\[
\theta_{n+1,0} = \theta_{n,0} + \frac{\delta \lambda}{P_{eh}(\Delta \sigma)^2} \frac{2D_L}{R^2} [\theta_{n,1} - \theta_{n,0}] - \frac{R_A \rho_B \Delta H L}{G_o(T_o - T_w)C_p}
\]

(Eq. 13)

**Equation of change for mass balance**

The general equation for steady state mass transfer, assuming the packed bed represents a continuous homogeneous medium, is given by:

\[
[\nabla \cdot (-E \nabla C)] + [\nabla \cdot (C \mathbf{v})] + R_A \rho_B = 0
\]

(Eq. 14)

where

- \( E \) = total diffusivity for mass transfer
- \( C \) = concentration of limiting reactant
- \( v \) = velocity of fluid
- \( R_A \rho_B \) = mass generation
In terms of cylindrical coordinates, Equation 14 reduces to

\[
\frac{1}{r} \frac{\partial}{\partial r} \left( -Er \frac{\partial C}{\partial r} \right) + \frac{\partial}{\partial z} \left( Cv_z \right) + R_A \rho_B = 0 \quad \text{(Eq. 15)}
\]

with the boundary conditions

(1) \( \frac{\partial C}{\partial r} \bigg|_{r=0} = 0 \)

(2) \( \frac{\partial C}{\partial r} \bigg|_{r=r_w} = 0 \)

In terms of the dimensionless variables

\[ \sigma = \frac{r}{R}, \quad \lambda = \frac{z}{L}, \quad = C/C_1, \]

Equation 15 can be transformed into

\[
\frac{\partial}{\partial \lambda} = \frac{1}{Pe_M v^* \sigma} \left( E^* \frac{\partial}{\partial \sigma} \right) - \frac{R_A \rho_B L}{v_0 C_1 v^*} \quad \text{(Eq. 16)}
\]

where

\[ C_1 = \text{initial concentration of limiting reactant}, \]

\[ v_{z_o} = \text{velocity at the center of the packed bed in} \]
the axial direction,

\[ E_0 = \text{mass diffusivity at the center of the packed bed,} \]

\[ v^*_z = \frac{v_z}{v^*_z}, \]

\[ E^* = \frac{E}{E_0}, \]

\[ Pe^*_M = \left( \frac{r_w v^*_z}{E_0} \right) \left( \frac{r_w}{L} \right) = \text{Peclet number for mass transfer}, \]

Equation 16 can then be reduced to a finite difference equation analogous to Equation 10 for heat transfer:

\[ \Gamma_{n+1,m} = \Gamma_{n,m} + \frac{\delta \lambda}{2Pe^*_M \delta \sigma} \left[ \frac{E^*}{m+\frac{1}{2}}(1 + 1/2m) \right] \]

\[ \left( \Gamma_{n+1,m} + \Gamma_{n,m+1} - \Gamma_{n+1,m} - \Gamma_{n,m} \right) - \frac{E^*}{m-\frac{1}{2}}(1 - 1/2m) \]

\[ \left( \Gamma_{n+1,m} + \Gamma_{n,m} - \Gamma_{n+1,m-1} - \Gamma_{n,m-1} \right) - \frac{R_A \rho E L}{v^*_z \delta i \delta m} \]

\[ (\text{Eq. 17}) \]

Equation 17 is not valid at the center of the bed, as it becomes indeterminant at \( \sigma = 0 \), or at the wall, since the slope of the concentration versus radial position curve must be zero (no mass transferred through the wall). As in the case for heat transfer, the value of the center temperature is
determined by application of the Crank-Nicolson finite difference technique to a radial position, \((\Delta \sigma/4)\), away from the center. Thus Equation 16 has the following form, similar to that for heat transfer,

\[
\Gamma_{n+1,0} = \Gamma_{n,0} + \frac{2(\delta \lambda)}{(\Delta \sigma)^2 \rho e_{M_v} v^{*}_L} \left[ E^{*}_{1/4} \right] (\Gamma_{n+1,1} - \Gamma_{n+1,0}) \\
+ [\Gamma_{n,1} - \Gamma_{n,0}] - \frac{R_A c_B L}{v_{z_0} c_{v^{*}_L}} (Eq. 18)
\]

where the subscripts, \(n\) and \(m\), again refer to the longitudinal distance from the origin and to the radial distance from the origin, respectively. A similar procedure is followed to obtain the equation at the wall except the derivatives are evaluated at \((M-\frac{1}{4})\) to give

\[
\Gamma_{n+1,M} = \Gamma_{n,M} + \frac{(\delta \lambda)(M-\frac{3}{4})}{\rho e_{M_v} v^{*}_L (M-\frac{1}{4}) (\Delta \sigma)^2 (M-\frac{1}{4})} \left[ E^{*}_{M-\frac{1}{2}} (\Gamma_{n+1,M-1} \\
+ [\Gamma_{n,M-1} - \Gamma_{n+1,M-1}] - \frac{R_A c_B L}{v_{z_0} c_{v^{*}_L} (M-\frac{1}{4})} \right] (Eq. 19)
\]

For the special case when constant bed properties \((E^{*} = v^{*} = 1.0)\) and plug-flow are assumed, and when a first
order approximation of the derivative is used, Equation 17 reduces to the form,

\[ \Gamma_{n+1,m} = \Gamma_{n,m} + \frac{\delta \lambda}{2 \text{Pe}_M^* (\Delta \sigma)^2} \left[ (1 + 1/2m)(\Gamma_{n,m+1} - \Gamma_{n,m}) \right] \\
- (1 - 1/2m)(\Gamma_{n,m} - \Gamma_{n,m-1}) - \frac{R_A \rho_B \rho_L}{v_{Z_0} C_1} \]  
(Eq. 20)

Similarly Equations 18 and 19 reduce to

\[ \Gamma_{n+1,0} = \Gamma_{n,0} + 2(\delta \lambda)/(\Delta \sigma)^2 \text{Pe}_M^* (2 \Gamma_{n,1} - 2 \Gamma_{n,0}) - \frac{R_A \rho_B \rho_R}{v_{Z_0} C_1} \]  
(Eq. 21)

and

\[ \Gamma_{n+1,M} = \Gamma_{n,M} + \frac{(\delta \lambda)(M-\frac{1}{2})}{\text{Pe}_M^* (\Delta \sigma)^2 (M-\frac{1}{2})} \left[ 2 \Gamma_{n,M-1} - 2 \Gamma_{n,M} \right] - \frac{R_A \rho_B \rho_L}{v_{Z_0} C_1} \]  
(Eq. 22)

respectively.

For the special case of constant bed properties and uniform velocity profile Equation 16 can be re-written to express the concentration of the limiting component in terms of the conversions, \( x \), where
\[ x = \frac{\text{moles limiting reactant consumed}}{\text{moles limiting reactant in feed}} \]

If \( C_i \) is the concentration entering the reactor,

\[ x = \frac{C_i - C}{C_i} \]

and if the entering mole fraction of the limiting component is \( y_0 \), then

\[ C_i v_z = G_0 y_0 / \text{MW}_{\text{ave}} \]

where \( \text{MW}_{\text{ave}} \) is the average molecular weight of the mixture. Thus Equation 16 becomes

\[
\frac{\partial x}{\partial \lambda} = \frac{1}{P_e M_o} \left( \frac{\partial}{\partial \sigma} \left( \frac{\partial x}{\partial \sigma} \right) - \frac{R_A \rho B \text{LMW}_{\text{ave}}}{G_0 y_0} \right) \quad (\text{Eq. 23})
\]

and Equation 17 has the form,

\[
x_{n+1, m} = x_{n, m} + \frac{\delta \lambda}{P_e M_o (\Delta \sigma)^2} \left[ (1 + 1/2m)(x_{n, m+1} - x_{n, m}) \\
- (1 - 1/2m)(x_{n, m} - x_{n, m-1}) \right] - \frac{R_A \rho B \text{LMW}_{\text{ave}}}{G y_0} \quad (\text{Eq. 24})
\]
Equations 18 and 19 which are valid for the center of the tube and at the wall, respectively, can be transformed in a similar manner:

\[ x_{n+1,0} = x_{n,0} + \frac{2(6\lambda)/(Pe_o^2)}{2} \left( 2x_{n,1} - 2x_{n,0} \right) \]  
(Eq. 25)

\[ x_{n+1,M} = x_{n,M} + \left[ \frac{(6\lambda)(M-\frac{1}{2})}{Pe_o^2} \right] \left( 2x_{n,M-1} - 2x_{n,M} \right) \]  
(Eq. 26)

**Heat Transfer**

The accurate design of fixed bed tubular, catalytic reactors can only be attempted with a full knowledge of the heat transfer characteristics of the system. In most industrial applications, heat transfer considerations largely determine the size of the reactor and the limits of profitable or safe operating conditions once the former has been decided upon.

Even with simplified methods of reactor design, considerable uncertainty is attached to the predictions of heat transfer rates to or from the wall of tubular exchangers packed with granules, as well as from point to point within
the packed bed. The effects of gas flow rate, catalyst pellet size and shape, tube diameter and length are shown to be important by the literature, yet these effects cannot be quantitatively evaluated with any certainty.

The equation

$$k\left[\frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\partial T}{\partial r}\right) + \frac{\partial^2 T}{\partial r^2}\right] = k\left[\frac{\partial^2 T}{\partial r^2} + \frac{1}{r} \frac{\partial T}{\partial r} + \frac{\partial^2 T}{\partial z^2}\right] = \frac{\partial T}{\partial z}$$

(Eq. 27)

is the heat balance equation for rod-like flow of a fluid flowing through a cylindrical tube exchanging heat with the fluid. This equation has been widely used to interpret heat transfer to fluids in streamline flow through pipes.

Equation 27 has been used to describe the heat transfer process for a fluid flowing through a heat-exchanging cylindrical tube containing packing, in which $k$ is replaced by $k_e$ defined as an equivalent thermal conductivity of the system of fluid and packing.

It has been generally assumed that rod-like flow, or no radial variation in the velocity, is reasonably valid for values of $D_T/D_p$ greater than ten; however, this assumption often leads to large deviations between predicted and experimental values of $k_e$. The term,

$$k_e \frac{\partial^2 T}{\partial z^2},$$
representing axial heat flow by conduction may generally be
neglected for low conductivity packing.

Equation 27 has been the basis for the work of many in­
vestigators to obtain effective thermal conductivity from
experimental data and then correlate the results in terms of
the important variables. Two methods have been used in this
connection.

In the first, Equation 27 has been integrated for a
point solution of the temperature within the bed. A further
integration of the point equation has been performed to give
a solution in terms of the mean fluid temperature entering
and leaving the bed. Then from the measured values of these
latter two quantities, an average overall thermal conductivity
could be computed.

The second method involves actual measurement of tem­
peratures across the diameter of the bed. The experimental
values of the temperature profiles have then been used to
establish what are called point values of the effective
thermal conductivity.

**Overall effective thermal conductivity**

Using the integral mean temperature to replace the
radial point fluid temperatures, the integration of Equation
1 gives a solution which is a converging infinite series.
This method has been used by Singer and Wilhelm (44) who presented the solution of Equation 1 in terms of point solutions for solids and fluid temperature and also in terms of mixed terminal fluid temperatures. The equations have been set up for the following cases:

1. fixed bed heat exchangers,
2. moving bed heat exchangers,
3. fixed bed chemical reactors,

assuming constant thermal properties within the system and also plug flow, or no radial variation in the velocity. In the case of chemical reaction this heat generation term is the form of an Arrhenius equation and it is temperature dependent only and therefore it is expressed as a linear function of the solids temperature.

Singer and Wilhelm have pointed out that heat transfer in packed beds is brought about by a number of mechanisms which are capable of separate evaluation. These mechanisms are described generally as:

1. molecular diffusion as for a stagnant fluid,
2. turbulent or eddy diffusion due to dividing and mixing of fluid passing around particles of packing, and
3. a series mechanism involving heat transfer through the solid packing followed by transfer to neighbouring particles by point contact, convection,
conduction, and radiation.

The contributions of these three mechanisms are in parallel or additive and may be estimated independently.

The contribution of mechanism 1 may be calculated from the molecular diffusivity or thermal conductivity of the stagnant fluid,

$$\text{Pe}_M^1 = \frac{D_p v}{\alpha} = \frac{D_p G C_p}{k} = (Re')(Pr) \approx 0.74 \text{ Re}'$$, for gases

The contribution of mechanism 2 has been measured independently by Bernard and Wilhelm (6) who found that for values of $D_T/D_p$ greater than ten, modified Reynolds numbers, $\text{Re}'$, greater than 100, the modified Peclet number, $\text{Pe}_H^1$, has a value of 10 to 11.5 independent of the Reynolds number. Baron (3) has used a "random walk" method to show that this value of $\text{Pe}_{TD}^1$ is theoretically sound.

Using a number of simplifying assumptions the contribution of mechanism 3 has been estimated by Argo and Smith (1) from:

(a) the conductivity of the solid comprising the packing, $k_S$,

(b) the point-contact conductivity as calculated by Schumann and Voss (42) and modified by Wilhelm et al. (47),

(c) the convection heat transfer coefficients of
Hougen and Wilke (26) and Hougen et al. (24) for transfer from the particles to gas, \( h_c \), and (d) the radiation effect.

Mechanisms b, c, and d are in parallel and in series with mechanism a.

\( k_e/G \) is experimentally found to be greater than 0.001 at modified Reynolds numbers greater than 100 so that the first term only of the infinite series solution is needed.

The most exhaustive measurements reported are those of Molino and Hougen (34). The reported values differ from those of other workers largely due to the difference in the definition of \( k_e \). The effective conductivity as defined by Molino and Hougen is based on the assumption that only the void space is available for heat transfer.

Adopting the concept of total bed volume and assuming that the mean void volume in Molino and Hougen's work was 0.4, the equation given by these workers becomes:

\[
\frac{k_e}{k_g} = 1.23 \left( Re_m \right)^{0.43} = 1.23 \left( \sqrt{a_p} \right)^{0.43} \quad \text{(Eq. 28)}
\]

where the characteristic length of \( Re_m \) is the square root of the surface area of the solid particle, \( \sqrt{a_p} \).

Calderbank and Pogorski (10) found the functional relationship suggested by Molino and Hougen to be a satisfactory
method for correlating the results for overall equivalent thermal conductivity. Their work tended to substantiate the fact that the tube diameter and particle diameter also influenced the k values considerably.

Hougen and Piret (22) collected extensive data on the cooling of air during downward flow through radially cooled beds of granular solids over a 50-fold range in mass velocities and initially varying air temperatures. They presented generalized correlation in terms of a modified Reynolds number in which the characteristic length was the square root of the particle surface area and the viscosity was evaluated at the mean temperature of the packed bed. As reported by Molino and Hougen (34), these workers did not observe any effect of bed diameter and particle size.

Argo and Smith (1) have developed a method for predicting the effective thermal conductivity based on the proposed mechanisms suggested by Singer and Wilhelm (44).

For the case of packing materials of high thermal conductivity the calculation of effective thermal conductivity proposed by Singer and Wilhelm may involve several terms of the solution to Equation 1 while that of Argo and Smith (1) remains relatively simple.
Point effective thermal conductivity

Some of the earliest evaluations of point effective thermal conductivity were presented by Bunnell et al. (8). These workers determined point values of the conductivity in a 2-inch reactor packed with 1/8-inch alumina cylinders over a 3-fold range in gas mass velocities. The values of thermal conductivity were depressed at the center of the pipe and increased with increasing radial position until the wall was approached where the values decreased rapidly. The magnitude of the effective thermal conductivity was about 10 times that of the fluid used, air, and the increase in effective thermal conductivity over the value at static condition, no flow of fluid through the bed, was observed to be directly proportional to the mass velocity of the gas. The predicted average $k_e$ was correlated as:

$$\frac{k_e}{k_g} = 5.0 + 0.061 \frac{D_p G}{\mu}$$  \hspace{1cm} (Eq. 29)

It was observed experimentally that there was no appreciable change in $k_e/k_g$ with respect to radial position until the tube wall was reached. However, there was a sizeable and inconsistent variation of $k_e/k_g$ with the packed bed depth.

Hall and Smith (20) postulated two effective thermal
conductivities, one which would represent the heat transfer resistance between bundles of gas and another which would represent the heat transfer resistance of the solid pellet and the gas region between pellets. The experimental gas and pellet temperatures, under conditions of no reaction, were used in the differential equations representing the two separate temperature distributions to calculate point values of each thermal conductivity for various positions in the packed bed. These workers observed large variations between the corresponding values of each of the postulated thermal conductivities and no consistent trend, which they attributed to differences between the gas and pellet temperatures. The difficulty in measuring these temperatures accurately and the fact that the difference between the gas and pellet temperatures was not large except near the wall of the tube suggested the use of an overall effective thermal conductivity for the packed bed. An additional factor which seemed to justify the use of a single value for the gas and the pellet temperatures, with a single value for the effective thermal conductivity was the fact that the experimental temperature curves for both the gas and catalyst temperature versus position had the same general shape. The values of \( k_e \) determined from the experimental gas and catalyst temperature profiles did show a decreasing trend with increase in radial position.
Irvin, Olson, and Smith (27) calculated the values of $k_e/C_p G$ by graphically evaluating the first and second order temperature differentials and substituting them into the differential equation representing the temperature distribution. Since the accuracy of these slopes could not always be determined with precision the $k_e/C_p G$ values calculated by this method were used in the numerical solution of the differential equation and the values adjusted until the temperature distribution obtained by the numerical solution agreed with the experimental temperature distribution.

Coberly and Marshall (12) also determined point values of $k_e$ by measuring the temperature gradients in an air stream being heated while flowing through a packed bed. Values of $k_e$ were calculated for three different particle sizes at various bed depth for a range of mass velocity from 175 to 1215 lb/(hr)(sq ft). They averaged the point values arithmetically across the diameter to arrive at an average overall effective thermal conductivity.

The authors reported that the experimental temperature gradients could be recalculated with good accuracy by using such an average value of $k_e$.

Radial heat transfer data in terms of effective thermal conductivities as they appear in a modified thermal diffusivity, $k_e/C_p G$, are presented by Schuler et al. (41). The data are presented for 1/8-, 3/16-, and 1/4-inch cylindrical
pellets in a 2-inch I.D. tube through which air was flowing at mass velocities from 150 to 750 lb/(hr)(sq ft). An approximate breakdown into separate contributions due to radiation, solid-solid conduction, and eddy transfer is also given. On the assumption that (a) the pressure drop through the bed does not vary with radial position and (b) the pressure drop in a non-isothermal bed is equal to the pressure drop in an isothermal bed operating at the same bulk mean temperature, the effect of the non-isothermal bed compared to that of an isothermal bed on the mass velocity is very small.

The values of $k_e/C_p G$ group were computed from the temperature data using Equation 27. The method of solution consisted of two steps: (a) an approximate value of $k_e/C_p G$ was obtained by differentiating the temperature profiles and substituting into Equation 27 and (b) the approximate values were improved by using them in a numerical solution of Equation 27 to reproduce the measured temperatures. This approach served to minimize the errors introduced by graphical differentiation of the temperature data. By this method it is possible to obtain $k_e/C_p G$ directly without the necessity of assuming uniform mass velocity across the tube.

These authors also determined the effect of assuming that the $k_e$ is constant across the diameter of the tube in Equation 27. If the term,
which is neglected in Equation 1, is included in the analysis, the values of $k_e/\rho C_p G$ may be determined using the velocity data of Morales and Spinn (35).

Calderbank and Pogorski (10) have measured effective conductivities for alundum and celite from experimental temperatures by the method of Coberly and Marshall (12) and correlated them as a linear function of a modified Reynolds number. Plautz and Johnstone (37) have also reported their data in a similar form.

Kwong and Smith (29) determined bed effective thermal conductivities by two methods: (a) the partial differential equation is numerically integrated to obtain the temperature profile by assuming a radial distribution of thermal conductivity values and using the known boundary conditions, and (b) assume the solution of the partial differential equation can be expressed as the product of two solutions according to the method of separation of variables. The separation constant is found to be a series of eigenvalues which are determined from the eigenfunctions from the known boundary conditions. The final solution is in the form of an infinite series. For large bed depths only the first term of the series is important and only the first eigenvalue need be determined. This method has the advantage over the
graphical procedure which involves the graphical determination of the second derivative of temperature versus radial position in the bed. It was found that it was usually necessary to measure temperature profiles for at least four bed depths to determine satisfactory values of $\frac{\partial T}{\partial z}$.

To obtain agreement between the experimentally observed values of the effective conductivity at high flow rates and those predicted theoretically by the work of Ranz (38) and Baron (3) it appears that a value of $Pe_{TD}^T = 7$ instead of the value of roughly 10 would be more acceptable.

Kwong and Smith (29) observed that the effective conductivity varied across the diameter of the tube, with the maximum value in some cases as much as 20% greater than that at the center of the tube as the radial position increased. These workers also noted a rapid decrease in the effective conductivity near the wall of the tube. The values of the thermal conductivity of different packing seemed to have only a minor effect on the effective conductivity.

Applying both the mechanism of lateral mixing proposed by Ranz (38),

$$k_{eTD} = \alpha GC_p/N,$$  \hspace{1cm} \text{(Eq. 30)}

where

$\alpha = \text{mass velocity of fluid flowing in the direction of}$
heat or mass transfer divided by the mass velocity of fluid based on cross-sectional area of empty tube in the direction of fluid flowing

\[ N = \text{number of solids in the unit length of packed bed, measured in the direction of heat flow,} \]

and their own assumptions, Yagi and Kunii (49) obtained theoretical formulas for the effective conductivity in terms of a static conductivity, for beds which contain a motionless fluid. Their final equation is given as:

\[
\frac{k_e}{k_g} = \frac{k^0}{k_g} + (\alpha\beta)/(D_p C_p G/k_g), \quad (\text{Eq. 31})
\]

where \( \beta \) is determined from the characteristics of the packing. The values of \( k^0/k_g \) were calculated from an equation developed earlier by Yagi and Wakao (50).

Yagi and Kunii (49) have also determined experimental values of the thermal conductivity using an experimental heater with an annular packed bed, wherein the heat flowed purely radially. The expression developed, Equation 31, by these workers was found to be of the same form as that obtained for cylindrical beds. For this geometry the values of \( (\alpha\beta) \) were correlated with the ratio of \( D_p/D_e \), where \( D_e \) is the equivalent diameter of the annular concentric cylinders.
Wall heat transfer coefficient

An equivalent wall heat transfer coefficient, $h_w$, has been introduced by many investigators as a means for presenting experimental data. It should be noted, however, that most all of the work on heat transfer within packed beds has shown that the resistance to heat flow is a property that is distributed throughout the bed rather than being concentrated at the container wall.

Following this general idea, Singer and Wilhelm (44) have expressed their analysis for heat transfer in a packed bed in terms of $h_w$ in the following way:

$$h_w = 5.79 \frac{k_e}{D_T} + 0.092 \frac{C_p G D_T}{L} \quad \text{(Eq. 32)}$$

which indicates a dependency of the overall heat transfer coefficient upon $D_T$, $D_p$, and $L$. This form of the equation is similar to those obtained by previous workers from dimensional analysis and experimental data, such as Colburn (13) and Leva (32).

The early work of Colburn and Leva was later extended by Chu and Storrow (11). The latter workers pointed out that the effect of column length was important. Unfortunately, they worked with tubes of small diameter and when the equations proposed by them were applied to tubes of 4-
or 5-inch diameter, impossibly high heat transfer coefficients result.

Steady state heat transfer experiments were carried out by Baumeister and Bennett (4) in a four-inch transite tube packed with 3/8-, 1/4-, and 5/32-inch steel spheres. Heat was generated in the pellets by means of a high-frequency induction coil surrounding the test section. The average heat transfer coefficient between the bed of spheres and a stream of air passing through the bed were correlated in the form:

\[ j_H = \frac{h}{C_p G} \left( \frac{k}{\mu} \right)^{1/3} = aRe^b \quad \text{(Eq. 33)} \]

It was found that a separate equation was needed for each particle size. \( a \) and \( b \) represent the effect of particle and tube size. An attempt was made to predict packed bed transfer rates from the rates for single particles after the method proposed by Ranz (38). Agreement was fair for low Reynolds' numbers but it was quite low compared to experimental data over the whole range of Reynolds' numbers.

The calculation of point thermal conductivity by evaluation of temperature profiles in a packed bed lead to the conclusion that the resistance to heat transfer in the vicinity of the retaining wall is quite different from that
in the main body of the tube. This phenomenon has led many investigators to determine a wall film heat transfer coefficient, $h_{wf}$. The overall wall heat transfer coefficient, $h_w$, is related to the wall film coefficient, and the effective thermal conductivity by

$$\frac{1}{h_w} = \frac{1}{k_e} + \frac{1}{h_{wf}}$$

(Eq. 34)

The experimental data of Coberly and Marshall (12) showed that the resistance to heat transfer in the gas film at the inside of the tube wall was not negligible. A mathematical analysis of the longitudinal temperature gradients enabled values of $h_{wf}$ to be determined from their final expression for the wall film coefficient, in terms of the gas mass velocity,

$$h_{wf} = 2.95 \, G^{0.33}$$

(Eq. 35)

Based on their experimental data, these authors have reported that substantially all of the resistance to heat transfer occurs at the wall when

$$\frac{D_p \, G}{\mu} > \text{about } 10^4.$$

Similar expressions have been reported by Calderbank
and Pogorski (10), Hanratty (21), and Flautz and Johnstone (37).

From experimental results using annular packed beds Yagi and Kunii (49) have reported the following equation for gases:

\[
\frac{h_{w_f} D_p}{k_g} = \frac{h^0_{w_f} D_p}{k_g} + \alpha_w PrRe' \quad \text{(Eq. 36)}
\]

where \( \alpha_w \) is the mass velocity of the fluid flowing in the direction of heat or mass transfer divided by mass velocity of the fluid based on the sectional area of empty tube in the direction of fluid flowing, \( h_{w_f} \) is the wall film coefficient of heat transfer on the surface of the inner tube in an annular packed bed, \( h^0_{w_f} \) is defined as before but for a stationary gas. The values of \( \alpha_w \) are larger for cylindrical beds than for annular beds. Also the values of \( h_{w_f} D_p / k_g \) differ; these differences are attributed to differences in the packing states for the particles.

**Stagnant thermal conductivity**

Kunii and Smith (28) have developed a theoretical model which is designed to be applicable both to packed beds and to beds of consolidated particles. This correlation is a function of the thermal conductivity of the solid and fluid
phases, void fraction, and if radiation is important, emissivity, mean temperature, and the diameter of particles. The correlations make use of a packing function which is developed on the basis of various possible geometrical configurations of spherical packing. The possible heat transfer mechanisms are based on those proposed by Wilhelm and Singer (44).

**Radiation in packed beds**

Schotte (40) has used a model similar to that of Argo and Smith (1) to develop an independent expression for radiation heat transfer between solid particles. This effect appears to be more important at high temperatures (above 400°C) particularly when the particles are large. The thermal conductivity of the bed is first predicted by correlations of bed conductivity versus gas and solid conductivity which also includes the effect of lower pressures on gas conductivity when the mean free path of the gas molecules is of the same order as the distance between particles. The radiation effect is additive. The experimental data of Yagi and Kunii (49) fit closely the predicted values.
Porosity in packed beds

Radial variations of porosity were investigated by Roblee et al. (39). For highly irregular shapes, i.e., Beryl saddles, results by these workers indicate that the void fraction decreases regularly from one at the wall to the average porosity at about one particle radius from the wall. For regularly shaped particles, i.e., spheres and cylinders, cycling was observed for more than two particle diameters into the bed, the amplitude decreasing as the distance from the wall was increased. The maxima and minima were observed at integral multiples of the particle radius. For Rashig rings a hump in the curve was observed at about 1/2 particle radius from the wall. The void fraction then decreased to its average value at one particle radius and then remained constant.

The average void fraction in packed beds have been determined by Calderbank and Pogorski (10) and by Leva (32) by the usual displacement methods. The experimental values of void fraction or porosity are correlated as a function of D_p/D_T; however, there is usually a considerable scattering of the data.

Schwartz and Smith (43) have determined the manner in which the void fraction varies with radial position near the container wall from experimental velocity profiles. The
void fractions were calculated by a numerical stepwise integration procedure using a semi-theoretical equation for the velocity in 2-, 3-, and 4-inch pipes packed with particle sizes ranging from 1/8- to 1/2-inch diameter. These data indicate a minimum value for the void fraction in the center of the pipe which does not seem to depend on the value of $D_p/D_T$. This minimum value for the void fraction does not change appreciably with a change in radial position of about 50%. As the wall of the pipe is approached, the void fraction tends to increase approximately exponentially to a theoretical value of 1.0 at the wall.

**Velocity distribution**

Morales et al. (35) using a circular, hot wire annemometer measured the point velocity in a standard 2-inch pipe packed with cylindrical pellets 1/8- and 3/8-inch diameter. These results showed a maximum near the wall, with a sharply decreasing value as the wall was approached more closely. The peak velocity was observed to be as much as 100% greater than the value at the center of the pipe. Similar gradients were observed for all three packing sizes, the smallest of which corresponded to a $D_T/D_p = 16$.

Schwartz and Smith (43) obtained data in 2-, 3-, and 4-inch standard pipes using 1/8-, 1/4-, 3/8-, and 1/2-inch
spherical and cylindrical pellets, corresponding to a range of \( D_T/D_p \) from 5 to 32. In order to include the range of flow rates encountered in commercial reactors, experimental velocities of 0.42 to 3.5 feet per second corresponding to 114 to 1110 pounds per hour per square feet, respectively, were investigated.

They observed a peak velocity at approximately 1 pellet diameter away from the pipe wall. For \( D_T/D_p > 30 \), this maximum velocity ranged from 30 to 100% greater than that at the center of the tube. Results indicated unless \( D_T/D_p \) is greater than the about 30, important velocity variations exist across a packed bed. Such variations would be important in analyzing the operation of catalytic reactors and packed bed heat exchangers.

Mass Transfer

Turbulent motion can arise in several different ways. In the case of a fluid flowing around a bluff object such as a cylinder or sphere, a boundary layer is formed next to the surface of the object, the fluid velocity varying in this layer from zero to the surface to the main stream velocity at the outer edge of the boundary layer. As the Reynolds number is increased further, eddies are continuously shed and reformed at the rear of the object, and the free eddies
maintain their form for some distance downstream, finally breaking up into an irregular motion. At still higher Reynolds numbers, the boundary layer itself becomes turbulent before it leaves the surface. For straight pipe or along flat surfaces, turbulence is generated within the boundary layer itself, as in the case of the bluff object at high velocities. The turbulence thus generated replenishes the energy of the turbulent core, which is being continuously dissipated by energy interchange within the core. Since turbulent heat transfer and turbulent material transfer take place by the same mechanism, the eddy diffusivity and the eddy thermal diffusivity are often assumed to have the same value.

From theoretical considerations, Baron (3) has predicted that the Peclet number should be between 5 and 13. The basis for this prediction is the so-called "random walk" theory in which statistical considerations are employed. The motion of a particle suspended in a fluid as it approaches a pellet is seen to suffer a lateral deflection, $\beta D_p$, where $\beta$ is of the order of one-half. After the particle has progressed a distance $z$, in the direction of flow, it has undergone $n = \gamma z/D_p$ deflections where $\gamma$ is approximately unity. The mean square deviation is

$$x^2 = n\beta^2 D_p^2 = az\beta^2 D_p$$  \hspace{1cm} (Eq. 37)
From the Einstein equation

\[ x^2 = 2Et \]  \hspace{1cm} (Eq. 38)

where \( t \) is the time corresponding to \( x^2 \). In the present case \( t = z/v \). Combining these relations we have

\[ Pe' = \frac{D_p v}{E} = \frac{2}{Y\beta^2} \]  \hspace{1cm} (Eq. 39)

In a typical bed, while \( Y \) is approximately unity, the value of \( \beta \) should range between 0.4 and 0.7. The predicted range of Peclet number is, therefore, between 5 and 13.

The random walk principle has been considerably extended by Latinen (30). He demonstrates the applicability of this theory to a body-centered cubic arrangement and for fully developed turbulence arrives at a value for the Peclet number of 11.3. However, no allowance is made for variation of velocity and void space with radial position.

Applying a different approach, Ranz (38) obtained a value of 11.2 for the Peclet number for a system of spherical particles packed with their centers at the corners of tetrahedrons.

Liquid phase experiments were performed by Bernard and Wilhelm (6) with a methylene blue solution diffusing from a point source into water flowing through a bed of packed
solids and gas phase experiments with CO₂ diffusing into an air stream in the bed. The basic differential equation was solved for the special case when the tube wall provides a boundary condition. Aqueous experiments were performed in a 2-inch tube for 1, 3, 5, 8-mm spheres, 3/16 x 3/16-inch and 1/4 x 1/4-inch cylinders; 1/8-inch cubes, 10-14 mesh granules; and a mixture of 1- and 3-mm spheres. Gaseous experiments were performed in an 8-inch tube with 3/8-inch spheres. Eddy diffusivities were correlated in terms of the modified mass Peclet number, \( Pe' = D_p v/E \), as a function of the modified Reynolds number, \( Re' = D_p G/\mu \). The experimental range of the Reynolds number was from 5 to 2,400.

These authors determined diffusivities directly by solution of the partial differential equation using a point source for the tracer material using the analytical solution for heat transfer. In addition to the assumption of constant properties for the system, it is necessary to assume plug flow or no radial variation in the velocity. When the mass Peclet number was plotted against the logarithm of the modified Reynolds number for four sizes of spherical packing used in the water apparatus, two regions were indicated for each size of packing. One region \( Re' > 40 \) was characterized experimentally by regular concentration profiles, the analysis of which led to reliable values of the eddy diffusivity. The second region \( Re' < 40 \) was characterized by
irregular concentration profiles leading to uncertainty in the values of the diffusivity. In this region it was observed, experimentally, that large eddies, with diameters several times that of the packing existed in the bed. The slope of the line through the experimental points was slightly negative for the smallest packing and increased through zero to small positive values as \( \frac{D_p}{D_T} \) was increased. The data indicated the break between the two regions moved forward with increasing values of the diameter ratio, and that the advance in position occurred roughly in direct proportion to \( \frac{D_p}{D_T} \).

The same plot for four types of non-spherical packing used in the water apparatus indicated there was no regularity in the shape of the curves in the broken-line region, but the solid lines in the stable region of higher Reynolds numbers all had small slopes, as with the spheres, and these slopes are about the same function of \( \frac{D_p}{D_T} \) as in the case of spherical packing. Also lines drawn for the non-spherical packing were displaced vertically downward toward the lower values of the Peclet number when compared to the results for spherical packing, and the displacement increases as the particle share deviates more and more widely from the spherical. The stable region was defined as being above a Reynolds number,

\[
Re' = 2150 \left( \frac{D_p}{D_T} \right) \quad (Eq. 40)
\]
For 3/8-inch spheres in the air apparatus, the curve through the experimental data has about the same slope as for mixed 1 mm and 3 mm spheres in the water apparatus, but is displaced downward about 15% or a difference in the Peclet number, \( \text{Pe'} = 2.0 \).

Experimental pressure drop data plotted in terms of a friction factor versus a modified Reynolds number showed that there was no sudden onset of turbulence beyond a certain value of the Reynolds number in a packed bed compared to the case for an empty pipe, which shows a sharp break across the transition region between viscous and turbulent flow. Observed pressure drops were correlated satisfactorily by the use of \( \frac{D_p}{D_T} \) having only a minor effect, so that turbulence in packed beds appears to be connected with the packing size and is probably interstitial in character. The fact that the unstable region in the plot of friction factor versus Reynolds number moves forward in proportion to \( \frac{D_p}{D_T} \) indicates that if the Reynolds number had been based on \( D_T \) instead of \( D_p \), these curves would have been pulled together.

Bernard and Wilhelm (6) state the above is an indication that the large eddies are a function of \( D_T \) rather than \( D_p \); that is, they are controlled by the wall of the containing tube, rather than the packing itself.

These workers present a highly simplified approach to stable turbulence in which it may be assumed that for any
particular shape of packing, geometric similarity always is maintained, regardless of the value of the modified Reynolds number. This means that the scale of turbulence is always proportional to the particle size and that the root mean square deviating velocity is proportional always to the velocity within the bed. This results in the prediction of a constant modified Peclet number at high Reynolds number which fits well with the experimental results for the turbulent region.

Plautz and Johnstone (37) have calculated eddy mass diffusivity from concentration data taken in an 8-inch tube packed with 1/2- and 3/4-inch glass spheres. The analysis was based on the assumptions: (a) there was no radial or axial variation in the velocity throughout the bed, and (b) the total diffusivity did not vary with radial position or bed depth. Superficial mass velocities ranged from 110 to 1,640 lb/(hr)(ft$^2$) corresponding to modified Reynolds numbers of 100 to 2,000. Air was the main stream fluid in this experiment. The modified Peclet number for mass transfer, $D_p \bar{V}/E_{TD}$, was found to have a constant value of about 12 in the region of fully developed turbulence. At lower Reynolds numbers, this group varied with the flow rate. Values of $E_{TD}$ and $Pe'$ were calculated for isothermal and non-isothermal packed beds with little or no significant variation observed.

Fahien and Smith (17) studied the effects of pipe and
packing sizes on mass diffusivities and also the effect of radial position in the bed. Measurements were made by introducing CO₂ tracer material into an air stream and the resulting mixtures were analyzed at various positions in the bed downstream from the point of injection. Data were taken using pipe sizes of 2-, 3-, and 4-inch which were packed with 5/32-, 1/4-, 3/8-, and 1/2-inch nominal diameter particles. The results were determined using an IBM calculator for the computation.

The results showed that the modified Peclet number increases from the center of the pipe toward the wall and that the increase is significant when $\frac{D_p}{D_T} > 0.05$. The variation in Peclet number seemed to be primarily dependent on the radial variation in porosity or void fraction of the packed bed. For modified Reynolds numbers above 40 to 100, these workers have developed the following equation for the radial variation of the Peclet number in terms of the corresponding void fraction which is valid for 81% of the radius of the bed:

$$\text{Pe'} = 8.0 + 100 (\varepsilon - \varepsilon_o),$$  \hspace{1cm} (Eq. 41)

where

$\varepsilon_o$ = void fraction at the center of the bed,

$\varepsilon$ = point void fraction, at any radial position.
At radial positions of \( r/r_w > 0.81 \), wall friction influences turbulence conditions and the Peclet number. Average values of the Peclet numbers were correlated by the equation

\[
Pe = (11.7) \, Re^{-0.06}(1 + 19.4 \, \frac{D_p}{D_T}),
\]

(Eq. 42)

where \( Re \) is a Reynolds number based on pellet size. The average values of the Peclet number as calculated by Equation 42 appeared to be in general agreement with those obtained by other investigators.

Dorweiler and Fahien (16) investigated mass transfer in packed columns using a tracer-injection technique to determine mass diffusivity and Peclet number as a function of radial position for flow rates in the laminar and transition regions. Gas velocity distributions were determined with a five-loop, circular hot-wire anemometer. The data were obtained using a 4-inch pipe, packed with 1.4-inch spherical, ceramic catalyst support pellets for eight mass velocities ranging from 50.2 to 985 lb/hr ft\(^2\). Average diffusivities and Peclet numbers were calculated from the solution of the differential equation representing the mass transfer which was an analytical solution in terms of Bessel functions. The values of point diffusivity and Peclet number were determined using a semi-numerical technique developed by Fahien (16). It was reported the eddy diffusivity could be
expressed by the equation

\[ E = D_m + 7.80 \cdot v^{1.12} \left( 1 + \sigma^{1.7} - 2\sigma^{3.4} \right) \]  
(Eq. 43)

The results on velocity profiles reported by these workers substantiates the findings of Schwartz (43) and Molino (34). The point diffusivity and Peclet number was found to be constant at the center of the bed, increased as the column wall was approached, reached a maximum near the wall and finally decreased at the wall.

These workers concluded that mass transfer in packed beds can be described as consisting of a parallel molecular and eddy transfer mechanism. The molecular contribution is fixed for a given system and is the only mechanism at low Reynolds' numbers with the eddy contribution (considered on a point basis) varying with the local flow conditions and correlates according to Equation 43. The variation of eddy diffusivity can be explained on the basis of velocity and void space variation within the system.

Aris and Amundson (2) investigated the effect of longitudinal mixing in fixed beds using as a model a bed packed with spheres in rhombohedral blocked-passage arrangement. The bed was visualized as being formed by stacking the spheres in layers so that there would be planes in the bed, normal to the flow, through which the velocity would be very
high followed by corresponding planes where the velocity would be considerably less. It was assumed that the free volume in each layer, the layer between the planes passing through the sphere centers, served as mixing cells for the fluid. The fluid entering a given cell would act as a jet mixer which would increase the turbulence in free volume. It was further assumed that there was no lag in the fluid flow from one cell to the next. These workers considered a series of well agitated mixing cells which, when extrapolated to an infinite number of cells represented a packed bed, predicted an axial Peclet number of 2 for high Reynolds numbers.

This representation of a packed bed has the advantage that it could be one component of an open loop computer control system for a non-linear fixed bed process. However, the introduction of a reaction rate term increases the mathematical complexity such that this representation has no advantage over the differential equation representation.

Kinetics of Gas-Solid Reactions

Hougen (22) has presented an extensive review of catalysis, solid-gas reactions, and the effect of the properties of catalysts on reaction in fixed bed reactors. In order of decreasing importance, five of the more important sources of error in the evaluation of kinetic models and
interpretation of rate data in catalytic, gas-solid, flow reactions are given by Hougen as:

1. Variation in catalyst activity.
2. Use of catalyst particles having effectiveness factors differing appreciably from unity.
3. Neglect of external resistances to mass and heat transfer. These effects are especially difficult to predict and evaluate when large gradients are present.
4. Apprreciable departure from plug flow.
5. Neglect of pressure drop due to flow.

The effect of varying catalyst particle size, which has a marked effect on the pressure drop, heat transfer, and catalyst activity, was also reviewed.
EXPERIMENTAL REACTION DATA, CONVERSIONS, AND TEMPERATURES

Source of Experimental Data

Experimental reaction rates for the oxidation of $\text{SO}_2$ to $\text{SO}_3$ by air for a 1-1/2 inch tubular stainless steel reactor were determined in this differential reactor and were reported by Hall and Smith (20). The reactor was packed with 1/8-inch cylindrical alumina catalyst pellets impregnated with 0.2 weight percent platinum. The bed depth was equivalent to two layers of catalyst, and the operating conditions were:

- gas mass velocity: $350 \text{ lb/hr ft}^2$
- temperature: $350$ to $475^\circ\text{C}$
- composition:
  - (a) before preconversion: 6.4 mole percent $\text{SO}_2$, 93.6 mole percent air
  - (b) preconversion range: 0 to 70 percent

The differential reactor was immersed in a constant temperature bath maintained at approximately $100^\circ\text{C}$ by boiling water. A preconverter tube containing 8 inches of platinum on alumina catalyst pellets was placed in the line before the gas entered the reactor proper to accomplish the desired amount of preconversion.
Integral reactor data for the oxidation of $\text{SO}_2$ to $\text{SO}_3$ using air were reported by Hall and Smith in the same work. The reactor consisted of a 2-inch stainless steel pipe, 24 inches long, packed with 8 inches of platinum on alumina catalyst. Several inches of dummy (plain alumina) pellets were placed on top of the active catalyst to eliminate a sudden change in the flow pattern and reduce heat transfer from the top of the catalyst bed to the walls by radiation. There was a flange or screen to hold the catalyst at the inlet but there was no indication of any prepacking. The reactor was operated for the following conditions:

(a) an inlet $\text{SO}_2$ concentration of approximately 6.4 mole percent $\text{SO}_2$

(b) gas mass velocity of approximately 350 lb/hr ft$^2$

(c) a constant wall temperature of approximately 100°C, maintained by a jacket containing boiling water which surrounded the reactor

Temperature data were reported for varying bed depths of 0-, 2-, 4-, 6-, and 8-inches and the mean conversion was obtained by analyzing the exit gases from the reactor. Both catalyst pellet temperatures and gas temperatures were obtained by placing thermocouples in the bed at a number of radial positions across the tube. Additional temperature and conversion data, for approximately the same operating conditions, were
reported by Irvin, Olson, and Smith (27) using essentially the same experimental equipment and procedure developed by Hall and Smith. These workers extended the original work for a number of other mass velocities and improved the earlier design procedure. It was noted by Irvin, Olson, and Smith that the bed depth at 0 corresponded to an actual distance from the reactor bed entrance of 0.063 inch where this distance corresponded to the position of the thermocouple junction imbedded halfway through pellets of catalyst in the lowest layer.

Experimental data for the oxidation of $SO_2$ to $SO_3$ using air were reported by Schuler, Stallings, and Smith (41) using the experimental equipment developed by Bunnell et al. (8). Temperatures were obtained for a 2-inch tubular reactor 38 inches long packed with 1/8-inch cylindrical alumina catalyst pellets coated with 0.2 weight percent platinum at bed depths of 0, 0.53, 0.88, 1.76, 4.23, and 5.68 inches. The pellets were supported in the tube by two stainless steel screens held by flanges, and the procedure was modified to include four inches of prepacking in the reactor. The temperatures were reported using thermocouples imbedded in the catalyst pellets. The mean conversion data were obtained by analyzing the exit and entering gases for $SO_2$ content for various bed depths. The entering gas composition was held nearly constant at 6.45 mole percent $SO_2$ and 93.55 mole percent air.
The reactor wall temperature was maintained at an average value of 197°C by surrounding the reactor tube by a bath of boiling glycol.

No Reaction

Temperature data were reported for air alone flowing through a 2-inch packed bed reactor with the wall temperature maintained at 100°C for a mass velocity of 350 lb/hr ft² by Hall and Smith (20). The temperatures were measured at the reactor entrance in addition to bed depths of 2, 4, 6, and 8 inches, and the air entered the packed bed after being heated to approximately 400°C. Additional data for the same mass velocity and wall temperature were obtained using the same reactor obtained by Irvin, Olson, and Smith (27) for heated air alone. The smoothed experimental temperature distributions for various bed depths, as shown in Figures 37 and 38 in Appendix A, represented the average of both catalyst pellet and gas temperatures.

Schuler, Stallings, and Smith (41) used the same reactor which was used to obtain reaction temperatures and conversions to get temperature data for heated air flowing at a mass velocity of 350 lb/hr ft². The air was heated to approximately 400°C before entering the reactor which was immersed in a boiling glycol bath to maintain the wall
temperature constant at approximately 197°C. These workers presented radial temperature variations for a single bed depth of 0.354 feet and the reactor entrance. These temperatures represented catalyst pellet temperatures since the thermocouples were imbedded in the 1/8-inch cylindrical pellets and sealed with water glass. The smoother distribution was obtained using the data available as shown in Figure 46 in Appendix A.

Initial Temperature Distribution

The experimental initial temperature distribution for the case of 100°C wall temperature was obtained from the data reported by Hall and Smith (20) and Irvin, Olson, and Smith (27) for heated air flowing through the reactor. Both catalyst pellet and gas temperatures were measured at the entrance to the packed bed (zero bed depth) or at an actual bed depth of 0.063 inch. The mass velocity of air was maintained at the same value as that for the reaction gas mixture. The data were used to obtain a smoothed temperature distribution as shown in Figure 39 in Appendix A. Hall and Smith (20) also obtained radial temperature distributions at the reactor entrance for the case of reaction occurring in the bed by measuring both the catalyst pellet and gas temperatures. The center catalyst temperature was approximately
15°C higher than the average temperature for the case of no reaction, but the catalyst temperatures for both cases appeared to have the same value after a radial position of \( r/r_w = 0.7 \) was reached. However, by comparing the smoothed or averaged temperatures for the case of reaction and no reaction it can be seen from Figure 39 in Appendix A that the former exceeds the latter at the center of the tube and the reverse is true at radial positions of \( r/r_w > 0.5 \).

Temperature for air flowing through a similar reactor at a mass velocity of 350 lb/hr ft² with a constant wall temperature of approximately 197°C was reported by Schuler, Stallings, and Smith (41). The temperatures were obtained using thermocouples imbedded in catalyst pellets, and the radial distribution at the reactor entrance was used to obtain a smooth initial temperature distribution as shown in Figure 46 in Appendix A.

Conversion

Hall and Smith (20) obtained the mean conversion for reactor bed depths of 2, 4, 6, and 8 inches. The conversion was calculated by analyzing gas samples for \( \text{SO}_2 \) content before and after the gas mixture passed through the reactor. Irvin, Olson, and Smith (27) reported additional data for a similar reactor operated under the same conditions and for
the same catalyst bed depths. The smoothed curve for mean conversion versus bed depth was obtained from the reported data as shown in Figure 40 in Appendix A.

Schuler et al. (41) reported mean conversion for catalyst bed depths of 0.53, 0.88, 1.76, 4.23, and 5.68 inches determined from a tubular reactor similar to that of Irvin et al. (27) operated at approximately the same conditions except for prepacking of the bed and a constant wall temperature of 197°C. The experimental data for conversion at the various bed depths were used to obtain smoothed data as shown in Figure 41 in Appendix A.

Reaction Temperatures

Experimental temperatures at various catalyst bed depths for an SO₂ tubular reactor operated at a constant wall temperature of 100°C were reported by Hall and Smith (20) and Irvin, Olson, and Smith (27). The data obtained by these workers were used for comparison with the temperatures calculated by the computer solution of the reactor design equations by taking the smoothed temperatures from the experimental radial distributions as shown in Figures 42, 43, 44, and 45 in Appendix A. These smoothed curves represent the temperature data, at a gas mass velocity of approximately 350 lb/hr ft² and an entering SO₂ composition of approxi-
mately 6.5 mole percent, reported by both of these sources. It should be noted that there were temperature differences of 20-30°C in some cases for the reactors although they were operated under the same conditions. Also both gas and catalyst pellet temperatures were used to obtain the smoothed experimental radial and longitudinal temperature distributions.

Experimental temperature data taken for a similar reactor operated at a constant wall temperature of 197°C were reported by Schuler, Stallings, and Smith (41). The experimental catalyst pellet temperatures at various bed depths were used to obtain smoothed radial temperature distributions as shown in Figures 46 and 47 in Appendix A.
DEVELOPMENT OF REACTOR DESIGN VARIABLES

Reaction Rate

Since the formation of \( \text{SO}_3 \) is a result of the reaction between \( \text{SO}_2 \) and \( \text{O}_2 \), probably on the surface of the catalyst, it is reasonable to assume that the partial pressure of \( \text{SO}_3 \) will be proportional to the conversion, \( x \), and can be expressed as

\[
P_{\text{SO}_3} = c_2 x \quad \text{(Eq. 44)}
\]

where \( c_2 \) is the proportionality constant. The \( \text{SO}_2 \) concentration of the entering gas mixture is small, less than seven mole percent; the remainder of the gas contains primarily oxygen and nitrogen whose concentrations will not vary appreciably throughout the reactor even though the conversion of \( \text{SO}_2 \) to \( \text{SO}_3 \) may be quite high. By expressing the partial pressure of \( \text{N}_2 \) and adsorbed \( \text{O}_2 \) as constant quantities by the constants \( c_3 \) and \( c_4 \), respectively, as

\[
p_{\text{O}_2} = c_3 \quad \text{and} \quad p_{\text{N}_2} = c_4 \quad \text{(Eq. 45)}
\]

the denominator of the theoretical rate equation
The $\text{SO}_2$ concentration may be expressed in terms of the conversion, $x$, and the initial $\text{SO}_2$ concentration, $y_0$, as

$$p_{\text{SO}_2} = C_5(1 - x)y_0$$  \hspace{1cm} (Eq. 48)

Expanding the denominator and substituting the equivalent expressions for the partial pressures, Equation 46 may be written as

$$R_A = \frac{kC_tK_0^{\frac{1}{2}}(p_{\text{SO}_2}p_{\text{O}_2}^{\frac{1}{2}} - \frac{1}{K}p_{\text{SO}_3})}{[1 + K_0^{\frac{1}{2}}p_{\text{O}_2}^{\frac{1}{2}} + K_{\text{SO}_3}p_{\text{SO}_3} + K_{\text{N}_2}p_{\text{N}_2}]^2}$$  \hspace{1cm} (Eq. 46)

can be written in the following form:

$$[1 + K_0^{\frac{1}{2}}c_3 + K_{\text{SO}_3}c_2x + K_{\text{N}_2}c_4]^2$$  \hspace{1cm} (Eq. 47)

Rearranging Equation 49, after collecting the terms involving the conversion, gives
The first term of the denominator of Equation 50 is a function of the initial $\text{SO}_2$ concentration and does not depend directly on the conversion; further, this term must represent the initial reaction rate corresponding to zero conversion. Therefore, Equation 31 may be written in a simplified form in the following manner:

$$R_A = \frac{R_A^0 - ax}{1 + bx + cx^2} \quad \text{(Eq. 51)}$$

where

$$R_A^0 = \frac{C_{1/2}K_{N2}C_3}{(1 + K_{O3}^2 + K_{N2}C_4)^2} v_o \quad \text{(Eq. 52)}$$

and

$$a = \frac{C_{1/2}K_{O3}C_3 v_o C_3 + C_{1/2}K_{O2}^2}{(1 + K_{O3}^2 + K_{N2}C_4)^2} \quad \text{(Eq. 53)}$$
No attempt was made to calculate each of the individual terms comprising the coefficients $R_A^0$, $a$, $b$, and $c$. Since $R_A^0$ is essentially a function of temperature, it should form an Arrhenius-type equation. The logarithm of the experimental reaction rate, for zero conversion, was plotted as ordinate against the reciprocal absolute temperature as abscissa. Although a single straight line gave adequate results over the entire temperature range, more accurate values were obtained by using two different lines for the high and low temperature range. The coefficients $a$, $b$, and $c$ were determined using the experimental reaction rates, and the resulting values were plotted against temperature. There appeared to be a definite interaction between coefficients $b$ and $c$, which was indicated by the fact that the $SO_3^-$ adsorption equilibrium constant appears, raised to a different exponent, in each of these coefficients. The
variations of $R_A^0$, $a$, $b$, and $c$ with temperature are shown in Figures 52, 53, 54, and 55, respectively, in Appendix A.

In order to utilize this information in a computer program, it is necessary to have the information in the form of an equation. A sub-routine was programmed on the Cyclone computer to calculate the reaction rate for any given temperature and conversion. The values calculated using Equation 51 may be compared with the experimental values of the reaction rate in Figure 2. The maximum deviation was about 8% and was generally about 2%.

In order to determine if the reaction rate equation was a valid expression for the reaction rate outside the range used to fit the coefficients, reaction rates were calculated for a temperature range of 350 to 680°C and a conversion range of 0 to 100%. The reaction rate versus temperature and reaction rate versus conversion are shown in Figures 3 and 4, respectively. Hougen and Watson (25) presented a plot of reaction rate versus temperature for the same reacting system but for a gas mass velocity of 600 lb/hr ft$^2$ and 3/8-inch cylindrical catalyst pellets. A comparison of the rates and the general form for rate versus temperature indicates that the semi-empirical rate equation can be used to predict rates over a wide range of conditions with reasonable accuracy. The least accurate calculated values for the rate are for conditions of low conversion ($x < 0.20$) above 600°C.
Figure 2. Comparison of experimental and estimated reaction rates for oxidation of sulfur dioxide
Conversion vs. Reaction rate, gm-moles catalyst hr

- 420°C
- 380°C
- 360°C
- 350°C

- O Calculated
- △ Experimental
Figure 3. Effect of temperature on reaction rate for the catalytic oxidation of $SO_2$ to $SO_3$. 
gm-moles/gm catalyst hr
Figure 4. Effect of conversion on reaction rate for the catalytic oxidation of $SO_2$ to $SO_3$
and for very high conversion \( (x > 0.95) \) for temperatures between 425 and 650°C. The computer sub-routine was designed to give the value of zero to the rate when the conversion reached 1.0 or 100% so that a conversion greater than 100% could not occur. The curves of reaction rate versus conversion at various temperatures show the correct trend as predicted from theoretical considerations except for a temperature range of 425 to 650°C where a small positive reaction rate is predicted at 100% conversion.

As a result of the experimental method used to obtain the rate data (20), Equation 51 can be expected to give reaction rates as a function of the bulk conversion or concentration and the catalyst temperature. For the mass velocity generally used the effect of diffusion may be considered to be included in the experimental values for the rate (36). Hougen and Watson (25) have shown that the correction for the temperature difference between bulk and catalyst surface should be small except at very high rates, corresponding to low conversion and high temperatures at the reactor entrance.

The fact that the rate equation developed from data for a limited range of temperatures and conversions, which was based on the theoretical equation for the \( \text{SO}_2 \) reaction, was very successful over almost the entire range of conversion and temperature possible lends additional support to the use of theoretical equations in place of arbitrary higher-order
polynomials when this type of information is available.

Velocity

Since a satisfactory equation could not be developed which would predict the radial variation of the gas velocity in a packed bed, the experimental data of Schwartz and Smith (43) were used. The data used were collected using air in a 2-inch tube packed with 1/8-inch cylindrical pellets for an average velocity of 2.68 feet per second compared to a reactor entering average velocity of 3.2 feet per second. The smoothed data are shown in Figure 48 in Appendix A as $(v/V)$ versus the dimensionless radial position, $r/r_w$. Since it is a necessary condition that the integrated average of $(v/V)$ with respect to the radius be equal to 1.0, the values of $v/V$ were adjusted until this requirement was satisfied. A sub-routine was written for the Cyclone computer which could be used to give point values of $v^*$, the smoothed point values being entered as tabular data.

Heat Transfer Peclet Number

The point values of the heat transfer Peclet number, $Pe_H$, were taken from the experimental data of Schuler, Stallings, and Smith (41). These data were obtained by
passing air, with a gas mass velocity of 350 lb/hr ft$^2$ and a
temperature of 400°C, through a 2-inch tube packed with 1/8-
ininch cyclindrical alumina catalyst pellets. The depth of
the active catalyst was six inches preceded by four inches
of dummy packing. The packed bed thermal diffusivity,
k_e/C_p, was estimated at each radial position by calculating
the first and second order temperature profiles, and then
the corresponding k_e/C_p-values were used in a numerical solu-
tion of the differential equation representing the temperature
distribution for no reaction and adjusted until the calcu-
lated and experimental profiles matched. The smoothed values
were taken from Figure 49 in Appendix A and put in tabular
form to be used by a sub-routine for the Cyclone computer
for the variable heat transfer Peclet number.

Mass Transfer Peclet Number

The point or variable mass Peclet numbers were calcu-
lated with the equation developed by Fahien and Smith (17),

$$Pe_M = Pe_{M_0} + F\sigma^m,$$

where the parameters, $F$ and $m$, are functions of $D_p/D_T$ as
given by these workers; $\sigma$ is the dimensionless radial posi-
tion. These equations were incorporated in a sub-routine
for the Cyclone computer which was designed to calculate the value of $\frac{Pe_M}{Pe_{M_0}}$ for a given value of $r/r_w$.

Effective Thermal Conductivity

The point effective thermal conductivities were taken from the experimental data reported by Kwong and Smith (29). These values were used because they were the only available data in which the velocity profile was taken into consideration. The data which approximated that of the $SO_2$ reactor were for the heating of air at a gas mass velocity of 300 lb/hr ft$^2$ in a 4-inch pipe packed with 1/4-inch alumina spheres. The smoothed data were obtained from a plot of experimental values of $k_e$ versus $\sigma$ as shown in Figure 50 in Appendix A. These workers obtained values for $k_e$ by considering the $k_e$ and $G$ values as functions of radial position and adjusting these values until the point $k_e$ values substituted into the differential equation representing the temperature distribution resulted in a temperature profile that matched the experimental profile. The smoothed values were entered as tabular data and a Cyclone sub-routine written that would extract the corresponding $k_e^*$ value for a given radial position.
Mass Eddy Diffusivity

The point mass eddy diffusivities for this system were based on the results of Dorweiler and Fahien (16). The experimental diffusivity data were determined for 1/4-inch spherical packing in a 4-inch tube and a gas mass velocity of 368 lb/hr ft² compared to 1/8-inch cylindrical pellets in a 2-inch tube and a gas mass velocity of 350 lb/hr ft² in the SO₂ reactor. However, the tube to pellet diameter ratio for both systems had the same value, $D_t/D_p = 16$, which is a more important criterion than either the tube or pellet size.

The calculated data of diffusivity versus dimensionless radial position are shown in Figure 51 in Appendix A. The smoothed data were used with a sub-routine in the computer program to give the point value of $E^*$ by entering the data in tabular form.

Average Heat and Mass Transfer Peclet Numbers

The turbulent diffusion contribution is a measure of heat transfer as a result of turbulent mixing of portions of the gas stream at different temperatures. Its value can be estimated from measurements of mass transfer, radially, by the same mechanism. The advantage of using mass-transfer data is that the transfer of mass radially in a packed bed
does not involve the series or radiation mechanism but is caused only by molecular conduction and turbulent diffusion. In addition the contribution of molecular conduction is small. Using the method of Argo and Smith (1) and the mass transfer correlations presented by Fahien and Smith (17) and Bernard and Wilhelm (6) gave a value of 4.4 for the average heat transfer Peclet number and a value of 9.6 for the average mass transfer Peclet number.

A second method of estimating the average Peclet numbers was to calculate the radial mean values from the point data. The average mass transfer Peclet number was formed by taking the equation point values of $\text{Pe}_M$ versus $r/r_w$ reported by Fahien and Smith (17) and averaging them with respect to the radius; the value of $\text{Pe}_M$ was found to be 11.1. The heat transfer Peclet number was determined by averaging the point thermal diffusivity data reported by Schuler, Stallings, and Smith (41) with respect to the radius; the value of $\text{Pe}_H$ determined in this way was 3.58.

Similar averages calculated from point mass eddy diffusivity reported by Dorweiler and Fahien (16) and point effective thermal conductivity data reported by Kwong and Smith (29) gave values of 14.7 and 3.02 for the mass transfer Peclet number and the heat transfer Peclet number, respectively.
The computer program was designed so that it could be used with variable gas density and heat of reaction; however, since the variation of the bed parameters was of more importance in this project average values were used. The gas density was found by assuming an average temperature of 400°C using the ideal gas law and neglecting the effect of the small amount of SO₂ in the entering reaction mixture. The average value of the heat of reaction throughout the packed bed was taken to be -22,700 cal/gram mole.

Use of Computer Program

The finite difference equation was programmed for the Cyclone computer using sub-routines to provide the calculation involved with the use of each of the parameters tested. The computer flow diagram is shown in Figure 56 in Appendix B.
RESULTS AND SUMMARY

Effect of Average Peclet Numbers

The conversion and temperature for various bed depths were calculated for four different combinations of average Peclet numbers:

(a) $Pe_H = 3.58$ and $Pe_M = 11.1$,
(b) $Pe_H = 3.58$ and $Pe_M = 9.6$,
(c) $Pe_H = 4.4$ and $Pe_M = 9.6$, and
(d) $Pe_H = 4.4$ and $Pe_M = 11.1$.

Effect on conversion

The results of these calculations can be compared with the experimental conversions in Table 1. It can be seen from Figure 5 that in all cases the experimental conversion rises to its maximum value much more rapidly than the calculated values. For a given heat transfer Peclet number of 3.58 the calculated mean conversion was increased slightly, 1.76%, by a decrease in the mass Peclet number; however, at higher heat transfer Peclet numbers the same decrease in the value of $Pe_M$ increased the conversion by 6.8%. It appeared that decreasing the mass transfer Peclet number increased the mass diffusion toward the wall which increased the mean
Table 1. Effect of average Peclet numbers on mean conversion for the oxidation of sulfur dioxide in a 2-inch packed tubular reactor for a wall temperature of 197°C

<table>
<thead>
<tr>
<th>Reactor length, feet</th>
<th>Pe_H=3.58</th>
<th>Pe_H=3.58</th>
<th>Pe_H=4.4</th>
<th>Pe_H=4.4</th>
<th>Experimental</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Pe_M=11.1</td>
<td>Pe_M=9.6</td>
<td>Pe_M=9.6</td>
<td>Pe_M=11.1</td>
<td></td>
</tr>
<tr>
<td>0</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>0.05</td>
<td>11.8</td>
<td>11.8</td>
<td>12.4</td>
<td>12.2</td>
<td>16.4</td>
</tr>
<tr>
<td>0.10</td>
<td>18.5</td>
<td>18.5</td>
<td>20.2</td>
<td>19.4</td>
<td>26.6</td>
</tr>
<tr>
<td>0.15</td>
<td>22.5</td>
<td>22.7</td>
<td>25.5</td>
<td>24.2</td>
<td>34.4</td>
</tr>
<tr>
<td>0.20</td>
<td>25.0</td>
<td>25.3</td>
<td>29.1</td>
<td>27.3</td>
<td>37.1</td>
</tr>
<tr>
<td>0.25</td>
<td>26.6</td>
<td>27.0</td>
<td>31.7</td>
<td>29.4</td>
<td>38.4</td>
</tr>
<tr>
<td>0.30</td>
<td>27.5</td>
<td>27.9</td>
<td>33.6</td>
<td>30.7</td>
<td>38.8</td>
</tr>
<tr>
<td>0.35</td>
<td>28.0</td>
<td>28.5</td>
<td>34.9</td>
<td>31.6</td>
<td>38.9</td>
</tr>
<tr>
<td>0.40</td>
<td>28.3</td>
<td>28.8</td>
<td>36.0</td>
<td>32.1</td>
<td>38.9</td>
</tr>
<tr>
<td>0.45</td>
<td>28.3</td>
<td>28.8</td>
<td>36.6</td>
<td>32.4</td>
<td>38.9</td>
</tr>
<tr>
<td>0.50</td>
<td>28.3</td>
<td>28.8</td>
<td>37.1</td>
<td>38.9</td>
<td>38.9</td>
</tr>
</tbody>
</table>

Conversion. At higher heat transfer Peclet numbers the decreased heat transfer away from the center of the tube appeared to increase the reaction rate which in turn caused greater conversion. The smallest deviation from the maximum
Figure 5. Effect of average Peclet numbers on mean conversion for a reactor wall temperature of 197°C
Mean Conversion, %

- o Experiment
- \( P_{e_M} = 9.6, P_{e_H} = 4.4 \)
- \( P_{e_M} = 9.6, P_{e_H} = 3.58 \)
- \( P_{e_M} = 11.1, P_{e_H} = 4.4 \)
- \( P_{e_M} = 11.1, P_{e_H} = 3.58 \)

Reactor bed
$\text{Pe}_H = 4.4$
$\text{Pe}_M = 9.6$
$\text{Pe}_u \approx 3.58$

- **Experimental**
  - $\{\text{Pe}_M = 9.6, \text{Pe}_H = 4.4\}$
  - $\{\text{Pe}_M = 9.6, \text{Pe}_H = 3.58\}$
  - $\{\text{Pe}_M = 11.1, \text{Pe}_H = 4.4\}$
  - $\{\text{Pe}_M = 11.1, \text{Pe}_H = 3.58\}$

Reactor bed depth
experimental conversion was for the case of $Pe_H = 4.4$ and $Pe_M = 9.6$ and the largest deviation was for the case of $Pe_H = 3.58$ and $Pe_M = 11.1$. For the lower value of mass transfer Peclet number increasing the heat transfer Peclet number from 3.58 to 4.4 increased the maximum conversion 28.8\% but for the larger mass transfer Peclet number increasing the heat transfer Peclet number in the same manner increased the maximum conversion 14.5\%. A change in the heat transfer Peclet number appears to have a much greater influence on the maximum conversion than a change in the mass transfer Peclet number.

**Effect on temperature distribution**

The effect of using different average Peclet numbers on the radial temperature distributions for a wall temperature of 197°C can be found in Table 5 in Appendix B; the longitudinal temperature distributions are reported in Table 6 in Appendix B. At low bed depths the calculated temperatures at every radial position for all four cases compared within 1 to 6°C; at higher bed depths the calculated values using a heat transfer Peclet number of 3.58 were 5 to 45°C lower than those calculated for a heat transfer Peclet number of 4.4. The best comparison of the experimental temperatures was for the case of $Pe_M = 9.6$ and $Pe_H = 4.4$; the poorest
overall comparison of the experimental values was for the case of $P_e_M = 11.1$ and $P_e_H = 3.58$.

The calculated temperatures were below the experimental values except at the center of the bed at large bed depths where the calculated values were greater than the experimental. The radial temperature distributions for various bed depths are shown in Figures 6 and 7. At low bed depths all the calculated values were about the same for all four cases giving a maximum deviation of $-79^\circ C$ or $-22.8\%$ near the wall and $-14^\circ C$ or $-3.12\%$ at the center. At high bed depths the deviation was $+17^\circ C$ or $+4.2\%$ at the center of the bed and $-2^\circ C$ or $-0.87\%$ next to the wall for the case of $P_e_M = 9.6$ and $P_e_H = 4.4$; the deviation was $-38^\circ C$ or $-9.4\%$ at the center of the bed and $-12^\circ C$ or $-5.2\%$ next to the wall for the case of $P_e_M = 11.1$ and $P_e_H = 3.58$.

The largest deviations occurred at bed depths corresponding to the point where the maximum temperature in the bed was obtained. The longitudinal temperature profiles are given in Figures 8, 9, and 10. It can be seen that the poorest comparison with the experimental values was near the wall at low bed depths.
Average bed properties

\[ z = 0.1467 \text{ ft} \]

\[ t_w = 197 \degree \text{C} \]
Figure 7. Effect of average Peclet numbers on radial temperature distribution at a longitudinal position of 0.4733 foot for a wall temperature of 197°C.
Average bed properties

\[ z = 0.4733 \text{ ft.} \]

\[ t_w = 197^\circ \text{C} \]

Experimental

\[ P_{e_M} = 11.1 \]

\[ P_{e_H} = 4.4 \]

\[ P_{e_M} = 9.6 \]

\[ P_{e_H} = 4.4 \]

\[ P_{e_M} = 11.1 \]

\[ P_{e_H} = 3.58 \]
Figure 8. Effect of average Peclet numbers on longitudinal temperature distribution at the center of the bed for a wall temperature of 197°C
Average bed properties
\( \frac{r}{r_w} = 0 \)
\( t_w = 197 \text{ C} \)

Experimental

\( Pe_M = 9.6 \)
\( Pe_H = 4.4 \)

\( Pe_M = 11.1 \)
\( Pe_H = 3.58 \)
Figure 9. Effect of average Peclet numbers on longitudinal temperature distribution at a dimensionless radial position of 0.5 for a wall temperature of 197°C
Average bed properties

\( \frac{r}{r_w} = 0.5 \)

\( t_w = 197 ^\circ C \)

(a) Experimental
(b) \( Pe_M = 9.6 \)
\( Pe_H = 4.4 \)
(c) \( Pe_M = 11.1 \)
\( Pe_H = 3.58 \)
Figure 10. Effect of average Peclet numbers on longitudinal temperature distribution at a dimensionless radial position of 0.8 for a wall temperature of 197°C
Average bed properties
\( r/r_w = 0.8 \)
\( t_w = 197^\circ C \)

(a) Experimental
(b) \( rPe = 9.6 \)
    \( \beta Pe = 4.4 \)
(c) \( rPe = 11.1 \)
    \( \beta Pe = 3.58 \)
Effect of Variable Bed Properties with No Reaction

The effect of including the variable velocity with average Peclet numbers is to predict a higher radial temperature distribution. Since the non-uniform velocity decreases the value of the velocity at the wall, the temperatures are more uniformly distributed across the radius of the bed and the rate of heat loss through the wall tends to be reduced compared to the case of a uniform velocity distribution.

The calculated temperatures at bed depths of 0.167, 0.333, 0.5, and 0.667 feet for the case of no reaction and a reactor wall temperature of 100°C may be found with the smoothed experimental values in Table 7 in Appendix B. The radial temperature distributions for three solutions of the reactor design equations may be compared with the experimental distributions at various bed depths in Figures 11, 12, and 13. The following three cases were used:

(a) average heat and mass transfer Peclet numbers and a uniform velocity distribution;
(b) point effective thermal conductivity, mass eddy diffusivity, and velocity; and
(c) point heat and mass transfer Peclet numbers and a non-uniform velocity distribution.

The calculated radial distributions for the case of point Peclet numbers at low bed depths appears to match best the
Figure 11. Effect of variable bed properties on radial temperature distribution at a longitudinal position of 0.333 foot with no reaction for a wall temperature of 100°C.
No reaction

$z = 0.333 \text{ ft}$

$t_w = 100^\circ \text{C}$

(a) Experimental
(b) Point $Pe_H, v$
(c) Point $ke, v$
(d) Average prop.
Figure 12. Effect of variable bed properties on radial temperature distribution at a longitudinal position of 0.5 foot with no reaction for a wall temperature of 100°C
No reaction
\( z = 0.5 \text{ ft} \)
\( t_w = 100^\circ C \)

(a) Experimental
(b) Point Pe, v
(c) Point ke, v
(d) Average prop.
Figure 13. Effect of variable bed properties on radial temperature distribution at a longitudinal position of 0.667 foot with no reaction for a wall temperature of 100°C
No reaction

\( z = 0.667 \text{ ft} \)

\( t_w = 100^\circ \text{C} \)

(a) Point Pe, \(v\)
(b) Average prop.
(c) Point ke, \(v\)
(d) Experimental
experimental temperature distribution, with a deviation of $-6^\circ C$ or $-1.55\%$ at the center and $+63^\circ C$ or $+31.5\%$ next to the wall ($r/r_w = 0.9$). The temperature distribution for the case of point $k_e$, $E_-$, and $v$ does not compare with the experimental distribution at the low bed depth giving a deviation of $-20^\circ C$ or $-5.15\%$ at the center and $+21^\circ C$ or $+10.5\%$ next to the wall. However, at higher bed depths the temperature distribution for this same case gives the best approximation to the experimental values. At the largest bed depth, 0.667 feet, the two temperature profiles compare within $-5^\circ C$ or $-2.19\%$ at the center to $+28^\circ C$ or $+22.6\%$ next to the wall. There is a tendency for all the calculated temperatures to deviate from the experimental values near the wall of the reactor except for the case of average properties which predicted values quite close to the experimental.

The effect of using variable Peclet numbers and velocity compared to average values was quite pronounced. The use of the variable properties gives rise to center to wall deviations of +6 to $15^\circ C$ for moderate bed depths and 90 to $92^\circ C$ for the largest bed depth. The use of both point Peclet numbers and a non-uniform velocity distribution tends to over-correct for the inadequacies of using only average properties since the velocity effect is included also in the point Peclet numbers. The larger temperature gradient induced by the lower wall temperature did not seem to lower
the center temperatures for a bed depth of approximately 0.34 feet but only gave a steeper gradient at the wall—at radial positions of $r/r_w = 0.7$ and greater.

The comparison between the radial temperature distribution reported by Schuler et al. (41) and the calculated temperature distribution for a bed depth of 0.354 feet are shown in Figure 14 and the data are presented in Table 8 in Appendix B. The temperature distributions were calculated for three cases:

(a) average heat and mass transfer Peclet numbers and a uniform velocity distribution;
(b) point effective thermal conductivity, eddy mass diffusivity, and velocity; and
(c) average heat and mass transfer Peclet numbers and a non-uniform velocity distribution.

It can be seen from Figure 14 that the case in which average properties were used compared most favorably with the experimental values considering the total radius of the tube and the case in which the point $k_e$, $E_v$ and velocity-values were used compares closely at the center of the bed but tends to deviate from the experimental values away from the center. However, the favorable comparison of the temperature profile using the average bed properties is somewhat distorted in that the experimental temperatures determined for air flowing
Figure 14. Effect of variable bed properties on radial temperature distribution at a longitudinal position of 0.354 foot with no reaction for a wall temperature of 197°C
No reaction
\[ z = 0.354 \text{ ft} \]
\[ t_w = 100^\circ \text{C} \]

(a) Average \( \bar{P}_e \), point \( v \)
(b) Average \( \bar{P}_{eH} \) and \( \bar{v} \)
(c) Point \( ke, v \)
through the bed were used to calculate the point heat transfer Peclet numbers by Schuler et al. (41); these reported point values were averaged radially to find a suitable value for the average heat transfer Peclet number. The point velocities (43), point effective thermal conductivities (29), and point eddy mass diffusivities (16) which were determined from three independent sources combined to give a calculated radial temperature distribution which was quite similar to the experimental distribution. Since the temperatures were given for a single bed depth it is difficult to make a close comparison of the different methods with respect to the experimental values. The results for the higher wall temperature show the consistency of the point effective thermal conductivity data which was taken in a 4-inch tube but for a $D_T/D_p$ ratio of 16. The larger tube size and particle size will give rise to a wall effect which becomes apparent at the larger bed depths.

Effect of Variable Bed Properties on Conversion and Temperature Distribution

The effect of variable bed properties on mean conversion and point temperatures for a reactor of constant wall temperature of $100^\circ C$ and $197^\circ C$ were found for as many as seven different cases. The various combinations of the parameters
considered were:

Case IA: point effective thermal conductivity, eddy mass diffusivity, and point or variable velocity

Case IB: point effective thermal conductivity, eddy mass diffusivity, and average uniform velocity

Case IIA: constant average heat and mass Peclet numbers ($Pe_H = 3.58$, $Pe_M = 11.1$) and variable velocity

Case IIB: constant average heat and mass Peclet numbers and average uniform velocity

Case IIIA: point heat and mass Peclet numbers and point velocity

Case IIIB: point heat and mass Peclet numbers and average uniform velocity

Case IV: effective thermal conductivity and mass diffusivity proportional to velocity which varies with radial position [as recommended by Beek (5) in Advances in Chemical Engineering]
### Table 2. Effect of variable bed properties on the form of the radial term in the partial differential equations for heat and mass transfer

<table>
<thead>
<tr>
<th>Case</th>
<th>Form of radial heat transfer term</th>
<th>Form of radial mass diffusion term</th>
<th>Velocity</th>
</tr>
</thead>
<tbody>
<tr>
<td>IA</td>
<td>$\delta[k_e(r)r \frac{\partial T}{\partial r}]$</td>
<td>$\delta[B(r)r \frac{\partial x}{\partial r}]$</td>
<td>$v(r)$</td>
</tr>
<tr>
<td>IB</td>
<td>$\delta[k_e(r)r \frac{\partial T}{\partial r}]$</td>
<td>$\delta[B(r)r \frac{\partial x}{\partial r}]$</td>
<td>$\overline{v}$</td>
</tr>
<tr>
<td>IIIA</td>
<td>$\frac{\delta(r \frac{\partial T}{\partial r})}{P_e H \frac{\partial r}{\partial r}}$</td>
<td>$\frac{\delta(r \frac{\partial x}{\partial r})}{P_e M \frac{\partial r}{\partial r}}$</td>
<td>$v(r)$</td>
</tr>
<tr>
<td>IIB</td>
<td>$\frac{\delta(r \frac{\partial T}{\partial r})}{P_e H \frac{\partial r}{\partial r}}$</td>
<td>$\frac{\delta(r \frac{\partial x}{\partial r})}{P_e M \frac{\partial r}{\partial r}}$</td>
<td>$\overline{v}$</td>
</tr>
<tr>
<td>IIIIA</td>
<td>$\frac{\delta[\frac{1}{P_e H} r \frac{\partial T}{\partial r}]}{r \frac{\partial r}{\partial r}}$</td>
<td>$\frac{\delta[\frac{1}{P_e M} r \frac{\partial x}{\partial r}]}{r \frac{\partial r}{\partial r}}$</td>
<td>$v(r)$</td>
</tr>
</tbody>
</table>
### Table 2. (Continued)

<table>
<thead>
<tr>
<th>Case</th>
<th>Form of radial heat transfer term</th>
<th>Form of radial mass diffusion term</th>
<th>Velocity</th>
</tr>
</thead>
<tbody>
<tr>
<td>IIIB</td>
<td>$\frac{\partial}{\partial r} \left( \frac{1}{\text{Pe}_H} r \frac{\partial T}{\partial r} \right)$</td>
<td>$\frac{\partial}{\partial r} \left( \frac{1}{\text{Pe}_M} r \frac{\partial x}{\partial r} \right)$</td>
<td>$\bar{v}$</td>
</tr>
<tr>
<td>IV</td>
<td>$\frac{\partial}{\partial r} \left( \gamma v(r) r \frac{\partial T}{\partial r} \right)$</td>
<td>$\frac{\partial}{\partial r} \left( \gamma v(r) r \frac{\partial x}{\partial r} \right)$</td>
<td>$v(r)$</td>
</tr>
</tbody>
</table>

Because there are a number of ways of expressing the Peclet number for heat and mass transfer, the following definitions are used:

$$\text{Pe}_H = \frac{D \cdot C_p \cdot v}{k_e} \quad \text{and} \quad \text{Pe}_M = \frac{D \cdot v}{E}$$

$$\text{Pe}_H = \frac{\int_{0}^{r_w} \frac{D \cdot C_p \cdot v}{k_e} \, rdr}{\int_{0}^{r_w} \, rdr} \quad \text{and} \quad \text{Pe}_M = \frac{\int_{0}^{r_w} \frac{D \cdot v}{E} \, rdr}{\int_{0}^{r_w} \, rdr}$$
The coefficients $a$, $b$, and $g$ for Case IV may be determined by examining the radial term for heat transfer in Equation 9. If the effective thermal conductivity is assumed to be composed of two terms, a turbulent contribution, $k_{TD}$, and a stagnant or solid particle contribution, $k_p$, the radial term may be re-written as

$$\frac{1}{\text{Pe}_H} \frac{1}{G^*} \left[ \frac{1}{\sigma} \frac{\partial}{\partial \sigma} \left[ (k_{TD} + k_p)^* \frac{\partial G}{\partial \sigma} \right] \right] \quad (\text{Eq. 56})$$

where

$$k_e^* = k_{TD}^* + k_p^* \quad (\text{Eq. 57})$$

The thermal conductivities can also be expressed in terms of the corresponding Peclet numbers

$$\frac{1}{\text{Pe}_H^*} = \frac{1}{\text{Pe}_{TD}^*} + \frac{1}{\text{Pe}_p^*} \quad (\text{Eq. 58})$$
If the turbulent contributions for heat and mass transfer are assumed to have the same value, the turbulent heat transfer contribution can be evaluated from the average mass transfer Peclet number which will allow the particle Peclet number to be calculated from Equation 58 in the following manner:

\[
\frac{1}{\text{Pe}_p} = \frac{1}{\text{Pe}_H} - \frac{1}{\text{Pe}_M} \quad \text{(Eq. 59)}
\]

or

\[
\frac{1}{\text{Pe}_p} = \frac{1}{3.58} - \frac{1}{11.1}
\]

If the turbulent contributions for heat, mass, and momentum transfer are assumed to be equal or

\[
\text{k}^{*}_{TD} = \text{E}^{*} = \text{v}^{*} \quad \text{(Eq. 60)}
\]

Equation 57 can be written in terms of \(\text{v}^{*}\) and a constant,

\[
\text{k}^{*}_e = \text{v}^{*} + \frac{\text{k}_{ep}}{\text{k}_{eo}}
\]

\[
\text{k}^{*}_e = \text{v}^{*} + \frac{\text{Pe}_H}{\text{Pe}_p}
\]
From Equation 61 it can be seen that

\[ \alpha = \frac{1}{v_0} \quad \text{and} \quad \beta = 0.678 \]

By a similar analysis, it can be seen from Equation 61 that

\[ \gamma = \frac{1}{v_0} \]

**Effect on mean conversion**

In general all the calculated values were less than the experimental at low bed depths \((z < 0.15\ \text{foot})\). On the other hand at large bed depths the calculated conversions tended to be higher than the experimental values. It must be recognized that the experimental values at low bed depths are subject to great inaccuracies as indicated by Figure 40 in Appendix A. Increasing the wall temperature from 100 to 197°C increased the maximum conversion from 28.1% to 38.9% for essentially the same operating conditions in both cases.
When the reactor was operated at a constant wall temperature of 197°C the largest deviation from the maximum experimental value at the end of the reactor was +49.5% for case IIIA. The best comparison was obtained for case IA or case IB which resulted in a deviation of +6.2% from the experimental. However, the mean conversion versus bed depth for case IA does not predict the same conversion trend as the experimental values which was a rather pronounced damping of conversion after a bed depth of approximately 0.2 foot was reached as shown in Figure 15 and Table 3. The conversion for case IIIA did not predict the correct trend or seem to give a maximum value. The effect of using both a variable velocity and point Peclet number was to give a large over-correction resulting in high conversions with no apparent maximum. The effect of using the average velocity with the point Peclet numbers in place of a variable velocity was to increase the maximum conversion by 3.48% at a bed depth of 0.3 foot. This effect was just reversed when point thermal conductivity and mass diffusivity were used, giving a decrease of 1.69%. The change was fairly uniform after a bed depth of 0.1 foot was reached in both cases. The effect of using a variable velocity with average Peclet numbers was to increase the conversion almost uniformly with bed depth increasing the maximum conversion by 17.7%.

Case IV is recommended in a recent review article by
Figure 15. Effect of variable bed properties on mean conversion for a reactor wall temperature of 197°C
Case IIIA

Case IA

Experimental

Case IIB

Case IV

\[ t_w = 197^\circ C \]

Catalyst bed depth, ft
Table 3. Effect of variable bed properties on mean conversion for the oxidation of sulfur dioxide in a 2-inch packed tubular reactor for a wall temperature of 197°C

<table>
<thead>
<tr>
<th>Reactor length, feet</th>
<th>Case IIB</th>
<th>Case IIIA</th>
<th>Case IIIB</th>
<th>Case IA</th>
<th>Case IB</th>
<th>Case IIA</th>
<th>Case IV</th>
<th>Experimental (41)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>0.05</td>
<td>11.8</td>
<td>13.8</td>
<td>14.0</td>
<td>13.1</td>
<td>13.1</td>
<td>12.4</td>
<td>10.8</td>
<td>16.4</td>
</tr>
<tr>
<td>0.10</td>
<td>18.5</td>
<td>24.6</td>
<td>25.2</td>
<td>22.1</td>
<td>21.8</td>
<td>20.2</td>
<td>15.8</td>
<td>26.6</td>
</tr>
<tr>
<td>0.15</td>
<td>22.5</td>
<td>33.2</td>
<td>34.1</td>
<td>28.5</td>
<td>28.1</td>
<td>25.2</td>
<td>18.0</td>
<td>34.4</td>
</tr>
<tr>
<td>0.20</td>
<td>25.0</td>
<td>40.0</td>
<td>41.3</td>
<td>33.1</td>
<td>33.0</td>
<td>28.5</td>
<td>18.8</td>
<td>37.1</td>
</tr>
<tr>
<td>0.25</td>
<td>26.6</td>
<td>45.6</td>
<td>47.1</td>
<td>36.0</td>
<td>35.6</td>
<td>30.7</td>
<td>18.9</td>
<td>38.4</td>
</tr>
<tr>
<td>0.30</td>
<td>27.5</td>
<td>50.3</td>
<td>52.0</td>
<td>38.7</td>
<td>37.8</td>
<td>32.0</td>
<td>18.9</td>
<td>38.8</td>
</tr>
<tr>
<td>0.35</td>
<td>28.0</td>
<td>54.2</td>
<td>40.2</td>
<td>39.4</td>
<td>32.8</td>
<td>18.8</td>
<td>38.9</td>
<td></td>
</tr>
<tr>
<td>0.40</td>
<td>28.3</td>
<td>58.2</td>
<td>41.2</td>
<td>40.4</td>
<td>33.0</td>
<td>18.8</td>
<td>38.9</td>
<td></td>
</tr>
<tr>
<td>0.45</td>
<td>28.3</td>
<td>41.8</td>
<td>40.9</td>
<td>33.3</td>
<td>18.7</td>
<td>38.9</td>
<td></td>
<td></td>
</tr>
<tr>
<td>0.50</td>
<td>28.3</td>
<td>42.0</td>
<td>41.3</td>
<td>33.3</td>
<td>18.6</td>
<td>38.9</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Beek (5). The assumption was made that the eddy diffusivity and thermal conductivity are proportional to velocity which varies with radial position. This method predicted conversions that were too low and gave the poorest results of any of the cases tested.
For the reactor operated at a wall temperature of 100°C the best comparison of the maximum conversion was for case IA. The deviation for this case was -1.85%. Using case IIIA in the design equations gave a deviation of +35.6% and using average Peclet numbers the deviation was -34.9% when comparing the maximum conversions.

Using a variable velocity with average Peclet numbers caused a fairly uniform increase in the conversion at all radial positions, and increased the maximum conversion by 1.75%. For this lower reactor wall temperature the calculated conversions were larger than the experimental values for bed depths of 0 to 0.25 feet. The experimental and calculated curves also have the same trend for the lower wall temperature as shown in Figure 16 and Table 4.

Prediction of temperature profiles

Although the prediction of point temperatures in the bed is less important and considerably more difficult, the use of the correct values for the bed properties should predict these temperatures fairly accurately. There is a certain amount of difficulty in defining a temperature in the bed since there may be quite a deviation between the catalyst, gas, and the homogeneous effective temperature. This difficulty originated at the entrance where in this
Figure 16. Effect of variable bed properties on mean conversion for a reactor wall temperature of 100°C
Table 4. Effect of variable bed properties on mean conversion for the oxidation of sulfur dioxide in a 2-inch packed tubular reactor for a wall temperature of 100°C

<table>
<thead>
<tr>
<th>Reactor length, feet</th>
<th>Case IIB</th>
<th>Case IIIA</th>
<th>Case IA</th>
<th>Case IIA</th>
<th>Experimental (20, 27)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>0.05</td>
<td>8.81</td>
<td>10.2</td>
<td>9.57</td>
<td>9.48</td>
<td>7.1</td>
</tr>
<tr>
<td>0.10</td>
<td>13.5</td>
<td>17.6</td>
<td>15.4</td>
<td>15.2</td>
<td>13.0</td>
</tr>
<tr>
<td>0.15</td>
<td>16.1</td>
<td>23.3</td>
<td>18.9</td>
<td>18.5</td>
<td>17.8</td>
</tr>
<tr>
<td>0.20</td>
<td>17.4</td>
<td>26.8</td>
<td>20.1</td>
<td>20.2</td>
<td>21.1</td>
</tr>
<tr>
<td>0.25</td>
<td>18.0</td>
<td>31.2</td>
<td>22.0</td>
<td>21.3</td>
<td>23.2</td>
</tr>
<tr>
<td>0.30</td>
<td>18.2</td>
<td>33.8</td>
<td>22.4</td>
<td>21.7</td>
<td>24.8</td>
</tr>
<tr>
<td>0.35</td>
<td>18.2</td>
<td>35.9</td>
<td>22.4</td>
<td>21.8</td>
<td>26.0</td>
</tr>
<tr>
<td>0.40</td>
<td>18.3</td>
<td>37.4</td>
<td>22.3</td>
<td>21.7</td>
<td>27.2</td>
</tr>
<tr>
<td>0.45</td>
<td>18.3</td>
<td>38.4</td>
<td>22.2</td>
<td>21.7</td>
<td>27.8</td>
</tr>
<tr>
<td>0.50</td>
<td>18.3</td>
<td>39.1</td>
<td>22.2</td>
<td>21.5</td>
<td>28.1</td>
</tr>
</tbody>
</table>

work the average of the gas and catalyst temperatures were used. Such an assumption probably gave lower reaction rates at the entrance and this temperature deviation persists throughout the length of the reactor. Also large temperature deviations would have a definite adverse effect on the cal-
culated mean conversion.

In almost all cases the calculated temperature failed to predict the maximum experimental temperature even within 20 to 25°C. It should be noted that for the case of 100°C wall temperature there was some difference in the maximum temperature observed by Hall and Smith (20) and Irvin, Olson, and Smith (27). However, in most cases the calculated values did predict a maximum temperature at approximately the correct or experimentally observed bed depth. The experimental values used for comparison have been determined by taking the best smooth curve that fits all the experimental data from both sources.

The point temperatures were calculated by the computer program for wall temperatures of 100°C and 197°C for cases IA, IIA, IIB, and IIIA and for cases I through IV, respectively. The calculated temperature versus bed depth for constant radial position and temperature versus radial position at constant bed depth for both wall temperatures are presented in Tables 9, 10, 11, and 12 in Appendix B.

An overall comparison for both wall temperatures of the predicted and experimental temperatures may be made by Figures 17, 18, and 19 which show the deviations as a function of bed depth for \( r/r_w = 0, 0.5, \) and 0.8, respectively. Since the selected initial temperature profile was the same for all cases, the deviations were taken as zero at the
Figure 17. Deviations from experimental temperature at the center of the bed for variable bed properties with reaction.
$t_w = 197^\circ C$

$t_w = 100^\circ C$

$r/r_w = 0.5$

Percent deviation from experimental

Catalyst bed depth, ft
Figure 18. Deviations from experimental temperature at a dimensionless radial position of 0.5 for variable bed properties with reaction
Percent deviation from experimental
Figure 19. Deviations from experimental temperature at a dimensionless radial position of 0.8 for variable bed properties with reaction.
reactor entrance. Because this profile is subject to some error any comparison at low bed depths is not too meaningful. Since the calculated center temperatures were below the experimental in almost all cases the deviations were mostly negative up to a bed depth of 0.5 foot. The best comparison was obtained using cases IA and IIA. It can be seen that the average deviation for case IA with respect to bed depth is almost zero for all radial positions except possibly very close to the wall. Also, the deviations for case IIA are consistently negative, hence the average deviation with respect to bed depth is quite large negatively, although the absolute value of the deviations may compare with case IA at a number of longitudinal and radial positions. In addition, it has been shown that the choice of values for average heat and mass transfer Peclet numbers can influence the calculated temperatures and conversions considerably which makes the overall consideration of case IIA less desirable. The deviations observed for the other cases are generally very large at greater bed depths indicating a departure of the calculated temperatures from the experimental after the entrance effect has been overcome.

A more accurate model, such as the Danckwerts boundary condition, was not used since the problem becomes too unwieldy for the reactor design problem considered in this work. However, a number of attempts were made to calculate
the temperatures in the wall region where the reaction rates are generally low and the effective thermal conductivity is the least accurate, compared to other radial positions. This entering effect can become very important because of the influence it exerts on the temperatures calculated at larger bed depths. The use of boundary layer theory without reaction predicted too steep a temperature gradient and an abnormal heat loss at the beginning of the reaction. A Bessel solution which results when the usual simplifying assumptions are made or the assumption that the wall can be approximated as a flat plate will allow for a small reaction term but this solution becomes quite complicated because of the large number of terms necessary to make the series converge for low z-values. Another model which was used in an attempt to take into account the effect of reaction near the wall similar to the flow of a gas past a heated plate (33) was doomed to failure since the velocity effect for flow past a heated plate is just the reverse for the case of flow between a given radial position and the reactor wall.

**Effect of wall temperature** The effect of varying the bed parameters on the radial temperature distribution for a reactor wall temperature of 197°C may be compared for a number of these cases in Figures 20 and 21. The effect of using a variable velocity in place of a uniform average velocity was to cause an increase in the calculated
Figure 20. Effect of variable bed properties on radial temperature distribution with reaction at a longitudinal position 0.1467 foot for a wall temperature of 197°C
$z = 0.147 \text{ ft}$
$\theta_w = 197^\circ \text{C}$

Temperature, $^\circ \text{C}$

Case II B
Case III A
Case IV
Case III B
Case I A

$\frac{r}{r_o}$
Figure 21. Effect of variable bed properties on radial temperature distribution with reaction at a longitudinal position of 0.4733 foot for a wall temperature of 197°C.
temperatures for average Peclet numbers (case IIA) and point effective thermal conductivity and eddy mass diffusivity (case IA); however, for the case of point Peclet numbers (case IIIA) this effect was reversed at a value of $r/r_w = 0.5$.

The effect of a variable velocity used with average Peclet numbers was considerably more significant than for the case of point effective thermal conductivity and mass diffusivity and point Peclet numbers, but this is a result of the experimental method used to obtain the point values. The average heat transfer Peclet number was calculated by taking the space average of the point Peclet numbers as determined by Schuler et al. (41); the use of this weighted value with a velocity which varies radially has the effect of producing a radially varying $Peclet_H$ that does not appear inside the radial term of the partial differential equation. This method has been applied with some success in earlier catalytic reactor design calculations, although the form of the differential equation shows that the Peclet number does not actually enter into the equation in this manner.

At low and moderate bed depths case IV gave a radial temperature distribution quite similar to that for case IIB. At larger bed depths the temperatures for case IV are 97 to 10°C below those predicted by case IIB at $r/r_w = 0$ and $r/r_w = 0.9$, respectively.
The effect of varying the bed properties on the longitudinal temperature distribution for a wall temperature of 197°C may be compared in Figures 22, 23, and 24 for a number of cases. The calculated values were lower than the experimentally observed maximum temperature, which occurs at the center of the bed, for all seven cases. The longitudinal position at which the maximum temperature occurred (approximately 0.16 foot) was predicted quite closely except for cases IIIA, IIIB, and IV, within 0.01 foot. Next to the wall agreement between the experimental and calculated temperatures at all bed depths was poor but case IA appeared to give the best overall comparison.

The effect of including a variable velocity was to cause an increase in the predicted temperatures at all radial positions except at large bed depths where this effect is reduced; for the case of average Peclet numbers the effect of the variable velocity is not diminished at the larger bed depths.

The temperatures calculated for case IV diverge from the experimental to a greater degree as the bed depth increases. The use of point Peclet numbers with an average uniform velocity and a variable velocity had the effect of distorting the predicted temperature profile so that it does not give the correct trend as observed experimentally.

The results for a wall temperature of 100°C were
Figure 22. Effect of variable bed properties with reaction on longitudinal temperature distribution at the center of the bed for a wall temperature of 197°C
Figure 23. Effect of variable bed properties with reaction on longitudinal temperature distribution at a dimensionless radial position of 0.5 for a wall temperature of 197°C
\[ \frac{r}{r_w} = 0.5 \]
\[ t_w = 197^\circ C \]
Figure 24. Effect of variable bed properties with reaction on longitudinal temperature distribution at a radial position of 0.8 for a wall temperature of 197°C
\[ r/r_w = 0.8 \]
\[ t_w = 197^\circ C \]
similar to those observed for a wall temperature of \(197^\circ C\), indicating that a lower wall temperature reduces the maximum temperature obtainable but that the bed properties are essentially independent of temperature as well as conversion over the range investigated. The predicted radial temperatures for several bed depths may be compared in Figures 25, 26, and 27. The best overall comparison with the experimental temperatures was for case IA although the center temperatures tend to be low at all bed depths. This may well result from the fact that the entering temperature distribution was lower for the case of no reaction than for the case of reaction; the predicted temperatures were based on the temperature distribution for the case of no reaction.

The use of variable Peclet numbers and velocity (case IIIA) in place of point effective thermal conductivity, eddy mass diffusivity, and velocity (case IA) results in considerably higher temperatures away from the experimental values. Although case IIA compares favorably with the experimental temperatures consideration must be given to the fact that the value of the average heat transfer Peclet number has a large influence on the calculated temperatures. The effect of using a variable velocity with average Peclet numbers was to predict higher temperatures at all radial positions but the increase is not as great near the wall. This effect follows the trend of the velocity distribution which
Figure 25. Effect of variable bed properties with reaction on radial temperature distribution at a longitudinal position of 0.167 foot for a wall temperature of 100°C
$z = 0.167 \text{ ft.}$
$t = 100 \degree \text{C}$

Graph showing temperature vs. $r/r_w$. The graph includes lines for Case IIIA, Case II B, Case IA, and Experimental data.
Figure 26. Effect of variable bed properties with reaction on radial temperature distribution at a longitudinal position of 0.333 foot for a wall temperature of 100°C.
$z = 0.333 \text{ ft}$
$t_w = 100 \degree \text{C}$
Figure 27. Effect of variable bed properties with reaction on radial temperature distribution at a longitudinal position of 0.5 foot for a wall temperature of 100°C
Experimental

Case II A

Case II B

Case III A

$z = 0.5 \text{ ft}$

$t_w = 100^\circ \text{C}$

Temperature $^\circ \text{C}$

$\frac{r}{r_w}$
indicates reduced, almost uniform, heat transfer at the center of the reactor and a large increase in heat transfer near the wall.

The longitudinal temperature profiles for a number of radial positions may be compared in Figures 28, 29, and 30. The best overall comparison with the experimental temperature profile was for cases IA and IIA although the maximum temperature predicted by case IA fell below the experimentally determined value. The maximum calculated temperature for case IIA was greater than the experimental value for the lower wall temperature where the reverse was true at the higher wall temperature. The maximum predicted temperature for case IA was slightly lower than the experimental value as it was in the case of the higher wall temperature.

The longitudinal position of the maximum center temperature from the experimental data was approximately 0.185 foot compared to 0.1 foot for case IA, 0.125 foot for case IIB, and 0.15 foot for case IIA and IIIA.

The effect of using point Peclet numbers and velocity compared to point effective thermal conductivity, eddy mass diffusivity, and velocity was to cause the temperature to remain too high even at large bed depths by restricting the rate of heat transfer through the bed toward the wall.

The effect of using a variable velocity with average bed properties was to predict considerably higher tempera-
Figure 28. Effect of variable bed properties with reaction on longitudinal temperature distribution at the center of the bed for a wall temperature of 100°C
Figure 29. Effect of variable bed properties with reaction on longitudinal temperature distribution at a dimensionless radial position of 0.5 for a wall temperature of 100°C
\[ \frac{r}{r_w} = 0.5 \]
\[ t_w = 100 \, ^\circ C \]

Temperature, °C

Catalyst bed depth, ft

Case III B
Experimental
Case I A
Case II B
Case II B

136b
Figure 30. Effect of variable bed properties with reaction on longitudinal temperature distribution at a dimensionless radial position of 0.8 for a wall temperature of 100°C.
$r/r_w = 0.8$
$\theta_w = 100 \degree C$
tures except near the wall, which would be expected from the form of the velocity distribution. As the larger bed depths, where the entrance effects do not allow for comparison, case IA compares quite favorably with the experimental temperature distribution at most radial positions.

Effect of initial temperature distribution The effect of using a uniform average temperature distribution at the entrance to the reactor compared to a non-uniform distribution on conversion and temperature was calculated for a reactor wall temperature of $100^\circ$C. The mean conversion versus catalyst bed depth for both of these cases may be compared in Figure 31. Since the temperature near the wall for the case of a uniform initial temperature distribution is high enough to cause considerable reaction there is a greater increase in mean conversion near the wall at low bed depths, which causes a more rapid increase in mean conversion. The conversion at each longitudinal position is increased for a flat entering temperature profile, therefore the maximum mean conversion is increased even though the concentration gradients are of the same order of magnitude at the larger bed depths.

It appears that the use of the entering experimental catalyst or gas temperature profile for the case of $100^\circ$C wall temperature would have the effect of increasing the maximum center temperature with a resulting increase in
Figure 31. Effect of initial temperature distribution on mean conversion for a wall temperature of 100°C
$t_0 = 400 \, ^\circ C$

$t_w = 100 \, ^\circ C$

Mean conversion vs. Reactor bed depth, ft

- Uniform entering temperature
- Experimental
- Non-uniform entering temperature
conversion although the entering temperatures near the wall were also lower than for the case of no reaction. Since there was no initial temperature data available for the case of reaction with a reactor wall temperature of 197°C, the temperature distribution for the case of no reaction was chosen for the calculations involving a reactor wall temperature of 100°C and 197°C in order to have a consistent entering condition.

The case of uniform entering temperature does give higher center temperatures along the length of the reactor as shown by Figure 32. However, the calculated temperature for the case of a uniform entering temperature exceeded the experimental value at most bed depths.

**Effect of Initial Center to Wall Temperature Ratio**

In the formulation of Equation 9, the introduction of the dimensionless temperature, \( \theta \), indicated that the entering center to wall temperature ratio, \( T_o/T_w \), was an important variable. A uniform entering temperature distribution was used so that the effect of the entering temperature distribution would be eliminated from the comparison. Case IA was used in order to compare the results for:

1. an entering temperature of 430°C and \( T_o/T_w = 1.33 \)
2. an entering temperature of 400°C and \( T_o/T_w = 1.33 \)
Figure 32. Effect of initial temperature distribution on the longitudinal temperature distribution with reaction at the center of the bed for a wall temperature of 100°C.
Uniform entering temperature

Experimental

Non-uniform entering temperature

$t_0 = 400^\circ C$
$t_w = 100^\circ C$

Temperature, °C

Reactor bed depth, ft
3. an entering temperature of 350°C and $T_o/T_w = 1.33$
4. an entering temperature of 400°C and $T_o/T_w = 4.0$

The effect of entering temperature and the center to wall temperature ratio may be observed by examining Figure 33. Increasing the entering temperature for a given value of $T_o/T_w$ increased the maximum conversion, but this effect was considerably less at higher entering temperatures. Since the reaction rate passes through a maximum value with increasing temperature, the conversion should pass through a maximum value but the entering temperatures used were not high enough to observe this result.

The effect of entering temperature for $T_o/T_w = 1.33$ is also shown in Figure 34. The tendency for the reacting system to approach an asymptotic conversion and a maximum temperature even though the entering center temperature is steadily increased is quite apparent. Although the same values of maximum center temperature and mean conversion are not obtained, the same trend exists for a non-uniform as for a uniform entering temperature distribution. However, the effect of the center to wall temperature ratio on the maximum temperature is much less pronounced than for the mean conversion as shown in Figure 35.

The effect of $T_o/T_w$ on the resulting temperature profiles is more difficult to interpret. However, as shown in
Figure 33. Effect of entering center and wall temperature on the mean conversion of sulfur dioxide and maximum center temperature for point velocity and heat and mass diffusivity
Mean conversion %
Figure 34. Effect of entering center temperature on mean conversion of sulfur dioxide and maximum center temperature for point velocity and heat and mass diffusivity.
Mean conversion vs entering center temperature for uniform entering temperature distribution

$\frac{t_o}{t_w} = 1.33$

Maximum center temperature

Mean conversion, %

Entering center temperature
Maximum center temperature

Mean conversion

Entering center temperature °C

Maximum center temperature
Figure 35. Effect of entering center to wall temperature ratio on the mean conversion of sulfur dioxide and maximum center temperature for point velocity and heat and mass diffusivity.
Mean conversion vs $t_c/t_w$

Entering center temperature = 400 °

$X =$ non-uniform entering temperature

$\Delta =$ uniform entering temperature

Ratio of center to wall temperature, $t_c/t_w$
Mean conversion vs $t_c/t_w$
Entering center temperature = 400 °C
$X$ = non-uniform entering temperature
$\Delta$ = uniform entering temperature

Maximum temperature
Mean conversion

Ratio of center to wall temperature, $t_0/t_w$
Figure 36, the longitudinal position of the maximum temperature is shifted toward the reactor entrance by an increase in entering temperature. The effect of using a uniform entering temperature distribution compared with a non-uniform distribution with the same $T_0/T_w$ was to shift the position of the maximum temperature away from the reactor entrance by as much as 50%. It is also apparent that a greater maximum temperature was obtained for the same increase in center temperature at constant $T_0/T_w$ as for the wall temperature at constant entering center temperature. However, the effect of the lower wall temperature has a stronger influence on the center temperature at large bed depths.
Figure 36. Effect of entering temperature distribution, entering center temperature, and wall temperature on the longitudinal temperature distribution.
Non-uniform entering temperature

Uniform entering temperature

Point $k_e$, $E$, and velocity

$T_{\text{MAX}} = 548.8 \, ^{\circ}\text{C}$

$T_{\text{MAX}} = 507.2 \, ^{\circ}\text{C}$

$T_{\text{MAX}} = 486.9 \, ^{\circ}\text{C}$

$T_{\text{MAX}} = 477.6 \, ^{\circ}\text{C}$

$T_{\text{MAX}} = 458.6 \, ^{\circ}\text{C}$

$T_{\text{MAX}} = 388 \, ^{\circ}\text{C}$

$T_{\text{MAX}} = 322.5 \, ^{\circ}\text{C}$, $t_0/t_w = 1.33$

$T_{\text{MAX}} = 300 \, ^{\circ}\text{C}$, $t_0/t_w = 1.33$

$t_w = 197 \, ^{\circ}\text{C}$, $t_0/t_w = 2$

$t_w = 197 \, ^{\circ}\text{C}$, $t_0/t_w = 2$

$t_w = 100 \, ^{\circ}\text{C}$, $t_0/t_w = 4$

$t_w = 263 \, ^{\circ}\text{C}$, $t_0/t_w = 1.33$

Reactor bed depth, ft
CONCLUSIONS

1. The mean conversion for a reactor wall temperature of 197°C was predicted with the greatest accuracy when point values of the eddy mass diffusivity, effective thermal conductivity, and velocity were used as a function of radial position. The next best agreement was obtained by the use of average values for the Peclet numbers combined with the use of a non-uniform velocity distribution.

2. The assumption of an average velocity and average diffusivities for heat and mass transfer gave better results than the assumption that the diffusivities were linear functions of velocity which varied with radial position. The latter case, in fact, gave the poorest comparison with the experimental data.

3. The effect of a variable velocity on the predicted conversion was not great when point diffusivities were used.

4. At a lower reactor wall temperature of 100°C, the use of point values of diffusivity and velocity also predicted the most accurate results.

5. The prediction of temperature profiles in the radial and longitudinal direction was carried out with reasonable accuracy (3 to 20%) by each method. This comparison was made difficult by the ambiguity of the experimental data consisting of catalyst and gas temperatures that differed by
as much as 15 to 20°C, and the difficulty of resolving the correct inlet temperature profile. In this work it was necessary to use average values of gas and catalyst temperature and this procedure did not predict sufficiently high temperatures in the entrance region. Also there was no attempt to choose either gas or catalyst pellet temperature for an absolute comparison. However, the use of variable thermal conductivity and mass diffusivity with a variable velocity as well as the use of constant average diffusivities with variable velocity gave better results than the case of variable Peclet numbers in predicting temperature profiles. The use of variable properties did give slightly better results at the higher bed depths. These conclusions are somewhat influenced by the fact that the experimental data were not available for the point diffusivities for the same tube size used in this work, although \( D_p/D_T \) and the mass velocity were the same. More extensive data would be expected to give better agreement. This speculation is borne out by the inability of the point effective thermal conductivity to predict temperature profiles in the wall region for this system with no reaction even though the general trend of the curves is correctly predicted.

6. The predicted mean conversion was found to increase with a decreased mass transfer Peclet number and to a greater degree with an increased heat transfer Peclet number.
7. At a constant inlet temperature, $T_0$, the mean conversion decreases with an increase in the ratio of inlet temperature to wall temperature.

The maximum temperature obtainable is almost entirely dependent on the entering center temperature, although a higher reactor wall temperature will tend to increase the maximum temperature.

The longitudinal position of the maximum temperature will be greater for an increase in entering center temperature at a constant entering to wall temperature ratio. The longitudinal position will be only slightly decreased for an increase in the entering wall temperature at a constant entering to wall temperature ratio.

8. At a constant ratio of entering to wall temperature, the mean conversion was found to increase with an increase in the entering temperature. The maximum temperature was also increased and its position moved further down the tube.
LITERATURE CITED


ACKNOWLEDGMENTS

The author wishes to express his sincere appreciation to Dr. R. W. Fahien for his guidance and constant encouragement in carrying out this project.
Figure 37. Experimental radial temperature distribution with no reaction for 100°C wall temperature.
Figure 38. Experimental radial temperature distribution with no reaction for 100°C wall temperature
No reaction

$\Delta z = 6" \ (20)$

$x z = 8" \ (27)$

$t_w = 100 \ ^\circ C$
No reaction

\[ \Delta \quad z = 6'' \quad (20) \]

\[ \times \quad z = 8'' \quad (27) \]

\[ t_w = 100 \, ^\circ \text{C} \]
Figure 39. Comparison of experimental entering radial temperature distribution with no reaction and with reaction for a reactor wall temperature of 100°C.
Figure 40. Experimental mean conversion of \( \text{SO}_2 \) for reactor wall temperature of 100°C
Figure 41. Experimental mean conversion of $SO_2$ for reactor wall temperature of 197°C
Figure 42. Experimental radial temperature distribution with reaction for reactor wall temperature of 100°C
Figure 43. Experimental radial temperature distribution with reaction for reactor wall temperature of 100°C
Figure 44. Experimental radial temperature distribution with reaction for reactor wall temperature of 100°C
Figure 45. Experimental radial temperature distribution with reaction for reactor wall temperature of 100°C
Figure 46. Experimental radial catalyst temperature distribution for reactor wall temperature of 197°C
Figure 47. Experimental radial catalyst temperature distribution for reactor wall temperature of 197°C
Figure 48. Experimental radial velocity distribution
Figure 49. Experimental radial effective thermal diffusivity distribution
\[ \bar{G} = 300 \text{ lb/hr ft}^2 \quad (29) \]

\[ \frac{D_p}{D_T} = 16 \]

Figure 50. Experimental radial effective thermal conductivity distribution
Figure 51. Experimental radial eddy mass diffusivity distribution

\[ G = 368 \text{ lb/hr ft}^2 \]  
\[ D_p/D_t = 16 \]
Figure 52. Graph of variation of reaction rate for zero conversion with temperature for the oxidation of sulfur dioxide
\[
\begin{align*}
\ln r_0 &= -\frac{8584.18}{T} + 9.813905 \quad T > 733 \\
\ln r_0 &= -\frac{7671.75}{T} + 8.569 \quad 663 < T < 733 \\
\ln r_0 &= -\frac{15019.73}{T} + 19.64871 \quad T < 663
\end{align*}
\]
Figure 53. Graph of variation of rate equation constant \( a \) with temperature for the oxidation of sulfur dioxide
\[
\ln \alpha = \frac{12.6165}{T} + 16.1505 \quad T < 640 \, ^\circ K
\]
\[
\alpha = 0.00117 T + 617 \quad 640 < T < 733
\]
\[
\ln \frac{1}{\alpha} = -0.013265 T + 11.6993 \quad T > 733
\]
Figure 54. Graph of variation of rate equation constant b with temperature for oxidation of sulfur dioxide
\[ b = 0 \quad T < 617 \]
\[ b = (-0.00079435 T + 0.601)(T-623) + 0.757 \quad 617 < T < 723 \]
\[ \ln b = -0.003144 T + 3.331 \quad T > 733 \]
Figure 55. Graph of variation of rate equation constant c with temperature for oxidation of sulfur dioxide
\[
\ln \left[ (b + c)^{1/2} r_0 \right] = 0.21599T - 16.744
\]
177

APPENDIX B
Table 5. Effect of average Peclet numbers on the radial temperature distribution for the oxidation of sulfur dioxide in a 2-inch packed tubular reactor for a wall temperature of 197°C

<table>
<thead>
<tr>
<th>$r/r_w$</th>
<th>Temperature, °C</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Pe_M=11.1</td>
</tr>
<tr>
<td>0</td>
<td>435</td>
</tr>
<tr>
<td>0.1</td>
<td>435</td>
</tr>
<tr>
<td>0.2</td>
<td>435</td>
</tr>
<tr>
<td>0.3</td>
<td>434</td>
</tr>
<tr>
<td>0.4</td>
<td>431</td>
</tr>
<tr>
<td>0.5</td>
<td>424</td>
</tr>
<tr>
<td>0.6</td>
<td>409</td>
</tr>
<tr>
<td>0.7</td>
<td>380</td>
</tr>
<tr>
<td>0.8</td>
<td>332</td>
</tr>
<tr>
<td>0.9</td>
<td>268</td>
</tr>
<tr>
<td>1.0</td>
<td>197</td>
</tr>
<tr>
<td>0</td>
<td>454</td>
</tr>
<tr>
<td>0.1</td>
<td>454</td>
</tr>
<tr>
<td>0.2</td>
<td>452</td>
</tr>
<tr>
<td>0.3</td>
<td>448</td>
</tr>
<tr>
<td>0.4</td>
<td>440</td>
</tr>
<tr>
<td>0.5</td>
<td>426</td>
</tr>
<tr>
<td>0.6</td>
<td>401</td>
</tr>
<tr>
<td>0.7</td>
<td>363</td>
</tr>
<tr>
<td>0.8</td>
<td>313</td>
</tr>
<tr>
<td>0.9</td>
<td>255</td>
</tr>
<tr>
<td>1.0</td>
<td>197</td>
</tr>
<tr>
<td>0</td>
<td>478</td>
</tr>
<tr>
<td>0.1</td>
<td>476</td>
</tr>
<tr>
<td>0.2</td>
<td>469</td>
</tr>
</tbody>
</table>

$z = 0.0442$ ft

$z = 0.0733$ ft

$z = 0.1467$ ft
Table 5. (Continued)

<table>
<thead>
<tr>
<th>r/r_w</th>
<th>Pe_M=11.1</th>
<th>Pe_M=9.6</th>
<th>Pe_M=9.6</th>
<th>Pe_M=11.1</th>
<th>Experimental</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Fe_H=3.58</td>
<td>Fe_H=3.58</td>
<td>Fe_H=4.4</td>
<td>Fe_H=4.4</td>
<td></td>
</tr>
<tr>
<td>0.3</td>
<td>457</td>
<td>457</td>
<td>473</td>
<td>468</td>
<td>503</td>
</tr>
<tr>
<td>0.4</td>
<td>437</td>
<td>437</td>
<td>456</td>
<td>450</td>
<td>490</td>
</tr>
<tr>
<td>0.5</td>
<td>409</td>
<td>410</td>
<td>429</td>
<td>423</td>
<td>471</td>
</tr>
<tr>
<td>0.6</td>
<td>374</td>
<td>374</td>
<td>394</td>
<td>387</td>
<td>443</td>
</tr>
<tr>
<td>0.7</td>
<td>332</td>
<td>332</td>
<td>349</td>
<td>343</td>
<td>413</td>
</tr>
<tr>
<td>0.8</td>
<td>287</td>
<td>287</td>
<td>300</td>
<td>295</td>
<td>375</td>
</tr>
<tr>
<td>0.9</td>
<td>241</td>
<td>241</td>
<td>248</td>
<td>245</td>
<td>319</td>
</tr>
<tr>
<td>1.0</td>
<td>197</td>
<td>197</td>
<td>197</td>
<td>197</td>
<td>197</td>
</tr>
</tbody>
</table>

\[ z = 0.3525 \text{ ft} \]

<table>
<thead>
<tr>
<th>r/r_w</th>
<th>Pe_M=11.1</th>
<th>Pe_M=9.6</th>
<th>Pe_M=9.6</th>
<th>Pe_M=11.1</th>
<th>Experimental</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Fe_H=3.58</td>
<td>Fe_H=3.58</td>
<td>Fe_H=4.4</td>
<td>Fe_H=4.4</td>
<td></td>
</tr>
<tr>
<td>0</td>
<td>418</td>
<td>419</td>
<td>463</td>
<td>450</td>
<td>452</td>
</tr>
<tr>
<td>0.1</td>
<td>414</td>
<td>416</td>
<td>460</td>
<td>446</td>
<td>446</td>
</tr>
<tr>
<td>0.2</td>
<td>405</td>
<td>406</td>
<td>449</td>
<td>436</td>
<td>435</td>
</tr>
<tr>
<td>0.3</td>
<td>389</td>
<td>390</td>
<td>431</td>
<td>418</td>
<td>419</td>
</tr>
<tr>
<td>0.4</td>
<td>367</td>
<td>368</td>
<td>407</td>
<td>394</td>
<td>398</td>
</tr>
<tr>
<td>0.5</td>
<td>341</td>
<td>342</td>
<td>377</td>
<td>364</td>
<td>374</td>
</tr>
<tr>
<td>0.6</td>
<td>313</td>
<td>314</td>
<td>343</td>
<td>331</td>
<td>347</td>
</tr>
<tr>
<td>0.7</td>
<td>284</td>
<td>284</td>
<td>308</td>
<td>297</td>
<td>315</td>
</tr>
<tr>
<td>0.8</td>
<td>254</td>
<td>254</td>
<td>271</td>
<td>262</td>
<td>278</td>
</tr>
<tr>
<td>0.9</td>
<td>224</td>
<td>225</td>
<td>234</td>
<td>229</td>
<td>239</td>
</tr>
<tr>
<td>1.0</td>
<td>197</td>
<td>197</td>
<td>197</td>
<td>197</td>
<td>197</td>
</tr>
</tbody>
</table>

\[ z = 0.4733 \text{ ft} \]

<table>
<thead>
<tr>
<th>r/r_w</th>
<th>Pe_M=11.1</th>
<th>Pe_M=9.6</th>
<th>Pe_M=9.6</th>
<th>Pe_M=11.1</th>
<th>Experimental</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Fe_H=3.58</td>
<td>Fe_H=3.58</td>
<td>Fe_H=4.4</td>
<td>Fe_H=4.4</td>
<td></td>
</tr>
<tr>
<td>0</td>
<td>366</td>
<td>368</td>
<td>421</td>
<td>403</td>
<td>404</td>
</tr>
<tr>
<td>0.1</td>
<td>363</td>
<td>365</td>
<td>418</td>
<td>400</td>
<td>402</td>
</tr>
<tr>
<td>0.2</td>
<td>355</td>
<td>357</td>
<td>408</td>
<td>391</td>
<td>395</td>
</tr>
<tr>
<td>0.3</td>
<td>343</td>
<td>345</td>
<td>393</td>
<td>375</td>
<td>385</td>
</tr>
<tr>
<td>0.4</td>
<td>328</td>
<td>328</td>
<td>371</td>
<td>355</td>
<td>369</td>
</tr>
<tr>
<td>0.5</td>
<td>308</td>
<td>309</td>
<td>346</td>
<td>331</td>
<td>346</td>
</tr>
<tr>
<td>0.6</td>
<td>287</td>
<td>287</td>
<td>319</td>
<td>305</td>
<td>318</td>
</tr>
<tr>
<td>0.7</td>
<td>264</td>
<td>264</td>
<td>289</td>
<td>278</td>
<td>289</td>
</tr>
<tr>
<td>0.8</td>
<td>241</td>
<td>241</td>
<td>258</td>
<td>250</td>
<td>259</td>
</tr>
<tr>
<td>0.9</td>
<td>218</td>
<td>218</td>
<td>228</td>
<td>223</td>
<td>230</td>
</tr>
<tr>
<td>1.0</td>
<td>197</td>
<td>197</td>
<td>197</td>
<td>197</td>
<td>197</td>
</tr>
</tbody>
</table>
Table 6. Effect of average Peclet numbers on the longitudinal temperature distribution for the oxidation of sulfur dioxide in a 2-inch packed tubular reactor for a wall temperature of 197°C

<table>
<thead>
<tr>
<th>$r/r_w$</th>
<th>$T_{E_{M}=11.1}$</th>
<th>$T_{E_{M}=9.6}$</th>
<th>$T_{E_{M}=9.6}$</th>
<th>$T_{E_{M}=11.1}$</th>
<th>$T_{E_{H}=3.58}$</th>
<th>$T_{E_{H}=3.58}$</th>
<th>$T_{E_{H}=4.4}$</th>
<th>$T_{E_{H}=4.4}$</th>
<th>Experimental (41)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$r/r_w = 0$</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>0</td>
<td>400</td>
<td>400</td>
<td>400</td>
<td>400</td>
<td>400</td>
<td>400</td>
<td>400</td>
<td>400</td>
<td>400</td>
</tr>
<tr>
<td>0.05</td>
<td>439</td>
<td>439</td>
<td>440</td>
<td>440</td>
<td>440</td>
<td>440</td>
<td>440</td>
<td>440</td>
<td>455</td>
</tr>
<tr>
<td>0.10</td>
<td>468</td>
<td>469</td>
<td>473</td>
<td>471</td>
<td>471</td>
<td>471</td>
<td>471</td>
<td>471</td>
<td>497</td>
</tr>
<tr>
<td>0.15</td>
<td>478</td>
<td>481</td>
<td>491</td>
<td>489</td>
<td>489</td>
<td>489</td>
<td>489</td>
<td>489</td>
<td>515</td>
</tr>
<tr>
<td>0.20</td>
<td>474</td>
<td>476</td>
<td>495</td>
<td>492</td>
<td>492</td>
<td>492</td>
<td>492</td>
<td>492</td>
<td>509</td>
</tr>
<tr>
<td>0.25</td>
<td>459</td>
<td>462</td>
<td>490</td>
<td>484</td>
<td>484</td>
<td>484</td>
<td>484</td>
<td>484</td>
<td>494</td>
</tr>
<tr>
<td>0.30</td>
<td>440</td>
<td>442</td>
<td>479</td>
<td>469</td>
<td>469</td>
<td>469</td>
<td>469</td>
<td>469</td>
<td>475</td>
</tr>
<tr>
<td>0.35</td>
<td>419</td>
<td>420</td>
<td>464</td>
<td>451</td>
<td>451</td>
<td>451</td>
<td>451</td>
<td>451</td>
<td>454</td>
</tr>
<tr>
<td>0.40</td>
<td>397</td>
<td>399</td>
<td>447</td>
<td>431</td>
<td>431</td>
<td>431</td>
<td>431</td>
<td>431</td>
<td>432</td>
</tr>
<tr>
<td>0.45</td>
<td>376</td>
<td>378</td>
<td>429</td>
<td>420</td>
<td>420</td>
<td>420</td>
<td>420</td>
<td>420</td>
<td>411</td>
</tr>
<tr>
<td>0.50</td>
<td>355</td>
<td>357</td>
<td>412</td>
<td>391</td>
<td>391</td>
<td>391</td>
<td>391</td>
<td>391</td>
<td></td>
</tr>
<tr>
<td>0.55</td>
<td>338</td>
<td>339</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>$r/r_w = 0.3$</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>0</td>
<td>400</td>
<td>400</td>
<td>400</td>
<td>400</td>
<td>400</td>
<td>400</td>
<td>400</td>
<td>400</td>
<td>400</td>
</tr>
<tr>
<td>0.05</td>
<td>437</td>
<td>437</td>
<td>439</td>
<td>438</td>
<td>438</td>
<td>438</td>
<td>438</td>
<td>438</td>
<td>452</td>
</tr>
<tr>
<td>0.10</td>
<td>456</td>
<td>456</td>
<td>465</td>
<td>461</td>
<td>461</td>
<td>461</td>
<td>461</td>
<td>461</td>
<td>488</td>
</tr>
<tr>
<td>0.15</td>
<td>456</td>
<td>457</td>
<td>473</td>
<td>468</td>
<td>468</td>
<td>468</td>
<td>468</td>
<td>468</td>
<td>503</td>
</tr>
<tr>
<td>0.20</td>
<td>445</td>
<td>446</td>
<td>470</td>
<td>463</td>
<td>463</td>
<td>463</td>
<td>463</td>
<td>463</td>
<td>494</td>
</tr>
<tr>
<td>0.25</td>
<td>428</td>
<td>430</td>
<td>460</td>
<td>451</td>
<td>451</td>
<td>451</td>
<td>451</td>
<td>451</td>
<td>475</td>
</tr>
<tr>
<td>0.30</td>
<td>409</td>
<td>411</td>
<td>447</td>
<td>436</td>
<td>436</td>
<td>436</td>
<td>436</td>
<td>436</td>
<td>454</td>
</tr>
<tr>
<td>0.35</td>
<td>390</td>
<td>391</td>
<td>432</td>
<td>419</td>
<td>419</td>
<td>419</td>
<td>419</td>
<td>419</td>
<td>434</td>
</tr>
<tr>
<td>0.40</td>
<td>370</td>
<td>372</td>
<td>416</td>
<td>401</td>
<td>401</td>
<td>401</td>
<td>401</td>
<td>401</td>
<td>413</td>
</tr>
<tr>
<td>0.45</td>
<td>351</td>
<td>353</td>
<td>400</td>
<td>390</td>
<td>390</td>
<td>390</td>
<td>390</td>
<td>390</td>
<td>393</td>
</tr>
<tr>
<td>0.50</td>
<td>335</td>
<td>336</td>
<td>384</td>
<td>373</td>
<td>373</td>
<td>373</td>
<td>373</td>
<td>373</td>
<td></td>
</tr>
<tr>
<td>0.55</td>
<td>320</td>
<td>321</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>$r/r_w = 0.5$</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>0</td>
<td>400</td>
<td>400</td>
<td>400</td>
<td>400</td>
<td>400</td>
<td>400</td>
<td>400</td>
<td>400</td>
<td>400</td>
</tr>
<tr>
<td>0.05</td>
<td>425</td>
<td>425</td>
<td>431</td>
<td>428</td>
<td>428</td>
<td>428</td>
<td>428</td>
<td>428</td>
<td>424</td>
</tr>
<tr>
<td>0.10</td>
<td>422</td>
<td>422</td>
<td>436</td>
<td>431</td>
<td>431</td>
<td>431</td>
<td>431</td>
<td>431</td>
<td>472</td>
</tr>
<tr>
<td>0.15</td>
<td>408</td>
<td>409</td>
<td>429</td>
<td>422</td>
<td>422</td>
<td>422</td>
<td>422</td>
<td>422</td>
<td>470</td>
</tr>
</tbody>
</table>
Table 6. (Continued)

<table>
<thead>
<tr>
<th>( \frac{r}{r_w} )</th>
<th>( r/\tau_w = 0.8 )</th>
<th>( r/\tau_w = 0.9 )</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \frac{r}{r_w} )</td>
<td>Temperature, °C</td>
<td></td>
</tr>
<tr>
<td>0.20</td>
<td>392</td>
<td>342</td>
</tr>
<tr>
<td>0.25</td>
<td>375</td>
<td>342</td>
</tr>
<tr>
<td>0.30</td>
<td>358</td>
<td>342</td>
</tr>
<tr>
<td>0.35</td>
<td>342</td>
<td>342</td>
</tr>
<tr>
<td>0.40</td>
<td>327</td>
<td>342</td>
</tr>
<tr>
<td>0.45</td>
<td>314</td>
<td>342</td>
</tr>
<tr>
<td>0.50</td>
<td>302</td>
<td>342</td>
</tr>
<tr>
<td>0.55</td>
<td>291</td>
<td>342</td>
</tr>
<tr>
<td>0</td>
<td>379</td>
<td>342</td>
</tr>
<tr>
<td>0.05</td>
<td>328</td>
<td>342</td>
</tr>
<tr>
<td>0.10</td>
<td>301</td>
<td>342</td>
</tr>
<tr>
<td>0.15</td>
<td>286</td>
<td>342</td>
</tr>
<tr>
<td>0.20</td>
<td>276</td>
<td>342</td>
</tr>
<tr>
<td>0.25</td>
<td>267</td>
<td>342</td>
</tr>
<tr>
<td>0.30</td>
<td>260</td>
<td>342</td>
</tr>
<tr>
<td>0.35</td>
<td>254</td>
<td>342</td>
</tr>
<tr>
<td>0.40</td>
<td>248</td>
<td>342</td>
</tr>
<tr>
<td>0.45</td>
<td>243</td>
<td>342</td>
</tr>
<tr>
<td>0.50</td>
<td>239</td>
<td>342</td>
</tr>
<tr>
<td>0.55</td>
<td>234</td>
<td>342</td>
</tr>
</tbody>
</table>

- \( \tau_w = 0.8 \)
- \( \tau_w = 0.9 \)
Table 7. Effect of variable bed properties on the radial temperature distribution for no reaction in a 2-inch packed tube for a wall temperature of 100°C

<table>
<thead>
<tr>
<th>$r/r_w$</th>
<th>Case IIB</th>
<th>Case IIIB</th>
<th>Case IIIA</th>
<th>Case IIIIA</th>
<th>Case IA</th>
<th>Experimental (20, 27)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>376</td>
<td>382</td>
<td>368</td>
<td>388</td>
<td>388</td>
<td></td>
</tr>
<tr>
<td>0.1</td>
<td>374</td>
<td>381</td>
<td>367</td>
<td>385</td>
<td>380</td>
<td></td>
</tr>
<tr>
<td>0.2</td>
<td>367</td>
<td>378</td>
<td>363</td>
<td>385</td>
<td>381</td>
<td></td>
</tr>
<tr>
<td>0.3</td>
<td>355</td>
<td>372</td>
<td>356</td>
<td>380</td>
<td>371</td>
<td></td>
</tr>
<tr>
<td>0.4</td>
<td>337</td>
<td>365</td>
<td>346</td>
<td>380</td>
<td>356</td>
<td></td>
</tr>
<tr>
<td>0.5</td>
<td>312</td>
<td>356</td>
<td>334</td>
<td>366</td>
<td>356</td>
<td></td>
</tr>
<tr>
<td>0.6</td>
<td>279</td>
<td>345</td>
<td>319</td>
<td>333</td>
<td>333</td>
<td></td>
</tr>
<tr>
<td>0.7</td>
<td>240</td>
<td>332</td>
<td>299</td>
<td>300</td>
<td>300</td>
<td></td>
</tr>
<tr>
<td>0.8</td>
<td>194</td>
<td>310</td>
<td>270</td>
<td>258</td>
<td>258</td>
<td></td>
</tr>
<tr>
<td>0.9</td>
<td>147</td>
<td>263</td>
<td>221</td>
<td>200</td>
<td>200</td>
<td></td>
</tr>
<tr>
<td>1.0</td>
<td>100</td>
<td>100</td>
<td>100</td>
<td>100</td>
<td>100</td>
<td></td>
</tr>
</tbody>
</table>

$z = 0.167$ ft

$z = 0.333$ ft

<table>
<thead>
<tr>
<th>$r/r_w$</th>
<th>Case IIB</th>
<th>Case IIIB</th>
<th>Case IIIA</th>
<th>Case IIIIA</th>
<th>Case IA</th>
<th>Experimental (20, 27)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>307</td>
<td>349</td>
<td>311</td>
<td>346</td>
<td>346</td>
<td></td>
</tr>
<tr>
<td>0.1</td>
<td>305</td>
<td>348</td>
<td>310</td>
<td>344</td>
<td>344</td>
<td></td>
</tr>
<tr>
<td>0.2</td>
<td>296</td>
<td>345</td>
<td>306</td>
<td>338</td>
<td>338</td>
<td></td>
</tr>
<tr>
<td>0.3</td>
<td>283</td>
<td>339</td>
<td>299</td>
<td>327</td>
<td>327</td>
<td></td>
</tr>
<tr>
<td>0.4</td>
<td>265</td>
<td>332</td>
<td>290</td>
<td>310</td>
<td>310</td>
<td></td>
</tr>
<tr>
<td>0.5</td>
<td>243</td>
<td>323</td>
<td>279</td>
<td>286</td>
<td>286</td>
<td></td>
</tr>
<tr>
<td>0.6</td>
<td>217</td>
<td>313</td>
<td>266</td>
<td>253</td>
<td>253</td>
<td></td>
</tr>
<tr>
<td>0.7</td>
<td>188</td>
<td>301</td>
<td>250</td>
<td>215</td>
<td>215</td>
<td></td>
</tr>
<tr>
<td>0.8</td>
<td>158</td>
<td>281</td>
<td>227</td>
<td>176</td>
<td>176</td>
<td></td>
</tr>
<tr>
<td>0.9</td>
<td>128</td>
<td>240</td>
<td>190</td>
<td>138</td>
<td>138</td>
<td></td>
</tr>
<tr>
<td>1.0</td>
<td>100</td>
<td>100</td>
<td>100</td>
<td>100</td>
<td>100</td>
<td></td>
</tr>
</tbody>
</table>
Table 7. (Continued)

<table>
<thead>
<tr>
<th>r/(r_{w})</th>
<th>Case IIB</th>
<th>Case IIIA</th>
<th>Case IA</th>
<th>Experimental (20, 27)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>244</td>
<td>318</td>
<td>261</td>
<td>286</td>
</tr>
<tr>
<td>0.1</td>
<td>242</td>
<td>317</td>
<td>260</td>
<td>282</td>
</tr>
<tr>
<td>0.2</td>
<td>236</td>
<td>314</td>
<td>257</td>
<td>273</td>
</tr>
<tr>
<td>0.3</td>
<td>227</td>
<td>309</td>
<td>252</td>
<td>259</td>
</tr>
<tr>
<td>0.4</td>
<td>213</td>
<td>302</td>
<td>244</td>
<td>239</td>
</tr>
<tr>
<td>0.5</td>
<td>197</td>
<td>294</td>
<td>236</td>
<td>215</td>
</tr>
<tr>
<td>0.6</td>
<td>179</td>
<td>285</td>
<td>226</td>
<td>192</td>
</tr>
<tr>
<td>0.7</td>
<td>159</td>
<td>274</td>
<td>214</td>
<td>170</td>
</tr>
<tr>
<td>0.8</td>
<td>139</td>
<td>257</td>
<td>196</td>
<td>145</td>
</tr>
<tr>
<td>0.9</td>
<td>119</td>
<td>221</td>
<td>168</td>
<td>123</td>
</tr>
<tr>
<td>1.0</td>
<td>100</td>
<td>100</td>
<td>100</td>
<td>100</td>
</tr>
</tbody>
</table>

\(z = 0.50 \text{ ft}\)

<table>
<thead>
<tr>
<th>r/(r_{w})</th>
<th>Case IIB</th>
<th>Case IIIA</th>
<th>Case IA</th>
<th>Experimental (20, 27)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>199</td>
<td>289</td>
<td>223</td>
<td>228</td>
</tr>
<tr>
<td>0.1</td>
<td>198</td>
<td>288</td>
<td>222</td>
<td>228</td>
</tr>
<tr>
<td>0.2</td>
<td>194</td>
<td>286</td>
<td>219</td>
<td>227</td>
</tr>
<tr>
<td>0.3</td>
<td>187</td>
<td>281</td>
<td>215</td>
<td>223</td>
</tr>
<tr>
<td>0.4</td>
<td>178</td>
<td>275</td>
<td>210</td>
<td>214</td>
</tr>
<tr>
<td>0.5</td>
<td>167</td>
<td>268</td>
<td>203</td>
<td>202</td>
</tr>
<tr>
<td>0.6</td>
<td>154</td>
<td>260</td>
<td>196</td>
<td>186</td>
</tr>
<tr>
<td>0.7</td>
<td>141</td>
<td>251</td>
<td>186</td>
<td>167</td>
</tr>
<tr>
<td>0.8</td>
<td>127</td>
<td>236</td>
<td>173</td>
<td>147</td>
</tr>
<tr>
<td>0.9</td>
<td>113</td>
<td>205</td>
<td>152</td>
<td>124</td>
</tr>
<tr>
<td>1.0</td>
<td>100</td>
<td>100</td>
<td>100</td>
<td>100</td>
</tr>
</tbody>
</table>

\(z = 0.667 \text{ ft}\)
Table 8. Effect of variable bed properties on the radial temperature distribution for no reaction in a 2-inch packed tube for a wall temperature of 197°C

<table>
<thead>
<tr>
<th>r/r_w</th>
<th>Case IIB</th>
<th>Case IA</th>
<th>Case IIA</th>
<th>Experimental (41)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>334</td>
<td>341</td>
<td>367</td>
<td>348</td>
</tr>
<tr>
<td>0.1</td>
<td>333</td>
<td>340</td>
<td>367</td>
<td>344</td>
</tr>
<tr>
<td>0.2</td>
<td>327</td>
<td>337</td>
<td>365</td>
<td>337</td>
</tr>
<tr>
<td>0.3</td>
<td>318</td>
<td>332</td>
<td>361</td>
<td>327</td>
</tr>
<tr>
<td>0.4</td>
<td>306</td>
<td>326</td>
<td>356</td>
<td>314</td>
</tr>
<tr>
<td>0.5</td>
<td>291</td>
<td>319</td>
<td>350</td>
<td>300</td>
</tr>
<tr>
<td>0.6</td>
<td>274</td>
<td>310</td>
<td>343</td>
<td>285</td>
</tr>
<tr>
<td>0.7</td>
<td>255</td>
<td>299</td>
<td>334</td>
<td>269</td>
</tr>
<tr>
<td>0.8</td>
<td>235</td>
<td>283</td>
<td>321</td>
<td>251</td>
</tr>
<tr>
<td>0.9</td>
<td>216</td>
<td>258</td>
<td>293</td>
<td>230</td>
</tr>
<tr>
<td>1.0</td>
<td>197</td>
<td>197</td>
<td>197</td>
<td>197</td>
</tr>
</tbody>
</table>

z = 0.354 ft
Table 9. Effect of variable bed properties on the radial temperature distribution for the oxidation of sulfur dioxide in a 2-inch packed tubular reactor for a wall temperature of 100°C

<table>
<thead>
<tr>
<th>( r/r_w )</th>
<th>Case IIB</th>
<th>Case IIIA</th>
<th>Case IIA</th>
<th>Case IA</th>
<th>Experimental (20, 27)</th>
</tr>
</thead>
<tbody>
<tr>
<td>z = 0.1675 ft</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>0</td>
<td>456</td>
<td>473</td>
<td>475</td>
<td>443</td>
<td>481</td>
</tr>
<tr>
<td>0.1</td>
<td>452</td>
<td>470</td>
<td>471</td>
<td>440</td>
<td>474</td>
</tr>
<tr>
<td>0.2</td>
<td>440</td>
<td>463</td>
<td>461</td>
<td>432</td>
<td>461</td>
</tr>
<tr>
<td>0.3</td>
<td>420</td>
<td>452</td>
<td>442</td>
<td>419</td>
<td>443</td>
</tr>
<tr>
<td>0.4</td>
<td>391</td>
<td>437</td>
<td>416</td>
<td>402</td>
<td>422</td>
</tr>
<tr>
<td>0.5</td>
<td>353</td>
<td>419</td>
<td>381</td>
<td>382</td>
<td>395</td>
</tr>
<tr>
<td>0.6</td>
<td>309</td>
<td>399</td>
<td>336</td>
<td>358</td>
<td>365</td>
</tr>
<tr>
<td>0.7</td>
<td>258</td>
<td>377</td>
<td>284</td>
<td>330</td>
<td>328</td>
</tr>
<tr>
<td>0.8</td>
<td>207</td>
<td>344</td>
<td>225</td>
<td>293</td>
<td>278</td>
</tr>
<tr>
<td>0.9</td>
<td>152</td>
<td>283</td>
<td>162</td>
<td>235</td>
<td>200</td>
</tr>
<tr>
<td>1.0</td>
<td>100</td>
<td>100</td>
<td>100</td>
<td>100</td>
<td>100</td>
</tr>
<tr>
<td>z = 0.333 ft</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>0</td>
<td>367</td>
<td>443</td>
<td>401</td>
<td>369</td>
<td>391</td>
</tr>
<tr>
<td>0.1</td>
<td>363</td>
<td>441</td>
<td>397</td>
<td>367</td>
<td>379</td>
</tr>
<tr>
<td>0.2</td>
<td>351</td>
<td>434</td>
<td>385</td>
<td>350</td>
<td>367</td>
</tr>
<tr>
<td>0.3</td>
<td>332</td>
<td>425</td>
<td>365</td>
<td>349</td>
<td>352</td>
</tr>
<tr>
<td>0.4</td>
<td>307</td>
<td>412</td>
<td>339</td>
<td>336</td>
<td>335</td>
</tr>
<tr>
<td>0.5</td>
<td>277</td>
<td>396</td>
<td>308</td>
<td>320</td>
<td>315</td>
</tr>
<tr>
<td>0.6</td>
<td>243</td>
<td>379</td>
<td>272</td>
<td>302</td>
<td>288</td>
</tr>
<tr>
<td>0.7</td>
<td>208</td>
<td>359</td>
<td>231</td>
<td>281</td>
<td>253</td>
</tr>
<tr>
<td>0.8</td>
<td>171</td>
<td>330</td>
<td>187</td>
<td>252</td>
<td>213</td>
</tr>
<tr>
<td>0.9</td>
<td>134</td>
<td>274</td>
<td>142</td>
<td>207</td>
<td>163</td>
</tr>
<tr>
<td>1.0</td>
<td>100</td>
<td>100</td>
<td>100</td>
<td>100</td>
<td>100</td>
</tr>
<tr>
<td>z = 0.50 ft</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>0</td>
<td>282</td>
<td>409</td>
<td>314</td>
<td>300</td>
<td>332</td>
</tr>
<tr>
<td>0.1</td>
<td>279</td>
<td>407</td>
<td>313</td>
<td>298</td>
<td>321</td>
</tr>
<tr>
<td>0.2</td>
<td>271</td>
<td>402</td>
<td>303</td>
<td>294</td>
<td>309</td>
</tr>
<tr>
<td>0.3</td>
<td>259</td>
<td>394</td>
<td>290</td>
<td>287</td>
<td>295</td>
</tr>
<tr>
<td>0.4</td>
<td>242</td>
<td>382</td>
<td>272</td>
<td>278</td>
<td>279</td>
</tr>
<tr>
<td>0.5</td>
<td>222</td>
<td>368</td>
<td>250</td>
<td>257</td>
<td>261</td>
</tr>
<tr>
<td>0.6</td>
<td>199</td>
<td>353</td>
<td>224</td>
<td>254</td>
<td>239</td>
</tr>
<tr>
<td>0.7</td>
<td>174</td>
<td>336</td>
<td>194</td>
<td>239</td>
<td>212</td>
</tr>
<tr>
<td>0.8</td>
<td>149</td>
<td>312</td>
<td>163</td>
<td>217</td>
<td>181</td>
</tr>
<tr>
<td>0.9</td>
<td>124</td>
<td>261</td>
<td>130</td>
<td>183</td>
<td>144</td>
</tr>
<tr>
<td>1.0</td>
<td>100</td>
<td>100</td>
<td>100</td>
<td>100</td>
<td>100</td>
</tr>
</tbody>
</table>
Table 10. Effect of variable bed properties on the longitudinal temperature distribution for the oxidation of sulfur dioxide in a 2-inch packed tubular reactor for a wall temperature of 100°C

<table>
<thead>
<tr>
<th>$r/r_w$</th>
<th>Case IIB</th>
<th>Case IIIA</th>
<th>Case IIA</th>
<th>Case IA</th>
<th>Experimental (20, 27)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>400</td>
<td>400</td>
<td>400</td>
<td>400</td>
<td>400</td>
</tr>
<tr>
<td>0.05</td>
<td>437</td>
<td>442</td>
<td>443</td>
<td>440</td>
<td>435</td>
</tr>
<tr>
<td>0.10</td>
<td>459</td>
<td>465</td>
<td>470</td>
<td>454</td>
<td>455</td>
</tr>
<tr>
<td>0.15</td>
<td>460</td>
<td>472</td>
<td>476</td>
<td>447</td>
<td>468</td>
</tr>
<tr>
<td>0.20</td>
<td>444</td>
<td>470</td>
<td>466</td>
<td>431</td>
<td>468</td>
</tr>
<tr>
<td>0.25</td>
<td>418</td>
<td>464</td>
<td>445</td>
<td>410</td>
<td>459</td>
</tr>
<tr>
<td>0.30</td>
<td>388</td>
<td>454</td>
<td>419</td>
<td>386</td>
<td>443</td>
</tr>
<tr>
<td>0.35</td>
<td>356</td>
<td>444</td>
<td>392</td>
<td>360</td>
<td>419</td>
</tr>
<tr>
<td>0.40</td>
<td>328</td>
<td>432</td>
<td>363</td>
<td>337</td>
<td>386</td>
</tr>
<tr>
<td>0.45</td>
<td>304</td>
<td>421</td>
<td>336</td>
<td>318</td>
<td>352</td>
</tr>
<tr>
<td>0.50</td>
<td>282</td>
<td>409</td>
<td>314</td>
<td>300</td>
<td>324</td>
</tr>
<tr>
<td>0.55</td>
<td>262</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
</tbody>
</table>

$r/r_w = 0.3$

<table>
<thead>
<tr>
<th>$r/r_w$</th>
<th>Case IIB</th>
<th>Case IIIA</th>
<th>Case IIA</th>
<th>Case IA</th>
<th>Experimental (20, 27)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>399</td>
<td>399</td>
<td>399</td>
<td>399</td>
<td>399</td>
</tr>
<tr>
<td>0.05</td>
<td>430</td>
<td>433</td>
<td>436</td>
<td>429</td>
<td>409</td>
</tr>
<tr>
<td>0.10</td>
<td>438</td>
<td>448</td>
<td>451</td>
<td>434</td>
<td>434</td>
</tr>
<tr>
<td>0.15</td>
<td>427</td>
<td>452</td>
<td>447</td>
<td>424</td>
<td>443</td>
</tr>
<tr>
<td>0.20</td>
<td>405</td>
<td>450</td>
<td>431</td>
<td>407</td>
<td>436</td>
</tr>
<tr>
<td>0.25</td>
<td>377</td>
<td>444</td>
<td>408</td>
<td>387</td>
<td>418</td>
</tr>
<tr>
<td>0.30</td>
<td>349</td>
<td>436</td>
<td>383</td>
<td>364</td>
<td>394</td>
</tr>
<tr>
<td>0.35</td>
<td>323</td>
<td>426</td>
<td>356</td>
<td>342</td>
<td>372</td>
</tr>
<tr>
<td>0.40</td>
<td>299</td>
<td>416</td>
<td>332</td>
<td>322</td>
<td>352</td>
</tr>
<tr>
<td>0.45</td>
<td>279</td>
<td>405</td>
<td>310</td>
<td>303</td>
<td>333</td>
</tr>
<tr>
<td>0.50</td>
<td>259</td>
<td>394</td>
<td>289</td>
<td>287</td>
<td>315</td>
</tr>
<tr>
<td>0.55</td>
<td>242</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>293</td>
</tr>
</tbody>
</table>

$r/r_w = 0.5$

<table>
<thead>
<tr>
<th>$r/r_w$</th>
<th>Case IIB</th>
<th>Case IIIA</th>
<th>Case IIA</th>
<th>Case IA</th>
<th>Experimental (20, 27)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>394</td>
<td>394</td>
<td>394</td>
<td>394</td>
<td>394</td>
</tr>
<tr>
<td>0.05</td>
<td>407</td>
<td>411</td>
<td>413</td>
<td>406</td>
<td>404</td>
</tr>
<tr>
<td>0.10</td>
<td>389</td>
<td>418</td>
<td>407</td>
<td>401</td>
<td>409</td>
</tr>
<tr>
<td>0.15</td>
<td>362</td>
<td>419</td>
<td>389</td>
<td>387</td>
<td>409</td>
</tr>
<tr>
<td>0.20</td>
<td>336</td>
<td>416</td>
<td>364</td>
<td>370</td>
<td>397</td>
</tr>
<tr>
<td>0.25</td>
<td>312</td>
<td>412</td>
<td>343</td>
<td>351</td>
<td>371</td>
</tr>
<tr>
<td>$r/r_w$</td>
<td>Case IIB</td>
<td>Case IIIA</td>
<td>Case IIA</td>
<td>Case IA</td>
<td>Experimental (20, 27)</td>
</tr>
<tr>
<td>--------</td>
<td>----------</td>
<td>-----------</td>
<td>----------</td>
<td>---------</td>
<td>----------------------</td>
</tr>
<tr>
<td>0.30</td>
<td>290</td>
<td>405</td>
<td>322</td>
<td>332</td>
<td>341</td>
</tr>
<tr>
<td>0.35</td>
<td>270</td>
<td>397</td>
<td>301</td>
<td>314</td>
<td>314</td>
</tr>
<tr>
<td>0.40</td>
<td>252</td>
<td>388</td>
<td>282</td>
<td>297</td>
<td>291</td>
</tr>
<tr>
<td>0.45</td>
<td>236</td>
<td>378</td>
<td>265</td>
<td>281</td>
<td>271</td>
</tr>
<tr>
<td>0.50</td>
<td>222</td>
<td>368</td>
<td>250</td>
<td>267</td>
<td>255</td>
</tr>
<tr>
<td>0.55</td>
<td>209</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>243</td>
</tr>
</tbody>
</table>

$r/r_w = 0.8$

<table>
<thead>
<tr>
<th>$r/r_w$</th>
<th>Case IIB</th>
<th>Case IIIA</th>
<th>Case IIA</th>
<th>Case IA</th>
<th>Experimental (20, 27)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.05</td>
<td>271</td>
<td>346</td>
<td>346</td>
<td>326</td>
<td>346</td>
</tr>
<tr>
<td>0.10</td>
<td>234</td>
<td>345</td>
<td>252</td>
<td>296</td>
<td>300</td>
</tr>
<tr>
<td>0.15</td>
<td>212</td>
<td>340</td>
<td>215</td>
<td>283</td>
<td>254</td>
</tr>
<tr>
<td>0.20</td>
<td>197</td>
<td>336</td>
<td>203</td>
<td>271</td>
<td>232</td>
</tr>
<tr>
<td>0.25</td>
<td>186</td>
<td>318</td>
<td>176</td>
<td>237</td>
<td>191</td>
</tr>
<tr>
<td>0.30</td>
<td>176</td>
<td>312</td>
<td>163</td>
<td>217</td>
<td>178</td>
</tr>
<tr>
<td>0.35</td>
<td>168</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>171</td>
</tr>
</tbody>
</table>

$r/r_w = 0.9$

<table>
<thead>
<tr>
<th>$r/r_w$</th>
<th>Case IIB</th>
<th>Case IIIA</th>
<th>Case IIA</th>
<th>Case IA</th>
<th>Experimental (20, 27)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.05</td>
<td>190</td>
<td>298</td>
<td>298</td>
<td>298</td>
<td>298</td>
</tr>
<tr>
<td>0.10</td>
<td>167</td>
<td>288</td>
<td>200</td>
<td>263</td>
<td>260</td>
</tr>
<tr>
<td>0.15</td>
<td>155</td>
<td>283</td>
<td>176</td>
<td>248</td>
<td>229</td>
</tr>
<tr>
<td>0.20</td>
<td>148</td>
<td>282</td>
<td>156</td>
<td>238</td>
<td>215</td>
</tr>
<tr>
<td>0.25</td>
<td>142</td>
<td>280</td>
<td>150</td>
<td>221</td>
<td>166</td>
</tr>
<tr>
<td>0.30</td>
<td>137</td>
<td>278</td>
<td>145</td>
<td>213</td>
<td>157</td>
</tr>
<tr>
<td>0.35</td>
<td>133</td>
<td>274</td>
<td>141</td>
<td>205</td>
<td>151</td>
</tr>
<tr>
<td>0.40</td>
<td>130</td>
<td>271</td>
<td>137</td>
<td>197</td>
<td>147</td>
</tr>
<tr>
<td>0.45</td>
<td>126</td>
<td>266</td>
<td>134</td>
<td>190</td>
<td>143</td>
</tr>
<tr>
<td>0.50</td>
<td>124</td>
<td>261</td>
<td>130</td>
<td>183</td>
<td>141</td>
</tr>
<tr>
<td>0.55</td>
<td>121</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>137</td>
</tr>
</tbody>
</table>
Table 11. Effect of variable bed properties on the radial temperature distribution for the oxidation of sulfur dioxide in a 2-inch packed tubular reactor for a wall temperature of 197°C

<table>
<thead>
<tr>
<th>$r/r_w$</th>
<th>Case IIB</th>
<th>Case IIIB</th>
<th>Case IIIA</th>
<th>Case IIA</th>
<th>Case IB</th>
<th>Case IA</th>
<th>Case IV (41)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>435</td>
<td>435</td>
<td>441</td>
<td>441</td>
<td>435</td>
<td>440</td>
<td>439</td>
</tr>
<tr>
<td>0.1</td>
<td>435</td>
<td>435</td>
<td>440</td>
<td>441</td>
<td>435</td>
<td>439</td>
<td>438</td>
</tr>
<tr>
<td>0.2</td>
<td>435</td>
<td>435</td>
<td>439</td>
<td>440</td>
<td>434</td>
<td>438</td>
<td>437</td>
</tr>
<tr>
<td>0.3</td>
<td>434</td>
<td>434</td>
<td>436</td>
<td>438</td>
<td>433</td>
<td>436</td>
<td>433</td>
</tr>
<tr>
<td>0.4</td>
<td>431</td>
<td>431</td>
<td>433</td>
<td>435</td>
<td>429</td>
<td>431</td>
<td>425</td>
</tr>
<tr>
<td>0.5</td>
<td>424</td>
<td>427</td>
<td>427</td>
<td>428</td>
<td>423</td>
<td>424</td>
<td>415</td>
</tr>
<tr>
<td>0.6</td>
<td>409</td>
<td>420</td>
<td>418</td>
<td>415</td>
<td>413</td>
<td>414</td>
<td>397</td>
</tr>
<tr>
<td>0.7</td>
<td>380</td>
<td>410</td>
<td>407</td>
<td>390</td>
<td>398</td>
<td>399</td>
<td>360</td>
</tr>
<tr>
<td>0.8</td>
<td>332</td>
<td>390</td>
<td>387</td>
<td>344</td>
<td>372</td>
<td>372</td>
<td>317</td>
</tr>
<tr>
<td>0.9</td>
<td>268</td>
<td>342</td>
<td>341</td>
<td>276</td>
<td>322</td>
<td>323</td>
<td>263</td>
</tr>
<tr>
<td>1.0</td>
<td>197</td>
<td>197</td>
<td>197</td>
<td>197</td>
<td>197</td>
<td>197</td>
<td>197</td>
</tr>
</tbody>
</table>

$z = 0.0442$ ft

<table>
<thead>
<tr>
<th>$r/r_w$</th>
<th>Case IIB</th>
<th>Case IIIB</th>
<th>Case IIIA</th>
<th>Case IIA</th>
<th>Case IB</th>
<th>Case IA</th>
<th>Case IV (41)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>454</td>
<td>454</td>
<td>461</td>
<td>463</td>
<td>453</td>
<td>459</td>
<td>453</td>
</tr>
<tr>
<td>0.1</td>
<td>454</td>
<td>453</td>
<td>460</td>
<td>462</td>
<td>452</td>
<td>458</td>
<td>452</td>
</tr>
<tr>
<td>0.2</td>
<td>452</td>
<td>452</td>
<td>458</td>
<td>460</td>
<td>450</td>
<td>455</td>
<td>447</td>
</tr>
<tr>
<td>0.3</td>
<td>448</td>
<td>449</td>
<td>454</td>
<td>456</td>
<td>446</td>
<td>450</td>
<td>437</td>
</tr>
<tr>
<td>0.4</td>
<td>440</td>
<td>445</td>
<td>448</td>
<td>449</td>
<td>439</td>
<td>442</td>
<td>423</td>
</tr>
<tr>
<td>0.5</td>
<td>426</td>
<td>438</td>
<td>438</td>
<td>435</td>
<td>429</td>
<td>431</td>
<td>405</td>
</tr>
<tr>
<td>0.6</td>
<td>401</td>
<td>429</td>
<td>427</td>
<td>414</td>
<td>415</td>
<td>417</td>
<td>373</td>
</tr>
<tr>
<td>0.7</td>
<td>363</td>
<td>417</td>
<td>414</td>
<td>379</td>
<td>396</td>
<td>399</td>
<td>338</td>
</tr>
<tr>
<td>0.8</td>
<td>313</td>
<td>395</td>
<td>392</td>
<td>327</td>
<td>365</td>
<td>367</td>
<td>297</td>
</tr>
<tr>
<td>0.9</td>
<td>255</td>
<td>345</td>
<td>343</td>
<td>263</td>
<td>316</td>
<td>318</td>
<td>250</td>
</tr>
<tr>
<td>1.0</td>
<td>197</td>
<td>197</td>
<td>197</td>
<td>197</td>
<td>197</td>
<td>197</td>
<td>197</td>
</tr>
</tbody>
</table>

$z = 0.0733$ ft

<table>
<thead>
<tr>
<th>$r/r_w$</th>
<th>Case IIB</th>
<th>Case IIIB</th>
<th>Case IIIA</th>
<th>Case IIA</th>
<th>Case IB</th>
<th>Case IA</th>
<th>Case IV (41)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>478</td>
<td>484</td>
<td>489</td>
<td>494</td>
<td>473</td>
<td>477</td>
<td>426</td>
</tr>
<tr>
<td>0.1</td>
<td>476</td>
<td>483</td>
<td>487</td>
<td>492</td>
<td>472</td>
<td>476</td>
<td>422</td>
</tr>
<tr>
<td>0.2</td>
<td>469</td>
<td>480</td>
<td>484</td>
<td>486</td>
<td>466</td>
<td>470</td>
<td>413</td>
</tr>
<tr>
<td>0.3</td>
<td>457</td>
<td>474</td>
<td>477</td>
<td>474</td>
<td>457</td>
<td>460</td>
<td>397</td>
</tr>
<tr>
<td>0.4</td>
<td>437</td>
<td>467</td>
<td>468</td>
<td>456</td>
<td>445</td>
<td>448</td>
<td>376</td>
</tr>
<tr>
<td>0.5</td>
<td>409</td>
<td>457</td>
<td>456</td>
<td>431</td>
<td>429</td>
<td>433</td>
<td>351</td>
</tr>
</tbody>
</table>

$z = 0.1467$ ft
Table 11. (Continued)

<table>
<thead>
<tr>
<th>$r/r_w$</th>
<th>Case IIB</th>
<th>Case IIIB</th>
<th>Case IIIA</th>
<th>Case IIA</th>
<th>Case IB</th>
<th>Case IA</th>
<th>Case IV (41)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.6</td>
<td>374</td>
<td>445</td>
<td>442</td>
<td>396</td>
<td>410</td>
<td>414</td>
<td>321</td>
</tr>
<tr>
<td>0.7</td>
<td>332</td>
<td>430</td>
<td>426</td>
<td>352</td>
<td>388</td>
<td>391</td>
<td>293</td>
</tr>
<tr>
<td>0.8</td>
<td>287</td>
<td>406</td>
<td>401</td>
<td>302</td>
<td>355</td>
<td>358</td>
<td>263</td>
</tr>
<tr>
<td>0.9</td>
<td>241</td>
<td>352</td>
<td>349</td>
<td>248</td>
<td>307</td>
<td>309</td>
<td>232</td>
</tr>
<tr>
<td>1.0</td>
<td>197</td>
<td>197</td>
<td>197</td>
<td>197</td>
<td>197</td>
<td>197</td>
<td>197</td>
</tr>
</tbody>
</table>

$z = 0.3525 \text{ ft}$

<table>
<thead>
<tr>
<th>$r/r_w$</th>
<th>Case IIB</th>
<th>Case IIIB</th>
<th>Case IIIA</th>
<th>Case IIA</th>
<th>Case IB</th>
<th>Case IA</th>
<th>Case IV (41)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>418</td>
<td>494</td>
<td>448</td>
<td>435</td>
<td>433</td>
<td>312</td>
<td>452</td>
</tr>
<tr>
<td>0.1</td>
<td>414</td>
<td>492</td>
<td>444</td>
<td>433</td>
<td>431</td>
<td>310</td>
<td>446</td>
</tr>
<tr>
<td>0.2</td>
<td>405</td>
<td>488</td>
<td>425</td>
<td>427</td>
<td>426</td>
<td>305</td>
<td>435</td>
</tr>
<tr>
<td>0.3</td>
<td>389</td>
<td>481</td>
<td>419</td>
<td>417</td>
<td>417</td>
<td>296</td>
<td>419</td>
</tr>
<tr>
<td>0.4</td>
<td>367</td>
<td>472</td>
<td>398</td>
<td>403</td>
<td>406</td>
<td>286</td>
<td>398</td>
</tr>
<tr>
<td>0.5</td>
<td>341</td>
<td>460</td>
<td>371</td>
<td>388</td>
<td>392</td>
<td>273</td>
<td>374</td>
</tr>
<tr>
<td>0.6</td>
<td>313</td>
<td>447</td>
<td>339</td>
<td>370</td>
<td>376</td>
<td>259</td>
<td>347</td>
</tr>
<tr>
<td>0.7</td>
<td>284</td>
<td>431</td>
<td>304</td>
<td>350</td>
<td>356</td>
<td>245</td>
<td>315</td>
</tr>
<tr>
<td>0.8</td>
<td>254</td>
<td>407</td>
<td>268</td>
<td>324</td>
<td>329</td>
<td>230</td>
<td>278</td>
</tr>
<tr>
<td>0.9</td>
<td>224</td>
<td>355</td>
<td>231</td>
<td>286</td>
<td>290</td>
<td>214</td>
<td>239</td>
</tr>
<tr>
<td>1.0</td>
<td>197</td>
<td>197</td>
<td>197</td>
<td>197</td>
<td>197</td>
<td>197</td>
<td>197</td>
</tr>
</tbody>
</table>

$z = 0.4733 \text{ ft}$

<table>
<thead>
<tr>
<th>$r/r_w$</th>
<th>Case IIB</th>
<th>Case IIIB</th>
<th>Case IIIA</th>
<th>Case IIA</th>
<th>Case IB</th>
<th>Case IA</th>
<th>Case IV (41)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>366</td>
<td>480</td>
<td>399</td>
<td>398</td>
<td>400</td>
<td>269</td>
<td>404</td>
</tr>
<tr>
<td>0.1</td>
<td>363</td>
<td>396</td>
<td>396</td>
<td>393</td>
<td>393</td>
<td>265</td>
<td>395</td>
</tr>
<tr>
<td>0.2</td>
<td>355</td>
<td>388</td>
<td>391</td>
<td>385</td>
<td>385</td>
<td>260</td>
<td>385</td>
</tr>
<tr>
<td>0.3</td>
<td>343</td>
<td>375</td>
<td>382</td>
<td>375</td>
<td>375</td>
<td>253</td>
<td>369</td>
</tr>
<tr>
<td>0.4</td>
<td>328</td>
<td>357</td>
<td>370</td>
<td>363</td>
<td>363</td>
<td>245</td>
<td>346</td>
</tr>
<tr>
<td>0.5</td>
<td>308</td>
<td>452</td>
<td>335</td>
<td>357</td>
<td>357</td>
<td>236</td>
<td>318</td>
</tr>
<tr>
<td>0.6</td>
<td>287</td>
<td>311</td>
<td>342</td>
<td>342</td>
<td>342</td>
<td>227</td>
<td>289</td>
</tr>
<tr>
<td>0.7</td>
<td>264</td>
<td>283</td>
<td>326</td>
<td>333</td>
<td>333</td>
<td>217</td>
<td>259</td>
</tr>
<tr>
<td>0.8</td>
<td>241</td>
<td>398</td>
<td>254</td>
<td>304</td>
<td>311</td>
<td>208</td>
<td>230</td>
</tr>
<tr>
<td>0.9</td>
<td>218</td>
<td>224</td>
<td>273</td>
<td>277</td>
<td>277</td>
<td>197</td>
<td>197</td>
</tr>
<tr>
<td>1.0</td>
<td>197</td>
<td>197</td>
<td>197</td>
<td>197</td>
<td>197</td>
<td>197</td>
<td>197</td>
</tr>
</tbody>
</table>
Table 12. Effect of variable bed properties on the longitudinal temperature distribution for the oxidation of sulfur dioxide in a 2-inch packed tubular reactor for a wall temperature of 197°C

<table>
<thead>
<tr>
<th>r/r_w</th>
<th>Case</th>
<th>Case</th>
<th>Case</th>
<th>Case</th>
<th>Case</th>
<th>Case</th>
<th>Case</th>
<th>Case</th>
<th>Experimental</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>IIB</td>
<td>IIIB</td>
<td>IIIA</td>
<td>IIA</td>
<td>IB</td>
<td>IA</td>
<td>IA</td>
<td>IV</td>
<td>(41)</td>
</tr>
<tr>
<td>0</td>
<td>400</td>
<td>400</td>
<td>400</td>
<td>400</td>
<td>400</td>
<td>400</td>
<td>400</td>
<td>400</td>
<td></td>
</tr>
<tr>
<td>0.05</td>
<td>439</td>
<td>445</td>
<td>439</td>
<td>445</td>
<td>439</td>
<td>444</td>
<td>443</td>
<td>455</td>
<td></td>
</tr>
<tr>
<td>0.10</td>
<td>468</td>
<td>475</td>
<td>468</td>
<td>479</td>
<td>465</td>
<td>470</td>
<td>455</td>
<td>497</td>
<td></td>
</tr>
<tr>
<td>0.15</td>
<td>478</td>
<td>490</td>
<td>485</td>
<td>495</td>
<td>474</td>
<td>478</td>
<td>435</td>
<td>515</td>
<td></td>
</tr>
<tr>
<td>0.20</td>
<td>474</td>
<td>497</td>
<td>493</td>
<td>495</td>
<td>471</td>
<td>473</td>
<td>404</td>
<td>509</td>
<td></td>
</tr>
<tr>
<td>0.25</td>
<td>459</td>
<td>498</td>
<td>496</td>
<td>486</td>
<td>462</td>
<td>463</td>
<td>369</td>
<td>494</td>
<td></td>
</tr>
<tr>
<td>0.30</td>
<td>440</td>
<td>497</td>
<td>496</td>
<td>469</td>
<td>450</td>
<td>449</td>
<td>338</td>
<td>475</td>
<td></td>
</tr>
<tr>
<td>0.35</td>
<td>419</td>
<td>494</td>
<td></td>
<td>449</td>
<td>436</td>
<td>434</td>
<td>313</td>
<td>454</td>
<td></td>
</tr>
<tr>
<td>0.40</td>
<td>397</td>
<td>490</td>
<td></td>
<td>428</td>
<td>421</td>
<td>419</td>
<td>292</td>
<td>432</td>
<td></td>
</tr>
<tr>
<td>0.45</td>
<td>376</td>
<td></td>
<td></td>
<td>408</td>
<td>405</td>
<td>404</td>
<td>275</td>
<td>411</td>
<td></td>
</tr>
<tr>
<td>0.50</td>
<td>355</td>
<td></td>
<td></td>
<td>388</td>
<td>390</td>
<td>389</td>
<td>262</td>
<td>391</td>
<td></td>
</tr>
<tr>
<td>0.55</td>
<td>338</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>253</td>
</tr>
<tr>
<td>0</td>
<td>400</td>
<td>400</td>
<td>400</td>
<td>400</td>
<td>400</td>
<td>400</td>
<td>400</td>
<td>400</td>
<td></td>
</tr>
<tr>
<td>0.05</td>
<td>437</td>
<td>441</td>
<td>437</td>
<td>442</td>
<td>436</td>
<td>439</td>
<td>435</td>
<td>452</td>
<td></td>
</tr>
<tr>
<td>0.10</td>
<td>456</td>
<td>471</td>
<td>462</td>
<td>467</td>
<td>454</td>
<td>457</td>
<td>432</td>
<td>488</td>
<td></td>
</tr>
<tr>
<td>0.15</td>
<td>456</td>
<td>479</td>
<td>476</td>
<td>474</td>
<td>457</td>
<td>461</td>
<td>407</td>
<td>503</td>
<td></td>
</tr>
<tr>
<td>0.20</td>
<td>445</td>
<td>484</td>
<td>482</td>
<td>468</td>
<td>452</td>
<td>455</td>
<td>376</td>
<td>494</td>
<td></td>
</tr>
<tr>
<td>0.25</td>
<td>428</td>
<td>485</td>
<td>484</td>
<td>456</td>
<td>443</td>
<td>445</td>
<td>346</td>
<td>475</td>
<td></td>
</tr>
<tr>
<td>0.30</td>
<td>409</td>
<td>484</td>
<td>484</td>
<td>439</td>
<td>431</td>
<td>432</td>
<td>319</td>
<td>454</td>
<td></td>
</tr>
<tr>
<td>0.35</td>
<td>390</td>
<td>482</td>
<td></td>
<td>420</td>
<td>417</td>
<td>418</td>
<td>298</td>
<td>434</td>
<td></td>
</tr>
<tr>
<td>0.40</td>
<td>370</td>
<td>480</td>
<td></td>
<td>402</td>
<td>403</td>
<td>404</td>
<td>280</td>
<td>413</td>
<td></td>
</tr>
<tr>
<td>0.45</td>
<td>351</td>
<td></td>
<td></td>
<td>383</td>
<td>389</td>
<td>389</td>
<td>265</td>
<td>393</td>
<td></td>
</tr>
<tr>
<td>0.50</td>
<td>335</td>
<td></td>
<td></td>
<td>365</td>
<td>374</td>
<td>374</td>
<td>253</td>
<td>373</td>
<td></td>
</tr>
<tr>
<td>0.55</td>
<td>320</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>247</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>0</td>
<td>400</td>
<td>400</td>
<td>400</td>
<td>400</td>
<td>400</td>
<td>400</td>
<td>400</td>
<td>400</td>
<td></td>
</tr>
<tr>
<td>0.05</td>
<td>425</td>
<td>430</td>
<td>429</td>
<td>430</td>
<td>425</td>
<td>426</td>
<td>411</td>
<td>444</td>
<td></td>
</tr>
<tr>
<td>0.10</td>
<td>422</td>
<td>447</td>
<td>447</td>
<td>437</td>
<td>431</td>
<td>434</td>
<td>387</td>
<td>472</td>
<td></td>
</tr>
<tr>
<td>0.15</td>
<td>408</td>
<td>457</td>
<td>457</td>
<td>430</td>
<td>429</td>
<td>432</td>
<td>359</td>
<td>470</td>
<td></td>
</tr>
</tbody>
</table>
### Table 12. (Continued)

<table>
<thead>
<tr>
<th>$r/r_w$</th>
<th>Case IIB</th>
<th>Case IIIB</th>
<th>Case IIIA</th>
<th>Case IIA</th>
<th>Case IB</th>
<th>Case IA</th>
<th>Case IV</th>
<th>Experimental (41)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.20</td>
<td>392</td>
<td>462</td>
<td>463</td>
<td>416</td>
<td>422</td>
<td>426</td>
<td>333</td>
<td>452</td>
</tr>
<tr>
<td>0.25</td>
<td>375</td>
<td>463</td>
<td>465</td>
<td>403</td>
<td>412</td>
<td>416</td>
<td>310</td>
<td>427</td>
</tr>
<tr>
<td>0.30</td>
<td>358</td>
<td>463</td>
<td>465</td>
<td>388</td>
<td>401</td>
<td>405</td>
<td>290</td>
<td>398</td>
</tr>
<tr>
<td>0.35</td>
<td>342</td>
<td>460</td>
<td></td>
<td>372</td>
<td>389</td>
<td>393</td>
<td>274</td>
<td>375</td>
</tr>
<tr>
<td>0.40</td>
<td>327</td>
<td>459</td>
<td></td>
<td>356</td>
<td>376</td>
<td>380</td>
<td>261</td>
<td>360</td>
</tr>
<tr>
<td>0.45</td>
<td>314</td>
<td></td>
<td></td>
<td>342</td>
<td>363</td>
<td>366</td>
<td>249</td>
<td>349</td>
</tr>
<tr>
<td>0.50</td>
<td>302</td>
<td></td>
<td></td>
<td>328</td>
<td>352</td>
<td>353</td>
<td>240</td>
<td>342</td>
</tr>
<tr>
<td>0.55</td>
<td>291</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>231</td>
</tr>
</tbody>
</table>

$r/r_w = 0.8$

<p>| | | | | | | | | |</p>
<table>
<thead>
<tr>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>379</td>
<td>379</td>
<td>379</td>
<td>379</td>
<td>379</td>
<td>379</td>
<td>379</td>
<td>379</td>
</tr>
<tr>
<td>0.05</td>
<td>328</td>
<td>388</td>
<td>391</td>
<td>340</td>
<td>369</td>
<td>371</td>
<td>312</td>
<td>393</td>
</tr>
<tr>
<td>0.10</td>
<td>301</td>
<td>396</td>
<td>400</td>
<td>316</td>
<td>361</td>
<td>364</td>
<td>284</td>
<td>393</td>
</tr>
<tr>
<td>0.15</td>
<td>286</td>
<td>402</td>
<td>406</td>
<td>301</td>
<td>354</td>
<td>358</td>
<td>268</td>
<td>374</td>
</tr>
<tr>
<td>0.20</td>
<td>276</td>
<td>406</td>
<td>410</td>
<td>291</td>
<td>347</td>
<td>352</td>
<td>256</td>
<td>350</td>
</tr>
<tr>
<td>0.25</td>
<td>267</td>
<td>408</td>
<td>412</td>
<td>282</td>
<td>340</td>
<td>345</td>
<td>246</td>
<td>325</td>
</tr>
<tr>
<td>0.30</td>
<td>260</td>
<td>408</td>
<td>412</td>
<td>275</td>
<td>332</td>
<td>338</td>
<td>237</td>
<td>300</td>
</tr>
<tr>
<td>0.35</td>
<td>254</td>
<td>407</td>
<td>-</td>
<td>268</td>
<td>324</td>
<td>330</td>
<td>231</td>
<td>280</td>
</tr>
<tr>
<td>0.40</td>
<td>248</td>
<td>406</td>
<td>-</td>
<td>262</td>
<td>316</td>
<td>321</td>
<td>225</td>
<td>267</td>
</tr>
<tr>
<td>0.45</td>
<td>243</td>
<td></td>
<td></td>
<td>256</td>
<td>308</td>
<td>313</td>
<td>220</td>
<td>261</td>
</tr>
<tr>
<td>0.50</td>
<td>239</td>
<td></td>
<td></td>
<td>251</td>
<td>300</td>
<td>305</td>
<td>216</td>
<td>258</td>
</tr>
<tr>
<td>0.55</td>
<td>234</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>212</td>
</tr>
</tbody>
</table>

$r/r_w = 0.9$

<p>| | | | | | | | | |</p>
<table>
<thead>
<tr>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>342</td>
<td>342</td>
<td>342</td>
<td>342</td>
<td>342</td>
<td>342</td>
<td>342</td>
<td>342</td>
</tr>
<tr>
<td>0.05</td>
<td>265</td>
<td>341</td>
<td>343</td>
<td>272</td>
<td>321</td>
<td>322</td>
<td>260</td>
<td>347</td>
</tr>
<tr>
<td>0.10</td>
<td>249</td>
<td>345</td>
<td>348</td>
<td>256</td>
<td>312</td>
<td>314</td>
<td>243</td>
<td>332</td>
</tr>
<tr>
<td>0.15</td>
<td>241</td>
<td>349</td>
<td>353</td>
<td>248</td>
<td>307</td>
<td>309</td>
<td>234</td>
<td>315</td>
</tr>
<tr>
<td>0.20</td>
<td>235</td>
<td>352</td>
<td>356</td>
<td>243</td>
<td>302</td>
<td>305</td>
<td>228</td>
<td>296</td>
</tr>
<tr>
<td>0.25</td>
<td>231</td>
<td>355</td>
<td>358</td>
<td>238</td>
<td>297</td>
<td>300</td>
<td>223</td>
<td>275</td>
</tr>
<tr>
<td>0.30</td>
<td>228</td>
<td>355</td>
<td>358</td>
<td>235</td>
<td>292</td>
<td>295</td>
<td>218</td>
<td>254</td>
</tr>
<tr>
<td>0.35</td>
<td>225</td>
<td>355</td>
<td></td>
<td>232</td>
<td>286</td>
<td>290</td>
<td>215</td>
<td>239</td>
</tr>
<tr>
<td>0.40</td>
<td>222</td>
<td>354</td>
<td></td>
<td>229</td>
<td>281</td>
<td>284</td>
<td>212</td>
<td>231</td>
</tr>
<tr>
<td>0.45</td>
<td>219</td>
<td></td>
<td></td>
<td>226</td>
<td>275</td>
<td>279</td>
<td>209</td>
<td>228</td>
</tr>
<tr>
<td>0.50</td>
<td>217</td>
<td></td>
<td></td>
<td>223</td>
<td>270</td>
<td>273</td>
<td>207</td>
<td>228</td>
</tr>
<tr>
<td>0.55</td>
<td>215</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>206</td>
</tr>
</tbody>
</table>
Figure 56. Information flow diagram for computer program
Calculate rate term

Test: var \( \Delta H \)

no

Calculate enthalpy term

Calculate \( \theta_n + 1 \), m

Collect \( \Gamma \)'s

Test: var \( Pe_h \)

Calculate \( Pe_h \) \( \theta \)'s

Calculate \( E \) \( \theta \)'s

no

Test: var \( E \)

Calculate variable rate term

\( \beta \)

Calculate constant rate term

Test: var vel

Calculate constant rate term

Test: var vel

Calculate variable rate term

Figure 56. (Continued)
Figure 56. (Continued)