# Theoretical Prediction of Heterogeneous One-Dimensional Heat Transfer Coefficients for Fixed-Bed Reactors

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The modeling of nonadiabatic fixed-bed tubular reactors has received considerable attention (Froment and Bischoff, 1979). Recently, Pereira Duarte et al. (1984a, 1984b) compared the behavior of different models and showed the importance of the heterogeneous ones, both in their one- and two-dimensional versions.

The heterogeneous two-dimensional model contains the basic heat transfer parameters that are believed to be significant and that can be determined experimentally (Dixon and Cresswell, 1979). The purpose of this work is to obtain analytic expressions for the heterogeneous one-dimensional heat transfer coefficients in terms of these basic parameters and to analyze its use in fixed-bed reactor models.

The steady state equations for a single reaction according to the heterogeneous one- and two-dimensional models were presented by Pereira Duarte et al. (1984b). In order to compare these models it is necessary to ensure equal heat fluxes. If the radial mean value of the convection heat fluxes is set equal it follows that

$$\bar{\theta}_f = \tau_f \tag{1}$$

Similarly, from the radial mean value of the interphase heat flux and from Eq. 1

$$\bar{\theta}_s = \tau_s \tag{2}$$

In addition, if the same heat flux to the surroundings is set in both models, it follows that

$$\alpha_i^f \tau_f = \alpha_w^f \theta_f|R \tag{3a}$$

$$\alpha_i^s \tau_s = \alpha_w^s \theta s|R \tag{3b}$$

and therefore

$$\alpha_i^f = \alpha_w^f / R_f \qquad \alpha_i^s = \alpha_w^s / R_s \tag{4a, b}$$

where

$$R_f = \bar{\theta}_f / \theta_{f|R} \quad \text{and} \quad R_s = \bar{\theta}_s / \theta_{s|R}$$

From these equations the heat transfer coefficients of the heterogeneous one-dimensional model can be calculated.

Since radial temperature profiles vary along a reactor, variable coefficients will be obtained from the numerical integration of the two-dimensional heterogeneous model equations. However, if the specific heat generation rate is uniform, analytic expressions for the heat transfer coefficients can be obtained. Following Olbrich's (1970) analysis, we will assume negligible axial variation of temperature and specific heat generation rate independent of the radial position. If we include a heat generation term in both phases, the dimensionless heat balance equations, according to the heterogeneous two-dimensional model, are

$$\frac{1}{x}\frac{d}{dx}\left(x\frac{d\theta_f}{dx}\right) + N_f(\theta_s - \theta_f) + Q_f = 0 \tag{5a}$$

$$\frac{1}{x}\frac{d}{dx}\left(x\frac{d\theta_s}{dx}\right) - N_s(\theta_s - \theta_f) + Q_s = 0 \tag{5b}$$

with boundary conditions

$$\frac{d\theta_f}{dx} = \frac{d\theta_s}{dx} = 0 \qquad \text{at} \qquad x = 0 \tag{6a,b}$$

$$-\frac{d\theta_f}{dx} = Bi_f\theta_f; \ -\frac{d\theta_s}{dx} = Bi_s\theta_s \qquad \text{at} \qquad x = 1 \tag{6c,d}$$

The system of Eqs. 5 and 6 can be solved following the procedure presented by Olbrich (1970) and the solution given by Hildebrand (1976). The following expressions for the radial temperature profiles arise:

$$\theta_f = \left[\frac{a_1}{k^2} I_o(kx)\left(\frac{1}{k^2} - \frac{1}{N_s}\right) + \frac{N_f}{k^2}\left(\frac{R_g}{N_s} - \frac{k^2}{N_s N_f} - \frac{x^2 R_g}{4}\right) + a_2\right] Q_s \tag{7a}$$

$$\theta_s = \left[\frac{a_1}{k^4} I_o(kx) - \frac{N_f R_g}{k^2}\frac{x^2}{4} + a_2\right] Q_s \tag{7b}$$

where

$$R_g = 1 + \frac{N_s Q_f}{N_f Q_s}; \quad a_1 = \frac{b_1}{Q_s}; \quad a_2 = \frac{b_2}{Q_s};$$

$$k^2 = N_f + N_s \tag{7c,d,e,f}$$

$$b_1 = \frac{k^2 Q_s R_g N_f (N_s^{-1} + 0.5\,Bi_s - 0.5\,Bi_f - k^2 N_s^{-1} N_f^{-1} R_g^{-1})}{\left[I_o(k) + \frac{kI_1(k)}{Bi_s}\right] - \left(1 - \frac{k^2}{N_s}\right)\left[I_o(k) + \frac{kI_1(k)}{Bi_f}\right]} \tag{7g}$$

$$b_2 = \frac{N_f Q_s R_g}{k^2}\left(\frac{1}{4} + \frac{1}{2Bi_s}\right) - \frac{b_1}{k^4}\left[I_o(k) + \frac{kI_1(k)}{Bi_s}\right] \tag{7h}$$

From Eqs. 7, expressions for $R_f$ and $R_s$ can be obtained:

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$$R_f = \frac{2a_3k^{-1}I_1(k) - N_fR_g/(8k^2) + a_2 + a_4}{a_3I_o(k) - N_fR_g/(4k^2) + a_2 + a_4} \quad (8a)$$

$$R_s = \frac{2(a_1/k^4)k^{-1}I_1(k) - N_fR_g/(8k^2) + a_2}{(a_1/k^4)I_o(k) - N_fR_g/(4k^2) + a_2} \quad (8b)$$

where

$$a_3 = \frac{a_1}{k^2}\left(\frac{1}{k^2} - \frac{1}{N_s}\right) \quad (8c)$$

$$a_4 = \frac{R_g}{N_s}\left(\frac{N_f}{k^2} - \frac{1}{R_g}\right) \quad (8d)$$

These are asymptotic values, since they are to be reached in a system with constant heat generation and heat transfer to the surroundings. It can be seen that both $R_f$ and $R_s$ are independent of the individual values of $Q_s$ and $Q_f$ and depend only on their ratio through $R_g$. Generally, in a catalytic reactor, $R_g = 1$, since there is no heat generation or consumption in the fluid phase.

## ANALYSIS OF THE LIMITING CASES OF THE ASYMPTOTIC COEFFICIENT

1. Infinitely large interphase resistance ($N_f, N_s \to 0$):

In this case both phases do not interact and Eqs. 5a and 5b are independent. Parabolic temperature profiles are obtained in each phase and

$$R_f = 1 + \frac{Bi_f}{4}; \quad R_s = 1 + \frac{Bi_s}{4} \quad (9)$$

2. Negligible interphase resistance ($N_f, N_s \to \infty$):

In these cases $\theta_f = \theta_s = \theta$ and a pseudohomogeneous model is strictly valid. It follows that

$$R_f = R_s = 1 + \frac{Bi}{4} \quad (10a)$$

where

$$Bi = \frac{\alpha_w^f + \alpha_w^s}{\lambda_{er}^f + \lambda_{er}^s} \cdot R = \frac{(N_s/N_f)Bi_f + Bi_s}{(N_s/N_f) + 1} \quad (10b)$$

The asymptotic values of $R_f$ and $R_s$ arising from Eqs. 8 have been represented in Figure 1 as a function of $N_f$. It can be seen that for $N_f$ less than 1 and greater than $10^6$ they coincide, respectively, with those arising from Eqs. 9 and 10 (limiting cases). However, the dimensionless parameters $N_f$ and $N_s$, which represent the degree of interaction between phases, depend on the system variables ($Re$, $dp$, etc.) and cannot be varied arbitrarily. For large $R/dp$ ratios, pseudo-homogeneous conditions are found, although $N_f$ and $N_s$ remain finite; on the other hand, for small ratios, the values of $N_f$ and $N_s$ decrease but seldom drop below 1. Thus, when the basic parameters that appear in the expressions for the heat transfer coefficients are calculated in actual conditions, from correlations found in the literature, differences are found between the asymptotic values of the heat transfer coefficients and those arising from the limiting equations. Nevertheless, these deviations are smaller when Eq. (10) is used to evaluate the heat transfer coefficient (Pereira Duarte et al., 1984b).

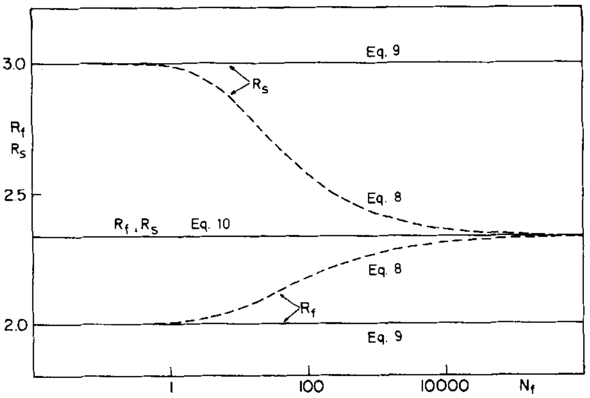


**Figure 1. Variation of $R_f$ and $R_s$ as a function of $N_f$. $Bi_s = 8$, $Bi_f = 4$, $N_s/N_f = 2$, $Q_f/Q_s = 1$.**

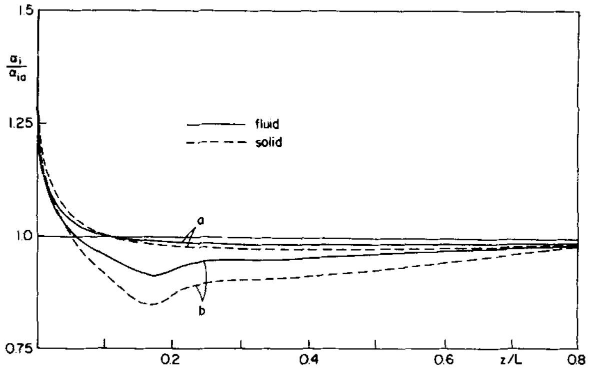


**Figure 2. Ratio between the variable and asymptotic coefficients. $Re = 500$, $R = 0.0127$ m, $d_p = 0.008$ m, $\beta = 1$, $a - \gamma_o = 10$, $b - \gamma_o = 25$.**

## INFLUENCE OF THE HEAT TRANSFER COEFFICIENTS ON THE BEHAVIOR OF THE HETEROGENEOUS ONE-DIMENSIONAL MODEL

The heterogeneous one-dimensional model for a nonadiabatic fixed-bed reactor was proposed by Pereira Duarte et al. (1984b) and, from the analysis made in this paper, three ways of evaluating the heat transfer coefficients arise: (a) from the numerical solution of the two-dimensional model equations and taking into account Eq. 4, and will be called variable coefficients; b) from Eqs. 4 and 8, namely, asymptotic coefficients; and (c) from Eqs. 4 and 10, namely, approximate coefficients. It is obvious that the variable coefficients will generate the closest response to the two-dimensional model, but their use is cumbersome. The ratio between the variable and the asymptotic coefficients is plotted in Figure 2 as a function of the reactor length for different activation energies. It can be seen that the largest differences are found at the inlet and close to the hot spot. The former do not depend on the kinetic conditions but they are due to the entrance effects (nondeveloped profiles), and the assumption of negligible axial variation of temperature becomes meaningless. This effect was analyzed by Li and Finlayson (1977) and showed that, although it can be neglected in the design of an industrial reactor, it must be considered in the analysis of experimental data. From our initial condition ($\theta_f = 0$), it is obvious that the ratio in Figure 2 must be greater than 1 close to the inlet. However, as we move along the reactor, this effect becomes less important and the chemical reaction plays the major role, especially close to the hot spot, since the heat generation rate is not uniform. This effect becomes more important as $\gamma_o$ increases, since the radial temperature profiles deviate from those arising from a uniform heat generation rate. It can also be seen from Figure 2 that for mild reactions this effect is small.

The differences between the conversions at the hot spot, obtained from the heterogeneous one- and two-dimensional models

$$e = \frac{X_o - \bar{X}_T}{\bar{X}_T} \cdot 100 \quad (11)$$

are shown in Table 1. As it can be expected, the variable coefficients generate better results, although the error increases with $\gamma_o$. From the analysis of Figure 3 it follows that the effect of $\beta$ is similar to that found for $\gamma_o$. It can also be seen that the errors introduced by the asymptotic coefficients are smaller than those for the approximate ones.

As the thermal sensitivity of the system increases, the errors, for any of the heat transfer coefficients, increase. Even for the variable coefficients significant errors are found, since in a one-dimensional model the reaction rate is evaluated in terms of the radial mean conditions.

TABLE 1. DIFFERENCES BETWEEN CONVERSIONS AT THE HOT SPOT

| Coefficient | $\gamma_o$ = 10 | 20 | 25 |
|---|---|---|---|
| Variable | −0.0949 | −2.915 | −6.849 |
| Asymptotic | 0.102 | −3.519 | −18.685 |

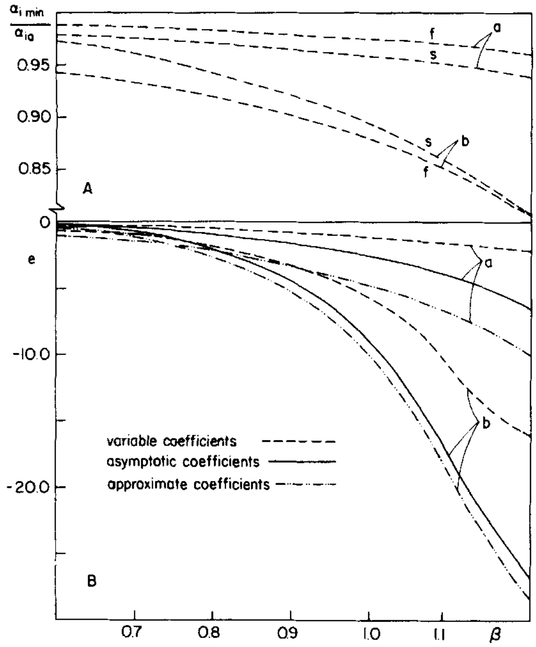


**Figure 3. Minimum values of the ratio between variables and asymptotic coefficients and error of the one-dimensional model. $\gamma_o$ = 20, $R$ = 0.0127 m, $\epsilon$ = 0.4, $s$: solid phase, $f$: fluid phase. $a$: $Re$ = 50, $d_p$ = 0.008 m. $b$; $Re$ = 500, $d_p$ = 0.002 m.**

## CONCLUSIONS

Analytic expressions have been obtained for the evaluation of the asymptotic radial heat transfer coefficients for a heterogeneous one-dimensional-fixed bed reactor model. From the analysis of the behavior of the one-dimensional reactor model it follows that the asymptotic heat transfer coefficients generate a better response, when compared with the two-dimensional model, than the approximate coefficients used before. It can also be established that, for any of the heat transfer coefficients used, the one-dimensional model must only be used for mild thermal conditions (moderate values of $\beta$ and $\gamma_o$).

This work shows the importance of the theoretical analysis in order to obtain the transfer coefficients of simplified models from more complete ones, like the heterogeneous two-dimensional. The close relationship between the transfer coefficients and the chemical reaction is also shown. It can also be concluded that it is important and necessary to obtain correct experimental values of the basic transfer parameters of the two-dimensional model. This will allow exact evaluation of the parameters in any simplified model derived from it.

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## NOTATION

- $a_v$ = external surface area of pellet per unit reactor volume, m$^{-1}$
- $Bi, Bi_f, Bi_s$ = Biot number, pseudohomogeneous, for fluid phase, for solid phase
- $d_p$ = pellet diameter
- $e$ = error in conversion, Eq. 11
- $h_f$ = heat transfer coefficient between pellet and fluid, kcal/h·m²·K
- $I_o, I_1$ = modified Bessel functions of zero and first order
- $k$ = parameter defined in Eq. 7f
- $L$ = bed length, m
- $N$ = dimensionless parameter, $= h_f a_v R^2/\lambda_{er}$
- $Q_G$ = heat generated per unit bed volume
- $Q$ = dimensionless heat generated, $= Q_G R^2/T_w \lambda_{er}$
- $R$ = tube radius
- $Re$ = Reynolds number
- $R_f, R_s$ = parameters defined in Eq. 4
- $R_g$ = parameter defined in Eq. 7c
- $t$ = temperature (one-dimensional model), K
- $T$ = temperature (two-dimensional model), K
- $x$ = dimensionless radial coordinate
- $X_o$ = conversion (one-dimensional model)
- $X_T$ = conversion (two-dimensional model)
- $Z$ = axial coordinate, m

### Greek Letters

- $\alpha_i^f, \alpha_i^s$ = one-dimensional heat transfer coefficient for the fluid phase; for the solid phase, kcal/m²·h·K
- $\alpha_w^f, \alpha_w^s$ = wall heat transfer coefficient for the fluid phase; for the solid phase, kcal/m²·h·K
- $\beta$ = adiabatic temperature rise
- $\gamma_o$ = dimensionless activation energy
- $\epsilon$ = bed porosity
- $\lambda_{er}^f, \lambda_{er}^s$ = radial effective thermal conductivity for the fluid phase; for the solid phase, kcal/h·m·K
- $\tau$ = dimensionless temperature, $= (t - t_w)/t_w$ (one-dimensional model)
- $\theta$ = dimensionless temperature, $= (T - T_w)/T_w$ (two-dimensional model)

### Subscripts and Superscripts

- $a$ = asymptotic value
- $f$ = fluid conditions
- $s$ = solid conditions
- $w$ = wall conditions
- $-$ = mean value

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