

Effective Thermal Conductivity in a Packed-Bed Radial-Flow Reactor

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The use of radial-flow packed-bed reactors has increased substantially in recent years, particularly for processes such as ammonia synthesis, reforming, and denitrogenation where high space velocities are required. Vek (1977) reported that full-scale radial-flow ammonia converters produced twice the specific output of conventional tubular reactors. Hence, a complete study of the heat transfer through the packed bed of such reactors is important for a better understanding and a more efficient design of these units. Furthermore, a precise knowledge of effective thermal properties is needed for the analysis of stability phenomena in fixed-bed exothermal reactors.

To date very few papers have presented engineering data that might be used for the design of radial flow reactors, possibly because consulting and design companies consider the data to be proprietary. Hlavacek and Vittoria (1977) recommended the use of data measured in tubular reactors for radial flow if a logarithmic average radius is adopted. More recent published work (Pinto and Kaye, 1979; Balakrishna and Luss, 1981; Chang and Calo, 1981; Ching et al., 1983) has been addressed mainly to the influence of imperfect radial velocity profiles on performance. An experimental study of heat transport in radial flow reactors was carried out by Vittoria and Hlavacek (1972). They provided experimental information about the effective thermal conductivity by making steady state measurements and concluded that the mean radius of the reactor would be the most important parameter under consideration. The present research looks into the heat transport characteristic of this type of reactor, develops an experimental method for estimating the effective thermal conductivity under nonsteady conditions, and compares results with tubular reactor data. Experiments were carried out at low Reynolds numbers of flow, which appears to be the most difficult conditions for determining axial thermal coefficients in tubular reactors. A literature search by computer has indicated so far that this study is the first experimental work done on a radial-flow reactor under nonsteady conditions.

Experimental Method

Figure 1 shows the flow diagram of the equipment used. It consists of a radial-flow prototype reactor placed inside a heat-insulated cylinder; a set of valves controlling not only the flow of warm and cool air but also the entrance temperature of the reactor feed; and an automatic data processing system connected to the reactor thermocouples to register temperature changes in the packed bed.

A detailed diagram of the reactor is shown in Figure 2. It is composed of two coaxial cylinders of different diameter constructed of stainless steel sheets, fixed by means of two discs with concentric grooves cut in them with dimensions corresponding to the major and minor circumferences of the reactor cylinders. A distributing tube, perforated with small, uniformly spaced orifices is placed in the axis to ensure correct radial flow of air through the packed bed. Temperatures were measured and reg-

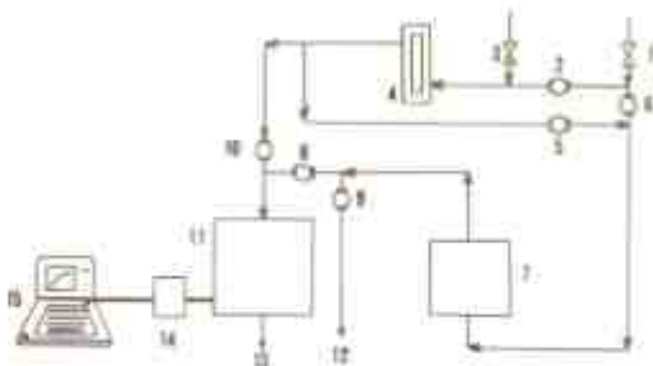


Figure 1. Equipment diagram.

- | | |
|--------------------------------|-----------------------------|
| 1, 3. Data valves | 11. Reactor vessel |
| 2, 5, 6, 8, 9, 10. Ball valves | 12. Drain |
| 4. Recorder | 13. Data acquisition system |
| 7. Electrical heater system | 14. Microcomputer |

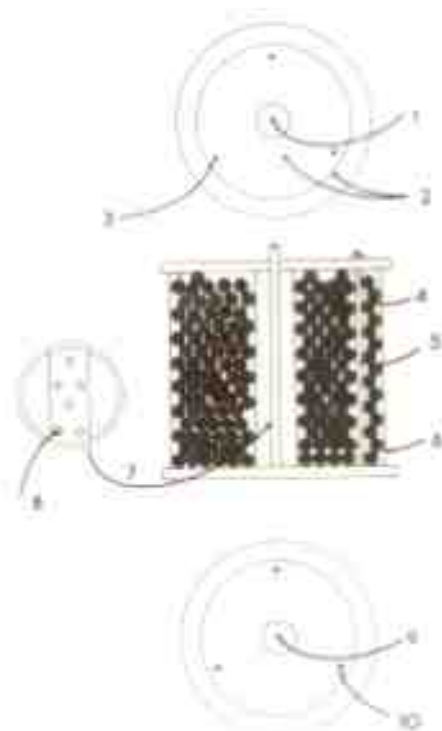


Figure 2. Radial-flow reactor.

- | | |
|-------------------------------------|----------------------------|
| 1. Distributor tube with upper disk | 7. Distributor tube |
| 2. Zones for perforated zones | 8. 1/2 in. holes (0.79 mm) |
| 3. Support struts | 9. Distributor base |
| 4. Reactor packing | 10. Bottom disk |
| 5. Stand | |

istered as shown in Figure 3. T-type thermocouples (copper-constantan) are placed in a small guide tube, sealing the edges with cement. An automatic data processing system, locally constructed (Tijez, 1985), entirely controlled by a microcomputer (Apple IIe) is connected to the thermocouples, allowing measurement of temperatures in a data base program for later analysis.

The packing material used was nonreacting polymer particles with an average diameter between 2×10^{-4} and 3×10^{-4} m. The maximum temperature of the warm air was limited by the melting point of the polymer; the optimum operation range found experimentally was 50 to 60°C. The cool air temperature

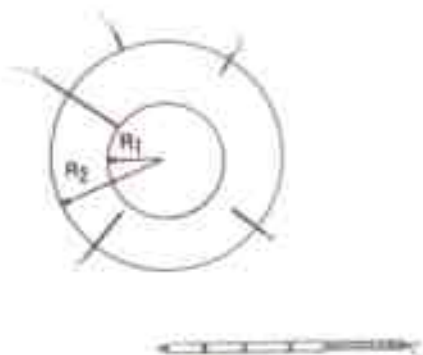


Figure 3. Reactor thermocouple positions.

- R_1 , inner radius = 1 cm.
 R_2 , outer radius = 11 cm.

was 23°C. The bed's axial and angular symmetry was verified for each experiment. In fact, temperature varied, in the worst case by 1.5°C. After everything was set, the temperature of the air going into the reactor was step-increased and the temperature response of the packed bed was measured as indicated above, repeating the experiment several times under different conditions of air flow and particle diameter. The air flow range was from 4.23 to 9.91 m^3/h , the particle diameter range was from 0.2 to 0.5 cm.

Mathematical Model

Temperature variations in the packed bed are analyzed using a pseudohomogeneous model that does not make any distinction between solid and fluid temperature. In effect, it has been shown (Sims, 1984) that local temperature equilibrium between phases is achieved if the following constraint is obeyed:

$$(\Delta p^2 k_f / (R_s - R_i)^2) (1/k_f + 1/k_s) < 1 \quad (1)$$

For the most critical conditions used in this work, the above constraint is less than 10^{-5} , which shows that the pseudohomogeneous model is a valid assumption. Therefore, a differential heat balance for this model is expressed by an equation such as:

$$\frac{\partial T}{\partial R^2} + \frac{1}{R} (1 - P_0) \frac{\partial T}{\partial R} - \frac{\partial T}{\partial t} \quad (2)$$

$$r = 0 \quad T = 1 \quad \text{at } R \quad (3)$$

$$r = 0 \quad \partial T / \partial R = P_0 T / (R - 1) \quad (4)$$

$$r = 0 \quad \partial T / \partial R = 0 \quad R = R_0 / R_1 \quad (5)$$

The most crucial assumption made in arriving at Eq. 2 was the invariability of the effective conductivity with position. Experimental results discussed later show that it indeed remains constant with radius. Initially, the reactor is at low temperature, T_0 , with air flowing at the same temperature. Afterward, the air

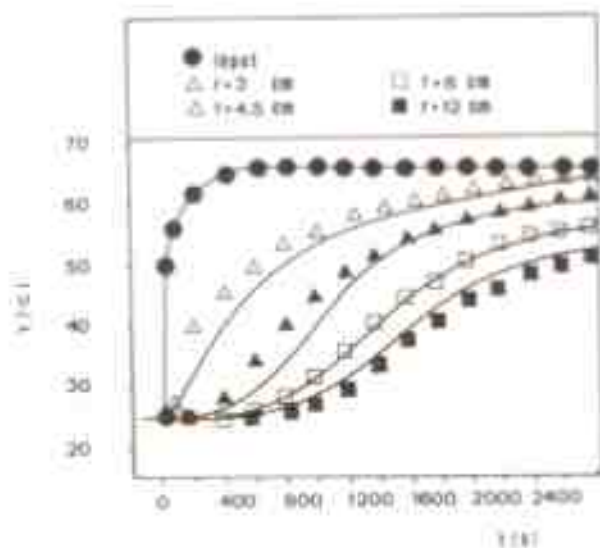


Figure 4. Bed temperature response.
 $\dot{V} = 0.00173 \text{ m}^3/\text{s}$; $d_p = 0.2 \text{ cm}$.

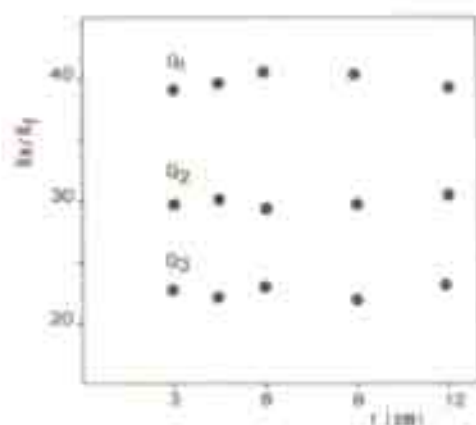


Figure 5. Variation of k_e/k_f with radial position.

$$\begin{aligned} Q1 &= 0.00113 \text{ m}^2/\text{s} \\ Q2 &= 0.00236 \text{ m}^2/\text{s} \\ Q3 &= 0.00571 \text{ m}^2/\text{s} \\ \Delta r &= 0.37 \text{ cm} \end{aligned}$$

temperature changes in a higher value, T_1 , and the response of the packed bed should follow the solution of Eqs. 2-5, which is given by an infinite series:

$$T(R, t) = \sum_{n=1}^{\infty} C_n [S_{n(1)}(R)]^{n(1)} J_{n(1)}(\lambda_n R) - V_{n(1)}(R)^{n(1)} J_{n(1)}(\lambda_n R) e^{-\lambda_n^2 t} \quad (6)$$

where the λ_n 's eigenvalues are roots of the following equation:

$$W_{n(1)} V_{n(1)} - S_{n(1)} U_{n(1)} = 0 \quad (7)$$

Details of the solution leading to a modified Bessel equation

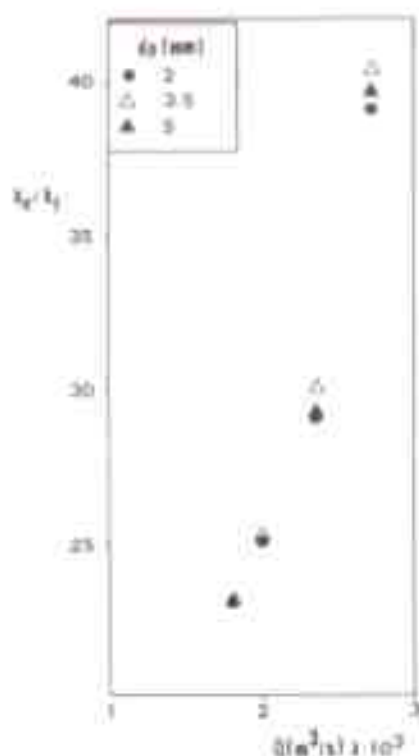


Figure 6. Particle diameter effect on k_e/k_f .

and the form of W , V , S , and U functions are given in supplementary material.

Equation 6 can be represented as: $T(R, t) = \sum_{n=1}^{\infty} a_n f_n(R) g_n(t)$ where the gradually decreased coefficient, a_n , is obtained from the initial condition. The $f_n(R)$ functions are solutions of the Sturm-Liouville system and they depend on the boundary conditions and the geometry of the problem. The $g(t)$ function is an exponential of negative increasing argument and is independent of the initial condition. Therefore, if an R value is set, then each term of the series is lower than the preliminary one, and for a given t value the infinite series approaches the first term. In effect, calculations indicated that the error made by taking only the first term is less than 4%. Thus, applying logarithm a straight line is obtained:

$$\ln T(R, t) = \ln f_1(R) - \lambda_1^2 t \quad (8)$$

A slope for Eq. 8 may be found by linear regression, and using the eigenvalue equation, Eq. 7, a value for the effective thermal conductivity k_e can be determined, since W , V , S , and U are only functions of the radial Peclet number. Furthermore, it was found that the slopes obtained from any thermocouple position were practically the same for each run. This method has the feature that no moment matching is required, nor is a complete solution needed for superimposition over the data to fit the thermal coefficient.

Results and Discussion

If the packed bed of the reactor and gas flowing through it are considered as a hypothetical homogeneous phase, then an effective thermal conductivity can be defined. It will depend certainly on the nature of the fluid, temperature, diameter of the packing particles, porosity of the bed, conductivities of solid and gas, fluid velocity, and probably on the geometry of the system. It is usually determined by comparing temperature profiles experimentally obtained from a step or pulse change in the entrance of the corresponding differential equation. In Figure 4 a typical radial bed temperature response to a step change is shown from this work for a given particle size and air flow. It is

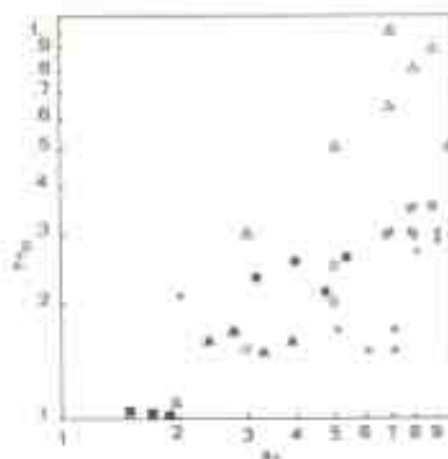


Figure 7. Comparison of results.

$$\begin{aligned} \bullet \blacksquare \text{ This work} \\ \triangle \text{ Ueno and De Souza (1974)} \\ \square \text{ Yagi et al. (1980)} \\ \times \text{ Yoneda and Hasegawa (1972)} \end{aligned}$$

observed that temperatures tend to become uniform, and at times greater than 2,000 s curves at different radial positions tend to approach each other, as expected for the new steady state. Although it is not necessary to find the complete solution to obtain a value of the thermal coefficient, a comparison between experimental data and the calculated values for each thermocouple position is shown with good agreement. Figure 5 shows a plot of k_a/k_f vs. radius for three volumetric flows, indicating that the effective thermal conductivity seems to be independent of position. Figure 6 shows a plot between k_a/k_f and the air volumetric flow for three particle sizes, and as expected the effective thermal conductivity increases with the flow. The figure also shows that particle size has no significant effect for volumetric flow lower than about $8 \text{ m}^3/\text{h}$ at standard conditions.

In Figure 7 results of this work are compared with values of particle Peclet number (Pe_p) obtained in tubular reactors for air and solid particles (unsteady data from Gunn and De Souza, 1974, steady data from Yagi et al., 1960). It is observed that values are of the same order of magnitude. For this comparison, an average radius between R_i and R_o of the radial flow reactor is taken as the equivalent radius for the tubular reactor. It seems from the results of this work that axial thermal conductivities obtained from tubular reactors can be used for the design of radial flow reactors and vice versa. However, reliable correlations for the axial thermal conductivity coefficients seem unavailable, particularly for low and intermediate Reynolds numbers (Dixon and Crewell, 1979). The unavailability comes mainly from the fact that in tubular reactors experiments, account has to be taken of radial thermal dispersion and wall heat transfer coefficients even for supposedly isolated systems. In this regard, the experimental method proposed in this work would certainly eliminate this inconvenience. In addition, in tubular reactors the variation in void fraction becomes significant as the wall is approached (larger near the wall compared to center); in a well-packed radial-flow reactor a constant void fraction is expected. Further, this method would be independent of the reactor to particle diameter ratio. Comparison is also made in Figure 7 with radial flow steady state data reported by Votruba and Hlavacek (1972). The general tendency of their results, considering only the working range of this research, is similar in this respect. Their main conclusion about the dependence on reactor radius has not yet been confirmed, since more data would be required. Finally, one must be aware of discrepancies when comparing results from steady and unsteady results, however, for the operating conditions of this work equivalence seems to apply.

Notation

- C_p = fluid heat capacity, $\text{kJ/kg} \cdot \text{K}$
 C_s = solid heat capacity, $\text{kJ/kg} \cdot \text{K}$
 d_p = particle diameter, m
 J_p = Bessel function of the first kind and order p
 k_e = effective thermal conductivity, $\text{W/m} \cdot \text{K}$
 k_f = fluid thermal conductivity, $\text{W/m} \cdot \text{K}$
 Pe = radial Peclet number = $\rho C_p v R_i / k_e$

- Pe_p = particle Peclet number = $\rho C_p v d_p / k_e$
 Q = volumetric flow, m^3/s
 R = dimensionless radius = r/R_i
 R_i = internal radius of reactor, m
 R_o = external radius of reactor, m
 Re = particle Reynolds number = $\rho v d_p / \mu$
 t = dimensionless time = $(k_e / (\rho C_p) R_i$
 T = dimensionless temperature = $(T) - T_o / (T_o - T_i)$
 T_o = cool air temperature, K
 T_i = warm air temperature, K
 v = superficial velocity for radius r , m/s
 v_o = superficial velocity at an average radius ($R_i + R_o$)/2

U, V, W, X = functions in the root k_a , γ defined in supplementary material

Greek letters

- ϵ = void fraction
 λ_e = a root of the eigenvalues equation
 μ = viscosity, $\text{Pa} \cdot \text{s}$
 ρ_o = fluid density, kg/m^3
 ρ_s = solid density, kg/m^3
 τ = time, s

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