EFFECTIVE THERMAL CONDUCTIVITY IN A RADIAL-FLOW PACKED-BED REACTOR[1]

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ABSTRACT

In this work a theoretical and experimental study of the heat transfer process in a radial flow reactor was carried out under steady and non-steady conditions in order to determine the effective thermal conductivity (k_e). One of the mathematical model proposed was the pseudo-homogeneous with effective thermal conductivity that varies with radial position. The second model studied was the two phase model with different thermal conductivities for gas and solid. For the pseudohomogeneous model, an analytical solution was obtained using the method of separation of variables and series approximation. In the two-phase model, the gas and solid temperature profiles were obtained by two numerical methods: orthogonal collocation and Runge-Kutta. Several experiments were made by changing particle diameter, gas flow and temperature input, reactor size and time-operation condition: steady and non-steady. Theoretical results were compared with experimental data in order to calculate the effective thermal conductivity. The values of k_e agree in general with the literature data. At low Reynolds numbers there is no appreciable difference between a pseudohomogeneous model and a two phase equation model. Constant thermal properties can be used at Re<5 with enough accuracy to predict the thermal behavior of a radial-flow reactor.

KEY WORDS: Effective thermal conductivity; packed-bed; pseudohomogeneous model; radial-flow; transient model; steady-state model.

1. INTRODUCTION

Radial-flow packed-bed reactors are used in certain processes where high space velocities are required [1]. 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. It is necessary a precise knowledge of effective thermal properties (*i. e.* effective thermal conductivity, k_e) in order to perform a stability phenomena analysis in the case of fixed-bed exothermal reactors.

Several authors have been working in this area. For example, Hlavácek and Votruba [2] recommended the use of data measured in tubular reactors for radial flow adopting a logarithmic average radius. Kunii and Smith, Swift, Kobayashi, Godbee and Ziegler, Kuzay, Bauer and Schünder, Jaguaribe and Beasly, Nozad *et al.* [3, 4, 5, 6, 7, 8, 9, 10, 11] recommended different methods for the evaluation of the stagnant effective thermal conductivity (k_e^0: effective thermal conductivity at zero velocity). Yagi *et al.*, Kunii and Smith, Votruba *et al.*, Gunn and De Souza, and Dixon and Cresswell [12, 13, 14, 15, 16] calculated the effective thermal conductivity in axial-flow packed-bed reactors using a steady-state model. Additionally, Juang and Weng, Levec and Carbonell, and Dixon and Creswell [17, 18, 19] worked with axial-flow packed-bed reactors but at transient conditions. Finally, Votruba and Hlavácek, Pulve *et al.*, López de Ramos and Pironti and Fuentes *et al.* [20, 21, 22, 23] studied the heat transfer process in radial-flow packed-bed reactors using stationary (the first two) and transient models (the last two authors).

The objective of this work was to calculate the effective thermal conductivity using steady state and transient models, considering that k_e depends on the radial position.

2. EXPERIMENTAL METHODS

The flow diagram of the equipment used (Fig. 1) consists of: a radial-flow reactor placed inside a heat insulated cylinder; a set of valves controlling the cold and hot air entrances to the reactor; an automatic data processing system connected to the reactor thermocouples to register temperatures changes in the packed bed. The reactor (Fig. 2) is composed of two coaxial cylinders of different diameter constructed of stainless steel sieves, fixed by means of two discs with concentric grooves cut in them with dimensions corresponding to the major and minor circumferences of the reactors cylinders. A distributing tube, perforated with small, uniformly spread orifices is placed in the axis to ensure correct radial flow of air through the packed bed. An electric tubular resistance is placed inside the distributing tube if the reactor works under steady state conditions. Temperatures were measured and registered in radial, angular and axial positions (16 ports in total). T-type thermocouples (copper-constantant) are placed is a small guide tube, sealing the edge with cement. The packing material used was non-reacting polymer and ceramic particles with average diameters between $2x10^{-3}$ and $5x10^{-3}$ 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 was 23°C. The bed's axial and angular symmetry was verified for each experiment. In fact, temperatures varied, in the worst case, by 1.5°C. In the transient case, the temperature of the air going into the reactor was step-increased. In the steady state case, the tubular electric resistance is set on using a Variac. The air flow range was from 6.23 to 9.91 m^3/h.

3. MATHEMATICAL MODEL

3.1 Homogeneous model

Temperature variations inside the bed are analyzed using a pseudohomogeneous model that does not make any distinction between solid and fluid temperature. A differential heat balance for this model is expressed by an equation such as:

$$\langle \rho Cp \rangle \frac{\partial T}{\partial t} + \rho_f Cp_f u \frac{\partial T}{\partial r} = \left(\frac{\partial k_e}{\partial r} + \frac{k_e}{r} \right) \frac{\partial T}{\partial r} + k_e \frac{\partial^2 T}{\partial r^2} \tag{1}$$

where, $\langle \rho Cp \rangle$ is the average heat capacity between solid and fluid, T is the temperature, t the time, ρ_f is the fluid density, Cp_f is the fluid heat capacity, u is the fluid superficial velocity, r is the radial position, and k_e is the effective thermal conductivity.

Yagi et al.[12] have found experimentally that the effective thermal conductivity for axial flow in tubular reactors varies linearly with fluid velocity according with the following expression:

$$\frac{k_e}{k_f} = \frac{k_e^0}{k_f} + \delta \, Pr \, Re \tag{2}$$

where, k_f is the thermal conductivity of the fluid, k_e^0 is the effective thermal conductivity for a stagnant fluid, δ is a correlation parameter, Prandtl number calculated as $Pr = \frac{Cp\mu}{k_f}$, Re is Reynolds number calculated as $Re = \frac{r_f u D_p}{m}$.

In equation (1) it is assumed that the effective thermal conductivity, k_e, is a function of the radial position through the fluid velocity as stated in equation (2), considering that in radial flow reactors the continuity equation is given by:

$$\frac{1}{r}\frac{\P(r\rho_f u)}{\P r} = 0 \tag{3}$$

3.1.1 Steady State Case:

The differential equation for the steady state case is the Eq. (1) without the first term. The boundary conditions applied to this problem were:

$$\begin{cases} T = T_1 \text{ at } r = R_1 \\ T = T_2 \text{ at } r = R_2 \end{cases} \tag{4}$$

The solution is:

$$\frac{T - T_2}{T_1 - T_2} = \theta = \frac{\left(R_2 + \delta Pe * d_p\right)^{Pe*} - \left(r + \delta Pe * d_p\right)^{Pe*}}{\left(R_2 + \delta Pe * d_p\right)^{Pe*} - \left(R_1 + \delta Pe * d_p\right)^{Pe*}} \tag{5}$$

where Pe* is a modified Peclet number given by: $Pe* = \frac{\rho_f urCp}{k_e^0}$

3.1.2 Non-steady Case:

Equation (1) is the differential equation for the non-steady case. The initial and boundary conditions applied were:

$$\begin{array}{lll} t = 0 & T = T_0 & R_1<r<R_2 \\ t > 0 & k_e \frac{\partial T}{\partial r} = \rho_f Cp_f u\left(T - T_1\right) & r = R_1 \\ t > 0 & \frac{\partial T}{\partial r} = 0 & r = R_2 \end{array} \tag{6}$$

The analytical solution of Eq. (1) with the initial and boundary conditions (6) is:

$$\phi(r,\tau)=\frac{T-T_1}{T_0-T_1}=\sum_{i=0}^{\infty}C_i\left[\phi_i^r(r)\right]\exp\left(-\frac{\lambda_i^2\tau}{1+H}\right) \tag{7}$$

where:

$$\phi_i^r(r)=C1_i\sum_{j=0}^{\infty}a_j(\lambda_i)\left(r+\delta\alpha Pe^*\right)^j+C2_i\sum_{j=0}^{\infty}b_j(\lambda_i)\left(r+\delta\alpha Pe^*\right)^{j+Pe^*}$$

$$H=\frac{\varepsilon\rho_f Cp_f}{(1-\varepsilon)\rho_s Cp_s} \tag{8}$$

$$\tau=\frac{tk_e^0}{\rho_f Cp_f R_1^2}$$

The coefficients a_j and b_j have the form of:

$$a_0=1 \qquad b_0=1$$

$$a_1=\frac{\delta\alpha Pe^*\lambda^2}{1-Pe^*} \qquad b_1=\frac{\delta\alpha Pe^*\lambda^2}{1+Pe^*} \tag{9}$$

$$a_j=\frac{\lambda^2\left(\delta\alpha Pe^* a_{j-1}-a_{j-2}\right)}{j\left(j-Pe^*\right)};\ j\geq 2 \qquad b_j=\frac{\lambda^2\left(\delta\alpha Pe^* b_{j-1}-b_{j-2}\right)}{j\left(j+Pe^*\right)};\ j\geq 2$$

The eigenvalues λ_i are calculated as the positive roots of the following equation:

$$U(\lambda_i)S(\lambda_i)-W(\lambda_i)V(\lambda_i)=0 \tag{10}$$

where,

$$U(\lambda_i)=\sum_{j=0}^{\infty}ja_j(\lambda_i)\left(\frac{R_2}{R_1}+\delta\alpha Pe^*\right)^{j-1} \qquad W(\lambda_i)=\sum_{j=0}^{\infty}\left(j+Pe^*\right)b_j(\lambda_i)\left(\frac{R_2}{R_1}+\delta\alpha Pe^*\right)^{j+Pe^*-1}$$

$$V(\lambda_i) = \sum_{j=0}^{\infty} \left(j - Pe^*\right) a_j(\lambda_i)\left(1 + \delta\alpha Pe^*\right)^j \qquad S(\lambda_i) = \sum_{j=0}^{\infty} j b_j(\lambda_i)\left(1 + \delta\alpha Pe^*\right)^{j+Pe^*} \tag{11}$$

The constants $C1_i$ and $C2_i$ and C_i can be calculated using the following expressions:

$$C1_i = -W(\lambda_i) \qquad C2_i = U(\lambda_i)$$

$$\underline{C_i} = \underline{\underline{A}}_{ij}^{-1} \underline{B}_i \tag{12}$$

where,

$$A_{i,j} = \int_1^{R_2/R_1} \phi_i^r(r)\phi_j^r(r)dr \qquad B_i = \int_1^{R_2/R_1} \phi_i^r(r)dr \tag{13}$$

3.3 Two-Phase Model:

Temperature variations inside the bed can be analyzed using a two-phase model that makes distinction between solid and fluid temperature. A differential heat balance for this model is expressed by the equations:

$$\frac{1}{r}\frac{\partial}{\partial r}\left(rk_{af}\frac{\partial T_f}{\partial r}\right) - \rho_f Cp_f u\frac{\partial T_f}{\partial r} - ah\left(T_f - T_s\right) = \varepsilon\rho_f Cp_f \frac{\partial T_f}{\partial t} \tag{14}$$

$$\frac{1}{r}\frac{\partial}{\partial r}\left(rk_{rs}\frac{\partial T_s}{\partial r}\right) + ah\left(T_f - T_s\right) = (1-\varepsilon)\rho_s Cp_s \frac{\partial T_s}{\partial t} \tag{15}$$

where k_{af} is the axial fluid phase effective thermal conductivity, k_{rs} is the solid phase effective thermal conductivity and h is the fluid-solid heat transfer coefficient.

The initial and boundary conditions for this model are:

$$t = 0 \rightarrow \begin{cases} T_f = T_0 \\ T_s = T_0 \end{cases} \qquad \forall r \tag{16}$$

$$t > 0 \rightarrow \begin{cases} \dfrac{\partial T_f}{\partial r} = 0 \\ \dfrac{\partial T_s}{\partial r} = 0 \end{cases} \qquad r=R_2 \tag{17}$$

$$t > 0 \rightarrow \begin{cases} k_{af} \dfrac{\partial T_{af}}{\partial r} = \rho_f Cp_f u\left(T_f - T_1\right) \\ T_s = T_f \end{cases} \qquad r=R_1 \tag{18}$$

The correlation reported by Zehner and Schlünder [24] was used to calculate the solid phase effective thermal conductivity, k_{rs}. This conductivity is assumed constant with position:

$$\frac{k_{rs}}{k_f} = \sqrt{1-\varepsilon}\,\frac{k_{rs}^0}{k_f} \tag{19}$$

To calculate the fluid phase effective thermal conductivity, k_{af}, the correlation proposed by Edwards and Richardson [25] was used. In this case, k_{af} is a function of the position through the fluid velocity:

$$\frac{1}{Pe_{af}} = \frac{0.73\varepsilon}{Re\,Pr} + \frac{0.5}{1+\dfrac{9.7\varepsilon}{Re\,Pr}} \tag{20}$$

The fluid-solid heat transfer coefficient, h, was calculated by Stuke's correlation [26] that assumed h as a function of the Reynolds number.

An analytical solution for this coupled system of nonlinear partial differential equations may be difficult to find, so an orthogonal collocation method (with 11 collocation points) was chosen combined with a Runge-Kutta method in order to obtain the corresponding temperature profiles.

4. RESULTS AND DISCUSSION

Figure 3.a shows a typical temperature response for the non-steady case after the temperature of the air was step-increased from T_0=23°C to T_1=66°C. The top line corresponds to the gas entrance temperature, close to a perfect step temperature input. Figure 3.b shows the temperature profiles corresponding to Fig. 3.a at different values of time. Initially the bed responds slowly to the change of temperature and after three hours the temperature inside the reactor was almost uniform and constant. For this reason, it was necessary to introduce an electric heater inside the reactor to study its thermal behavior under steady-state conditions.

Figure 4 shows three radial temperature profiles for the steady state case. The radial reactor used has a R_2/R_1 ratio equal to 6, a particle diameter of 3.3 mm and a gas input flow of 8.5, 13.8 and 16.3 m^3/h. The dotted lines represent the theoretical values obtained using Eq. (4). The value of δ adjusted for all steady experiments was 5. An acceptable agreement between experimental and theoretical values is observed. This behavior was found in all the experiments performed at different operating conditions.

Figure 5 contains all the k_e values calculated in this work for the steady state. For all the steady state cases the value of δ that best fit the temperature profile is 5 (in the range of Reynolds numbers studied). This value is one order of magnitude higher than the value reported by Yagi *et al.* [12] for axial flow in tubular reactors. As it was expected the variation of k_e as a function of RePr was linear (Eq.(2)).

Figure 6 shows the Peclet number values as a function of the Reynolds number including others experimental results previously reported, indicating good agreement. It can be

observed that non-steady experimental values are similar to those calculated under steady state conditions. This behavior agrees with the result obtained by Dixon and Cresswell [19] for the axial flow reactors at small Reynolds numbers. Then the effective parameter (k_e) can be considered the same for steady state and transient models for the radial-flow packed-bed reactors at Reynolds numbers less than 5.

Fig. 7 shows the temperature response (Fig. 7.a) and the temperature profile (Fig. 7.b) for a reactor with a ratio R_2/R_1 equal to 6 for two different Reynolds numbers (Re=8 and Re=400). The dimensionless temperatures were calculated using Eq. (6) and the numerical solution of Eqs. (9 and 13). There is an acceptable concordance between the pseudo-homogeneous and the two-phase models for low Reynolds number. The pseudo-homogeneous temperature profile is located between the solutions for solid and gas phases profiles. However, for high Reynolds number, a large difference is observed between the corresponding profiles, indicating disagreement between the two models. A pseudo-homogeneous equation can be used instead of the two-phase equations for low Reynolds numbers but it is not recommended for high Reynolds.

Fig. 8 shows a case where the effective thermal conductivity can be approximated by a constant along the reactor bed for radial flow. In this case the dimensionless temperature is almost the same when δ goes from 0 to 0.75 for low modified Peclet number (around 4). Nevertheless, for a Pe* equal to 40 the difference between taking k_e constant or variable is more notorious. Then, it is possible to simplify the model to one with k_e constant at low Reynolds (Pe*) numbers and use the analytical solution found by López de Ramos and Pironti [22].

5. CONCLUSIONS

For the pseudohomogeneous model, an analytical solution was obtained using the method of separation of variables and a series approximation. In the two-phase model, the gas and solid temperature profiles were obtained by two numerical methods: orthogonal collocation and Runge-Kutta. Theoretical results were compared with experimental data in order to calculate the effective thermal conductivity. The values of k_e agree in general with the literature data.

At low Reynolds numbers there is no appreciable difference between a pseudohomogeneous model and a two phase equation model. Constant thermal properties can be used at Re<5 with enough accuracy to predict the thermal behavior of a radial-flow reactor. Furthermore, there was no difference between steady state and transient methods for experimental determination of the effective thermal conductivity at low Reynolds numbers. At high Reynolds numbers it is recommendable to use a two-phase model with a variable fluid-phase effective thermal conductivity.

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FIGURE CAPTIONS

Fig. 1: Experimental set-up [1,3: Gate valves; 2,5, 6, 8, 9, 10: Ball valves; 4: Rotameter; 7: Electrical heater system; 11: Reactor vessel; 12, 13: Drains; 14: Data acquisition system; 15: Personal computer].

Fig. 2: Radial-flow packed-bed reactor.

Fig. 3. (a) Temperature response and (b) radial temperature profile for the non-steady case after a temperature air step from T_0=23°c to T_1=66°C. (Relation R_2/R_1 = 4, Particle diameter = 4.2 mm, Gas input flow = 19.51 m^3/h).

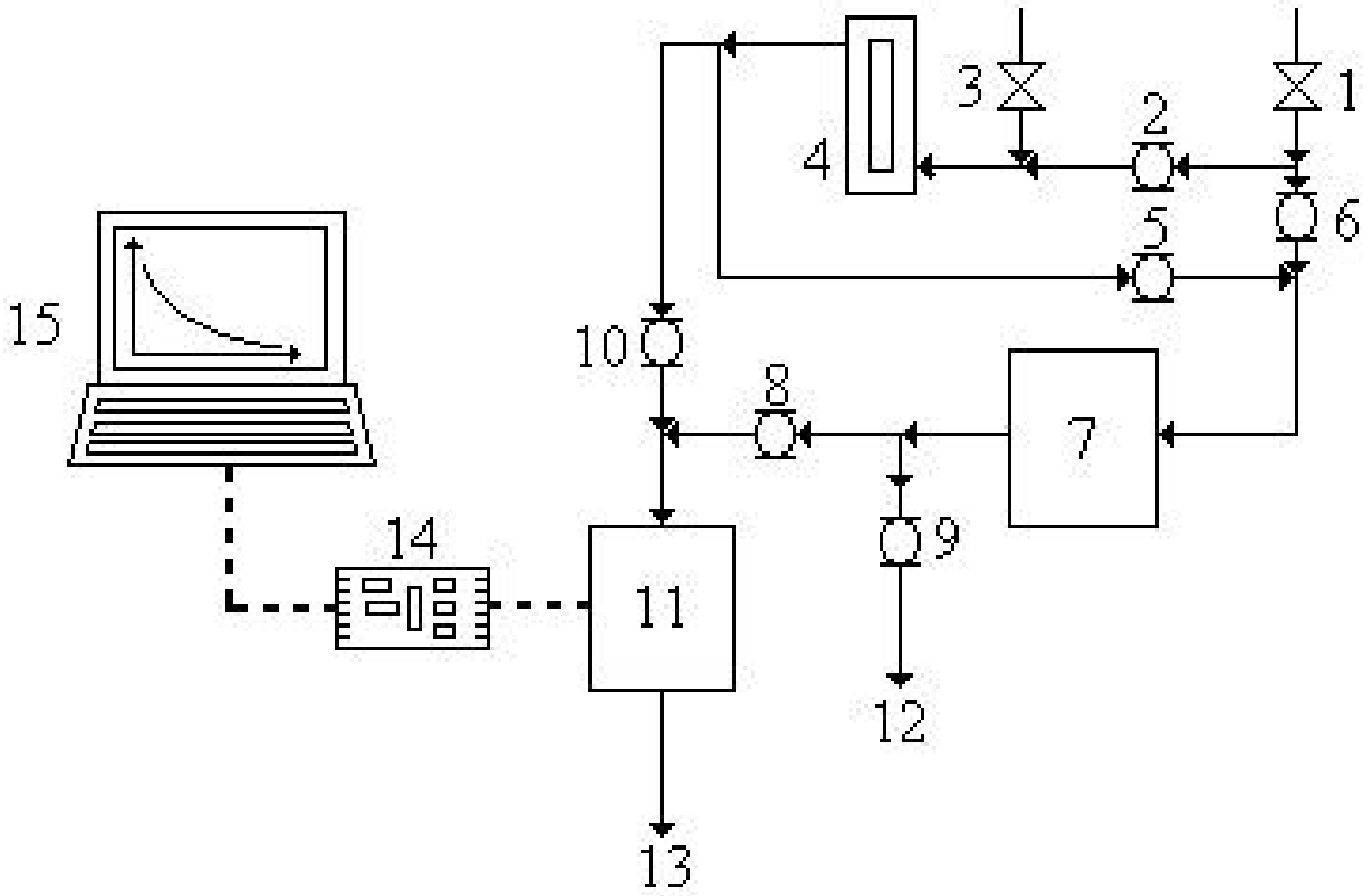
Fig. 4. Radial temperature profile for the steady state case (Relation R_2/R_1 = 6, Particle diameter = 3.3 mm, Gas input flow = 8.5, 13.8 and 16.3 m^3/h).

Fig. 5. k_e as a function of RePr using the steady state model.

Fig. 6. Pe as a function of Re for the steady state and transient models.

Fig. 7. (a) Temperature response and (b) temperature profile for Re = 50, R_2/R_1 = 6, and particle diameter = 4.2 mm for a non-steady method.

Fig. 8. Influence of δ values in temperature responses.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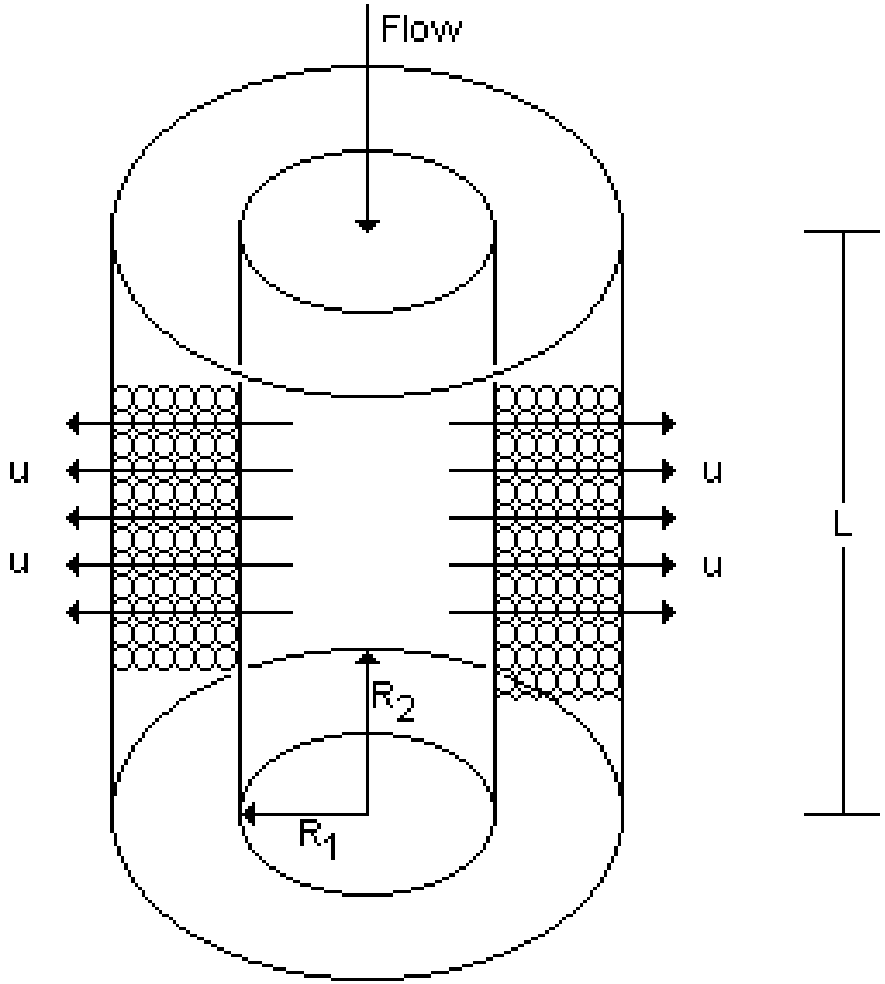
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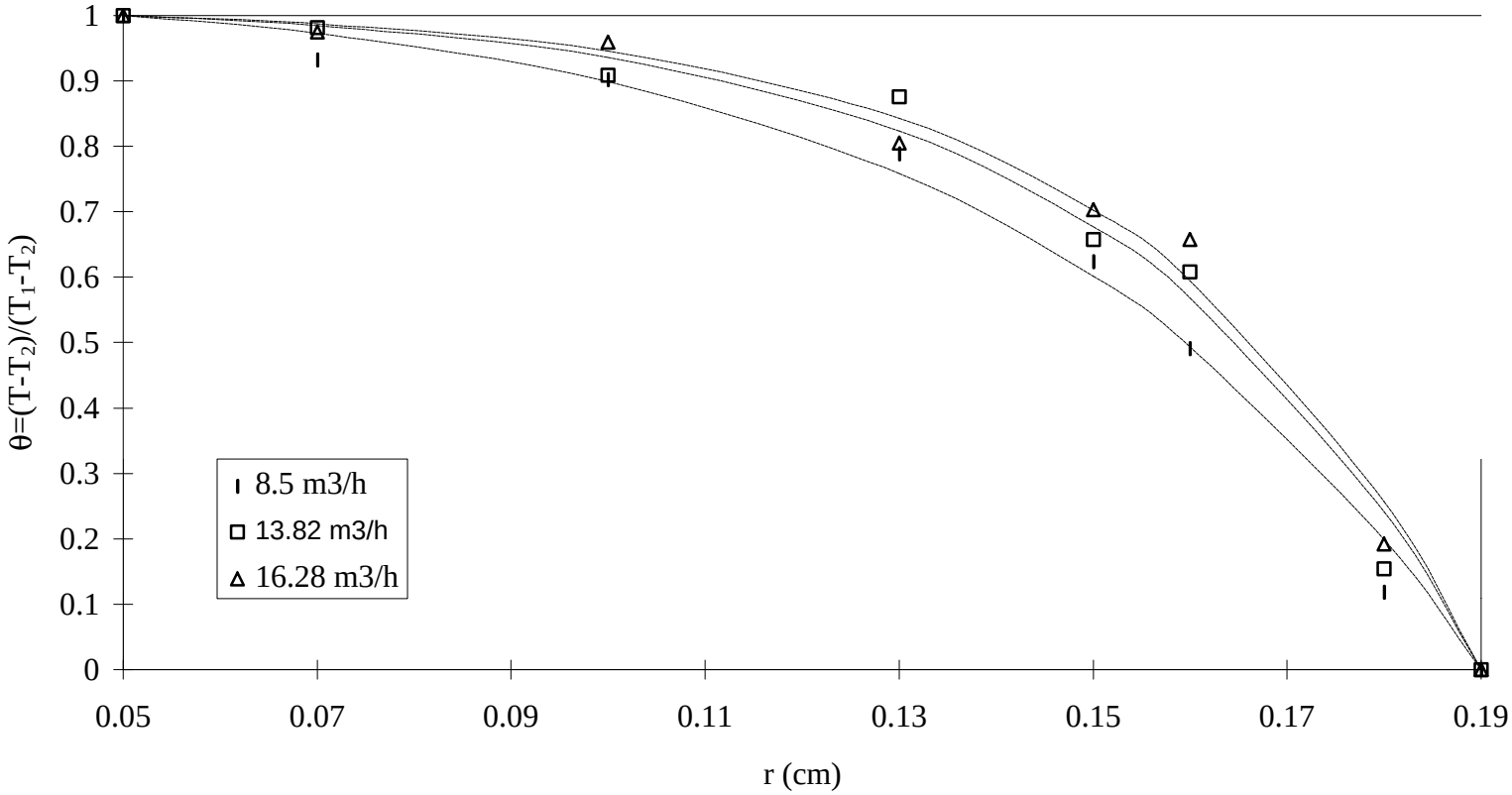
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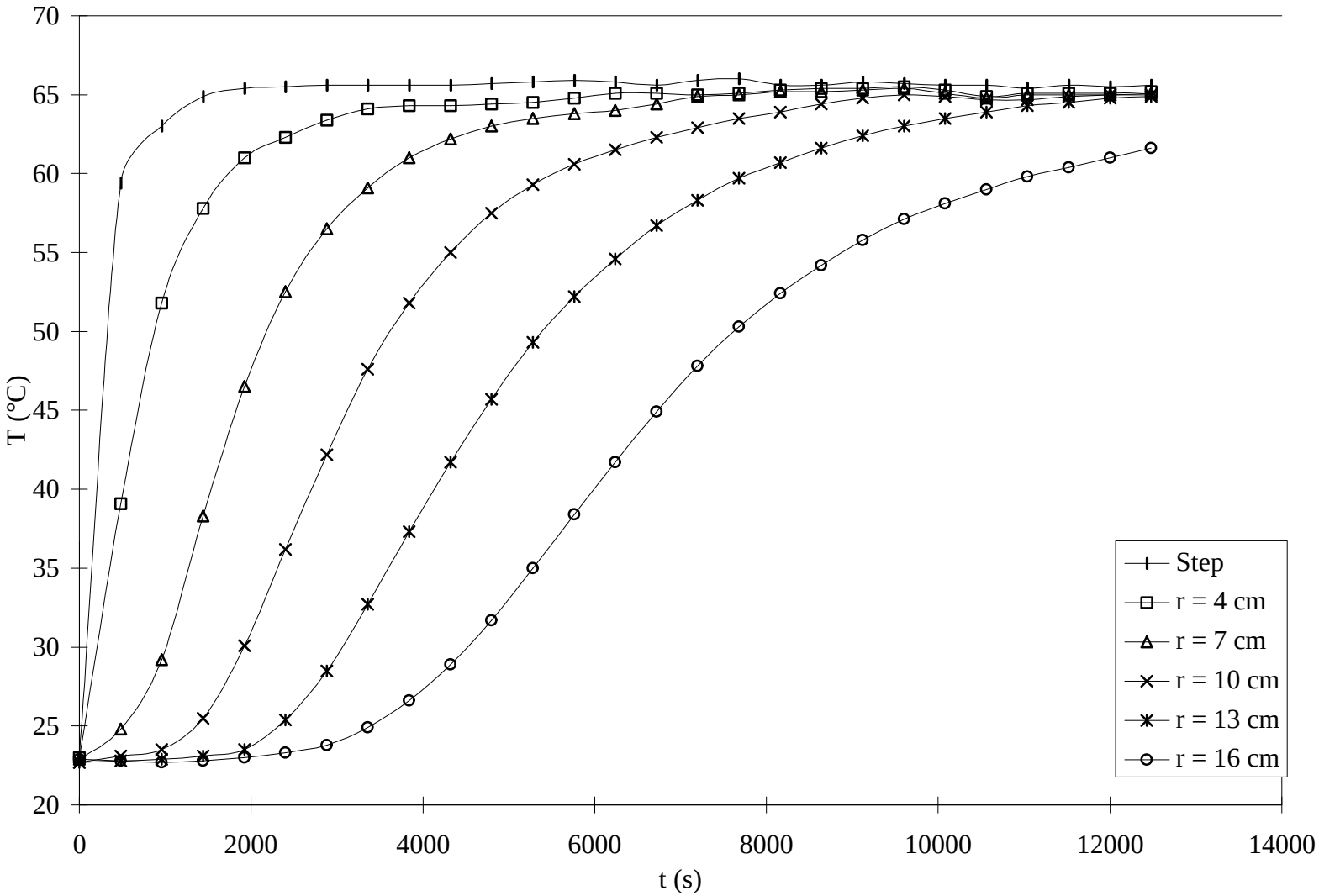
0.7
0.6
0.5
0.4
0.3
0.2
0.1
0
θ=(T-T2)/(T1-T2)
0.05
0.07
0.09
0.11
0.13
0.15
0.17
0.19
r (cm)
8.5 m3/h
13.82 m3/h
16.28 m3/h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60
55
50
45
40
35
30
25
20
T (°C)
0
2000
4000
6000
8000
10000
12000
14000
t (s)
Step
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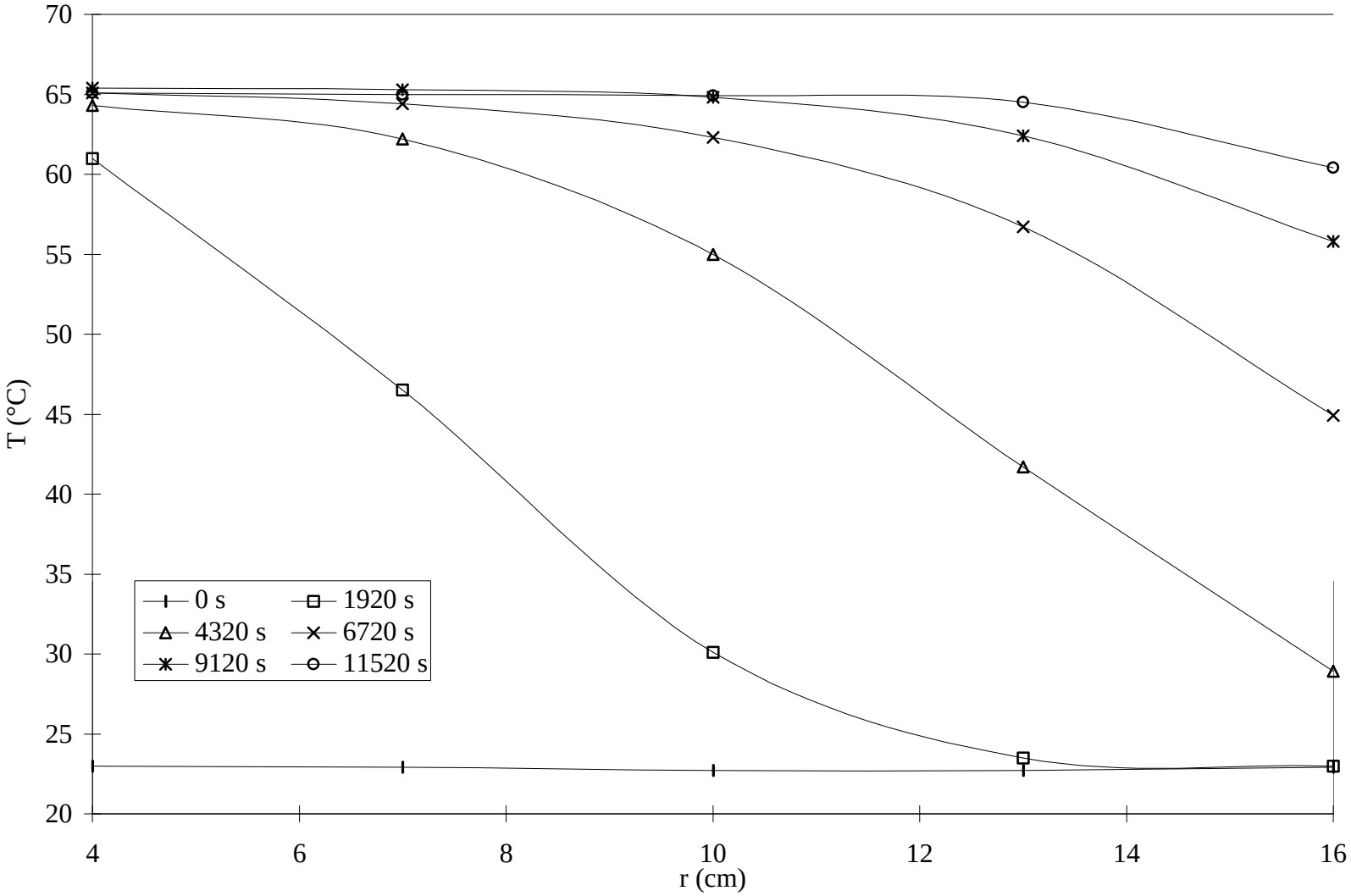
r = 7 cm
r = 10 cm
r = 13 cm
r = 16 cm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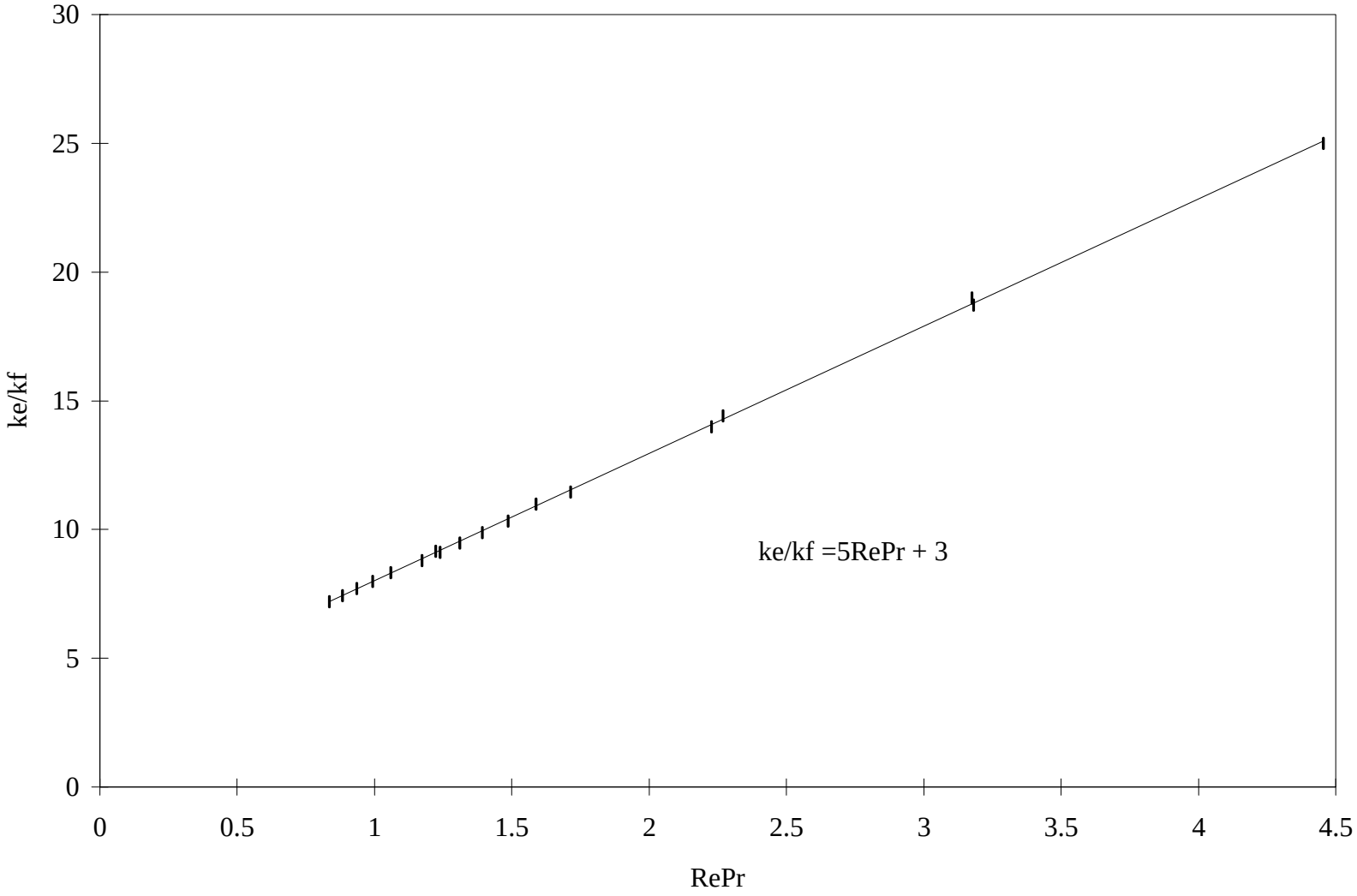
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10
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ke/kf
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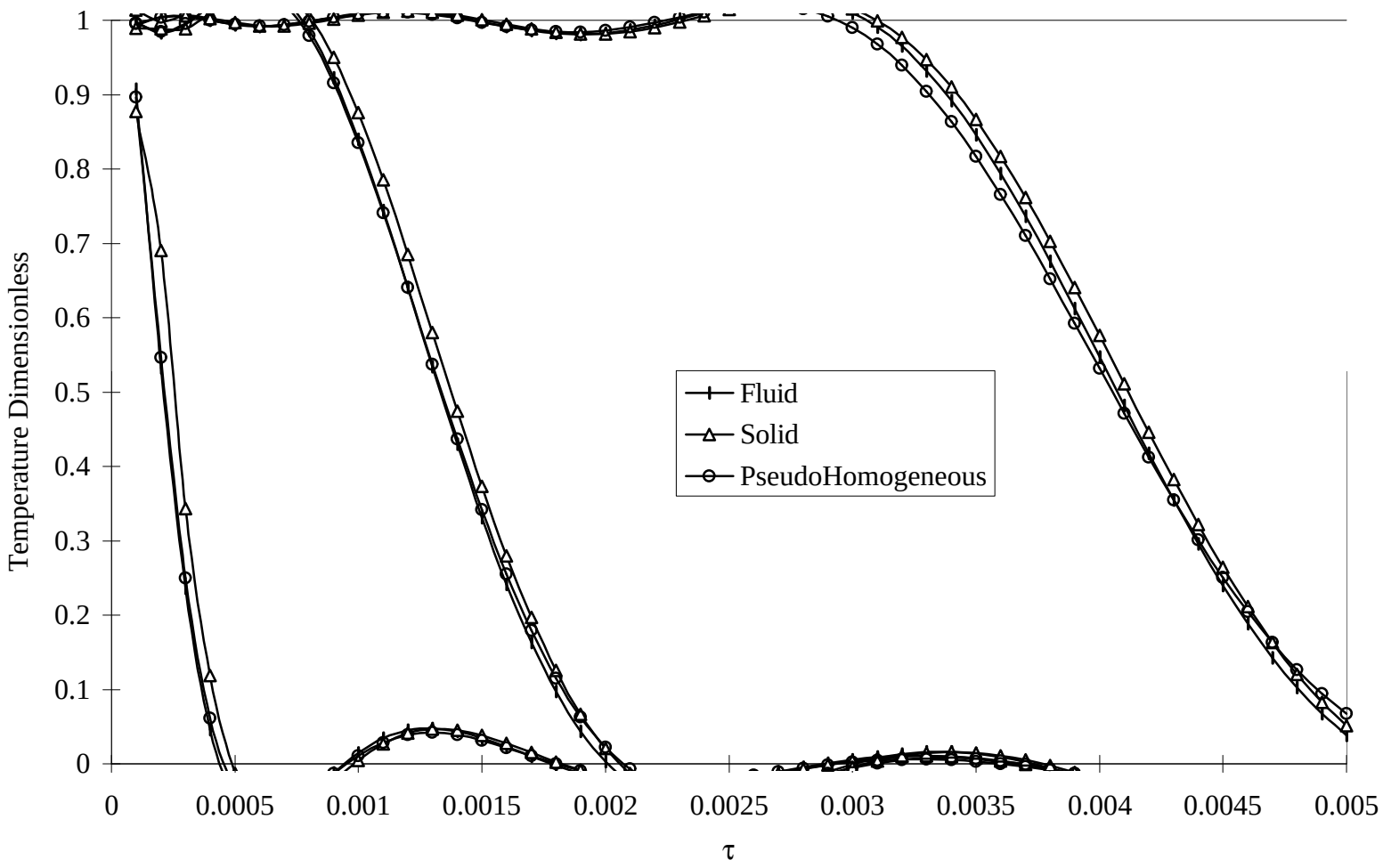
3
3.5
4
4.5
RePr
ke/kf =5RePr + 3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0.0005
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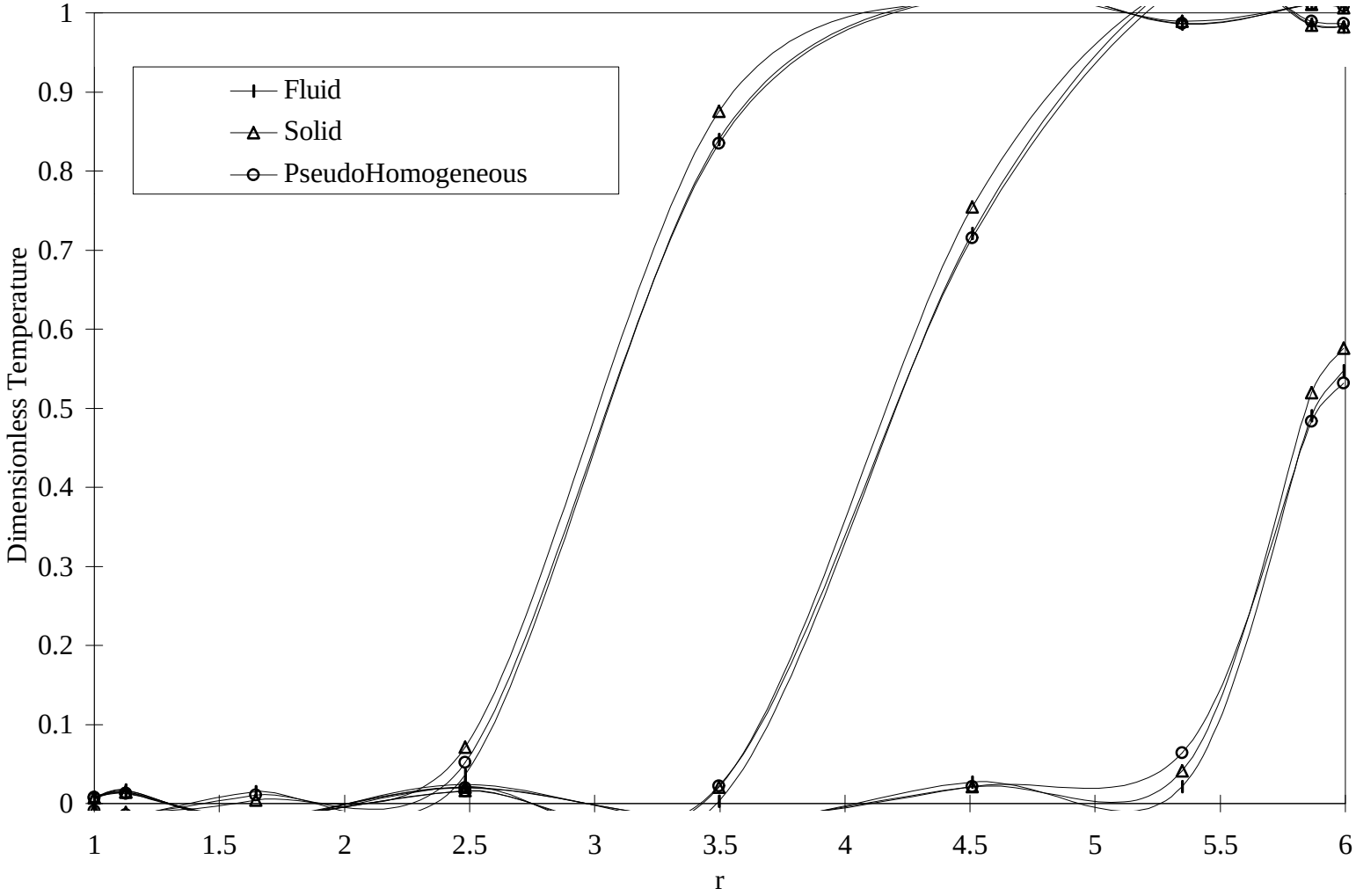
τ
Fluid
Solid
PseudoHomogeneous


Fluid
Solid
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Dimensionless Temperature
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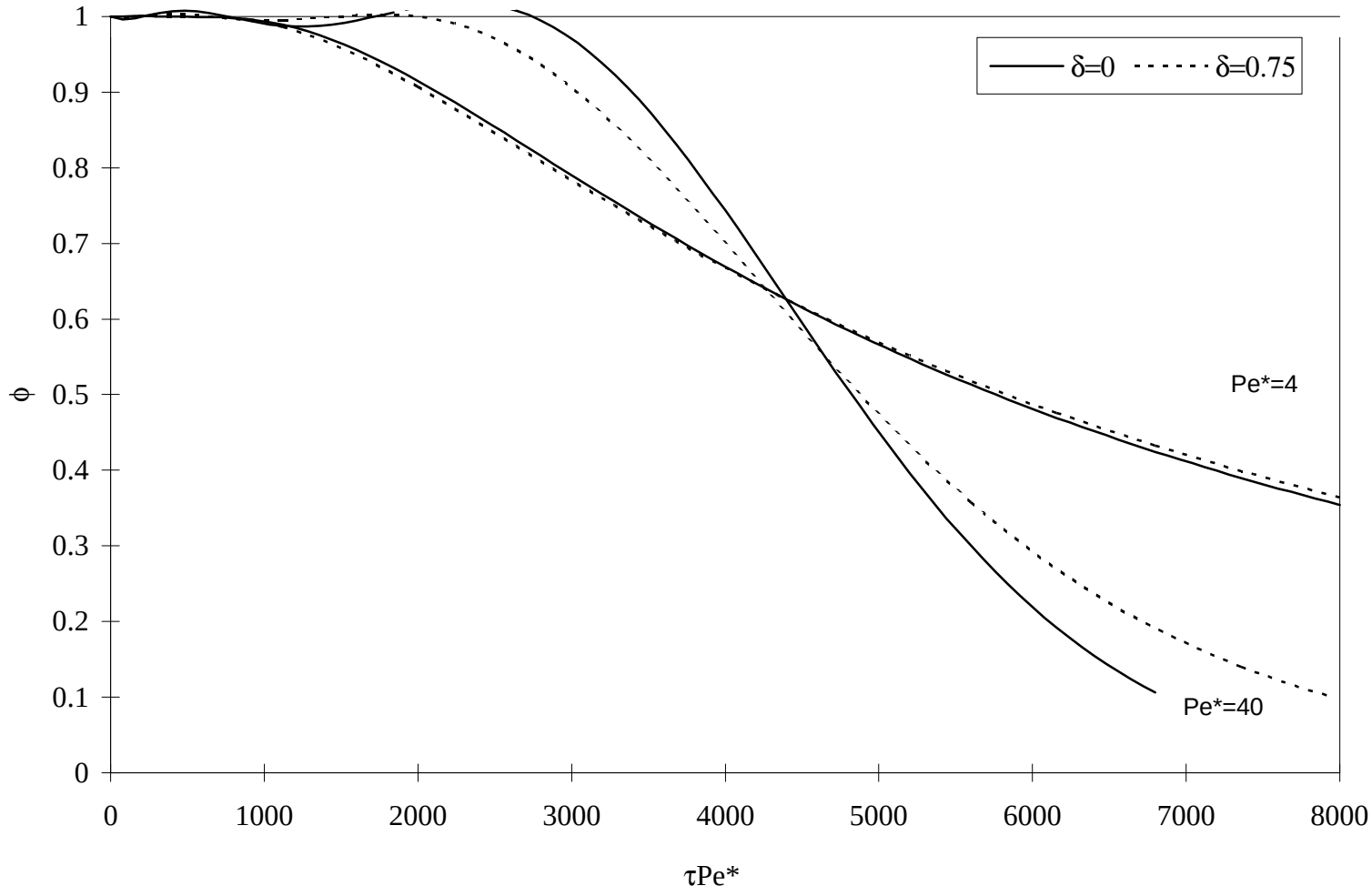
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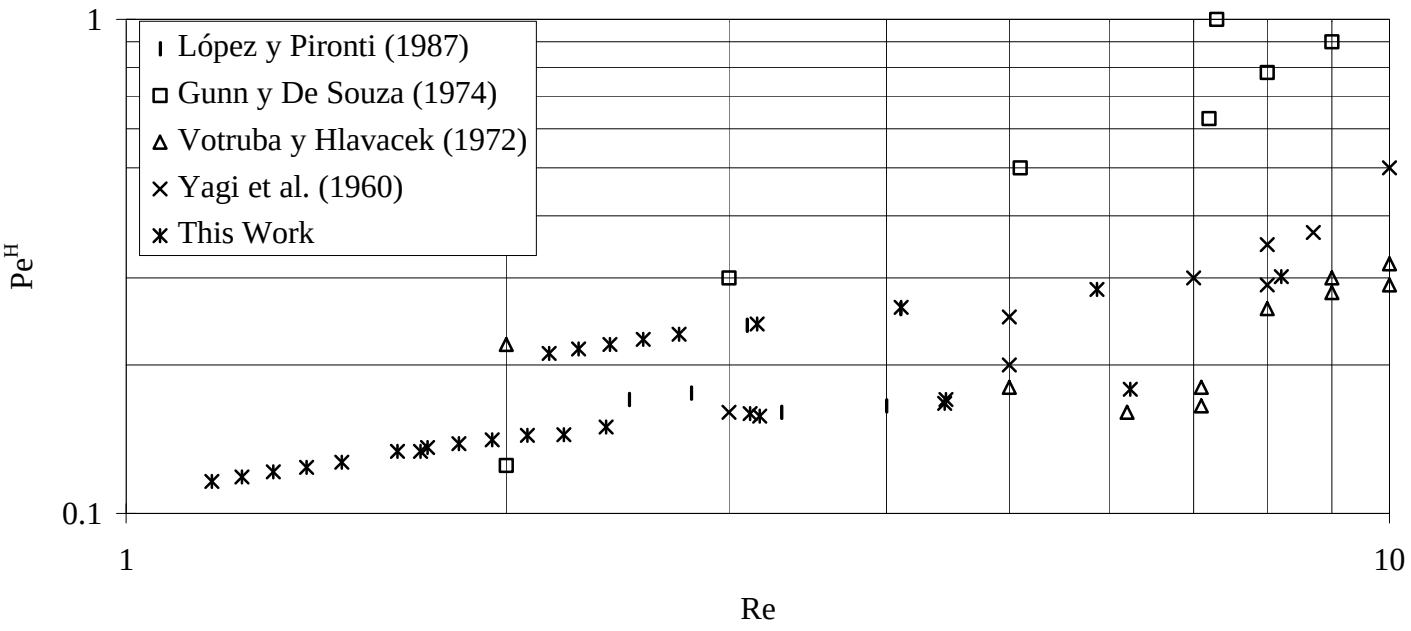
τPe*
0
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.8
0.9
1
0
1000
2000
3000
4000
5000
6000
7000
8000


López y Pironti (1987)
Gunn y De Souza (1974)
Votruba y Hlavacek (1972)
Yagi et al. (1960)
This Work
1
0.1
Pe^H
1
10
Re