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RADIAL HEAT TRANSPORT IN PACKED BEDS-II: MATHEMATICAL MODELING OF HEAT TRANSFER THROUGH PACKED BEDS AT ELEVATED PRESSURE

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ABSTRACT

Packed bed reactors are often used in the applications in the process which need for the removal or supplying of heat. Modeling of fluid flow, heat transfer and reaction in packed beds is an essential part of their designs, there is especially for endothermic or exothermic reactions that occur at high pressure.

Heat transfer in packed beds with fluid flow is important, whereas in these systems both reactor selectivity and velocity of reactions depend largely on bed temperature. Knowledge of temperature profile within the bed and the heat removal rate or which has been added is essential to achieve reactor productivity and product homogeneity. A mathematical model is developed to investigate the adequacy of the conventional equation used to describe the parameters which influences the temperature profile and wall heat transfer of a packed bed heat exchanger under high pressure up to 20 bars. The model equation provides comprehensive relation for predicting the wall heat transfer coefficient through the boundary layer that appears in the voidage between the wall and the random packing of solid particles in the bed and the effective radial thermal conductivity through the bed at elevated pressure. Predicted model equation have been compared with the experimental data of the temperature profile at different tube-particle diameter ratios for cylindrical pellets, inside the reactor tubes in which the only flow is air at high pressure.

Agreements between the theoretical model and experimental data results at high pressure are acceptable for estimating temperature profile through a fixed bed reactor, with different catalyst pellets and shapes. The highest average difference between the experimental and theoretical value was found approximately 11%.

Keywords: heat transfer, packed beds, pseudo-homogeneous model, boundary conditions

1. Introduction

Packed bed reactors are widely used in the industrial processing to carry out heterogeneous chemical reactions (endo- or exothermic reactions), and is usually accomplished by means of a multi-tubular reactor in which long narrow tubes are immersed in a heat exchange medium that facilitating radial heat loss to the tube wall. The study of radial heat transfer mechanisms through the packing and wall of fluid flowing through a single tube forms an important aspect to design of the packed bed reactors.

Many different models for wall-packed bed reactors have been developed over the past years, ranging from a simple one-dimensional homogeneous plug-flow model to more complex ones, such as the two-dimensional heterogeneous and axially dispersed plug-flow model. The parameters for heat and mass transport in these models lump different physical transport mechanisms of heat and mass, occurring at different scales, into simple overall coefficients. In the models, the driving forces for transport of heat and mass are averaged values which may actually vary strongly due to the heterogeneity of the packed bed. Many empirical correlations which have differ widely for the transport coefficients have been proposed in the literatures. These differences are probably attributed to the sensitivity of the model parameters to experimental errors, the use of different

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transport model concepts and to a poor consideration of the influence of the catalyst particles geometry and the packing. A difference between the values of the effective transport parameters obtained at reacting and non-reacting conditions has been reported in literature. There are several studies have been presented using two dimensional pseudo-homogeneous model and covering experimental statistical and theoretical aspects with quite conclusive results (e.g. Wellauer et al., 1982; Gun et al., 1987; Tsotsas and Schlünder, 1990; Freiwald and Paterson, 1992; Jorge et al., 1999; Thomeo and Freire, 2000). These studies usually contain effective heat transfer coefficients (K_{er} and h_w), which are preferably obtained from experimental without a chemical reaction. However, the adequate transfer coefficients values are greatly depending on the used scale of test equipments and the proper model assumptions that are used to evaluate the experimental data (e.g. the selection of boundary conditions will have a large impact on the accuracy of the estimated heat transport coefficient values. The two dimensional pseudo-homogeneous model for heat transfer in a packed bed used by the previous literature is:

$$GCp\frac{\partial T}{\partial Z} = K_{er}\left(\frac{\partial^2 T}{\partial r^2} + \frac{1}{r}\left(\frac{\partial T}{\partial r}\right)\right) \tag{1}$$

within boundary conditions;

$$z = 0,\ \text{all } r,\ T = T_o \tag{2.a}$$

$$r = 0,\ \text{all } z,\ \frac{\partial T}{\partial r} = 0 \tag{2.b}$$

$$r = R_t,\ \text{all } z,\ -K_{er}\frac{\partial T}{\partial r} = h_w\left(T_w - T\right) \tag{2.c}$$

In dimensionless form (Eqn.1) becomes:

$$\frac{\partial T'}{\partial \zeta} = \alpha'\frac{1}{r'}\frac{\partial}{\partial r'}\left(r'\frac{\partial T'}{\partial r'}\right) \tag{3}$$

So, boundary conditions given (Eqns. 2.a~c) respectively become:

$$\zeta = 0,\ \text{all } r',\ T' = 1 \tag{4.a}$$

$$r' = 0,\ \text{all } \zeta,\ \frac{\partial T'}{\partial r'} = 0 \tag{4.b}$$

$$r' = 1,\ \text{all } \zeta,\ -\frac{\partial T'}{\partial r'} = \beta_i T' \tag{4.c}$$

A world-wide well known solution for the (Eqn.3) is given by (Marshall & Coberly, 1951), when the effective thermal conductivity and wall heat transfer coefficient are taken as constants.

$$T'(r',\zeta) = 2\sum_{nj=1}^{\infty}\frac{J_o\left(\lambda_{nj}r'\right)*\exp\left(-\alpha'\lambda_{nj}^2\zeta\right)}{\lambda_{nj}J_1\left(\lambda_{nj}\right)*\left[\left(\lambda_{nj}/\beta_i\right)^2+1\right]} \tag{5}$$

Whereas;

$J_o\left(\lambda_{nj}r'\right)$ is the zeroth order Bessel function of the first kind, and the λ_{nj} are the roots to;

$$\lambda_{n_j}J_1(\lambda_{n_j}) = \beta_i J_o(\lambda_{n_j}) \tag{6}$$

Ziolkowski (1970) and, Thomeo and Grace (2004) show that the methods described above have some limitations, due to they were based on several simplifying assumptions (Eqn.5). These assumptions are:

First, the boundary condition (Eqn. 4.a) taken as the gas temperature $T' = 1$, and is uniform in initial cross-section of the bed, which is generally not valid, where a temperature gradient always exists along the radius (The temperature is larger near the wall).

Second, the limit of these methods consists in that the wall temperature is required to be uniform along the tube. This makes the experiments limited to rather low wall temperature.

The measurements of the radial temperature profiles above the bed for its different heights and the graphical differentiation of the radial temperature profiles, required for (Eqn.5) are the reason for further uncertainty in the estimation of (K_{er}), and (h_w) values and make the method rather time-consuming. Other limitation that, all methods are taken at atmospheric pressure, so the heat transfer phenomena of a packed bed are lumped into only a few effective parameters in such models, making it very difficult to extrapolate empirical correlations from laboratory conditions up to plant-size equipment.

Equation of Marshall and Coberly with their assumption are not good enough, due to it is depending on inlet temperature into packed bed is constant, but in reality of experimental investigations proved that, they are not constant value, but it is function of the radial distance (Thomeo and Grace, 2004; Al-Meshragi et al., 2009). This is what we have referred to in this study and depending on it greatly in formulation of a modified equation.

In this manuscript the influence is studied of the choice of boundary conditions, depending on the values obtained for the heat transport coefficients, for two-dimensional pseudo homogeneous model in packed bed at high pressure.

Generally, the main objective is investigating and obtaining the accuracy of a modified mathematical model to determine the temperature profiles within nonisothermal fixed bed gas reactor at high pressure. And as a consequence accurately estimating the characteristic parameters of heat transport namely effective radial thermal conductivity K_{er} and wall heat transfer coefficient h_w.

2. Modeling and Simulation

The main heat transport processes in a fixed bed may be represented by pseudo-homogeneous model, depending on radial temperature profile, where the two dimensional, two parameters continuum model for heat transfer in a cylindrical packed bed operated as steady state heat exchanger was used to evaluate the effective thermal conductivity and wall heat transfer coefficient, then the theoretical radial temperature profiles were evaluated.

General assumptions of the model equation of the heat transport can be summarized as the following;

- Steady state conditions and no chemical reactions were carried out.
- Uniform velocity of the fluid through tube diameter.
- Negligible temperature difference between solid and fluid.
- Effective thermal conductivity is uniform throughout the packed bed.
- There is no heat axial dispersion.
- No radiation occurred.
- There is no pressure drop throughout the packed bed.
- Physical properties of the gas and solid are independent on temperature.

The main assumption considered in the model equation, is that the bed inlet temperature is a function of radial distance at z = 0. There is no temperature profile available in the experimental data (z=0). The inlet temperature to the bed is taken as the first measured radial temperature profile and in addition the location as the bed entrance point (z_o). It was clear from the experimental data that the temperature profile at this point is a polynomial equation (Al-Meshragi et al., 2009; Ibrahim, 2010).

To solve (Eqn.3) the following modification will be made:

$$\theta = \frac{T'}{T'_c} = \frac{T_{w-}T}{T_w - T_c} \tag{7}$$

$$\delta = \frac{\zeta - \zeta_o}{1 - \zeta_o} = \frac{z - z_o}{L - z_o} \tag{8}$$

By modification (Eqn.3) becomes;

$$\frac{\partial \theta}{\partial \delta} = \frac{\alpha'}{r'} \frac{\partial}{\partial r'} \left(r' \frac{\partial \theta}{\partial r'} \right) \tag{9}$$

with modified boundary conditions can writing as:

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$$\theta(r') = a + b(r') + c(r')^2 + d(r')^3, \quad \text{at} \quad \delta = 0 \quad \text{for all} \quad r' \quad \text{values} \tag{10.a}$$

$$\frac{\partial\theta}{\partial\delta} = 0, \quad \text{at} \quad r' = 0 \quad \text{for all} \quad \delta \quad \text{values.} \tag{10.b}$$

$$\frac{\partial\theta}{\partial\delta} = -\beta_i{*}\theta, \text{ at} \quad r' = 1 \quad \text{for all} \quad \delta \quad \text{values.} \tag{10.c}$$

where:

a, b, c and *d* are constants obtained by fitting (Eqn.10.a) to the experimentally determined radial temperature profile at position (z=zo $\Rightarrow \delta = 0$). Eqn. (9) is solved by using the modified boundary conditions in (Eqn.10) to yield the final equation (more details, see Ibrahim, 2010);

$$\theta(r',\delta) = 2\sum_{n_j=1}^{\infty}\left[\frac{\left(A\lambda_{n_j}^4 - B\lambda_{n_j}^2 + C\frac{\lambda_{n_j}^4}{\beta_i} + (9d - b\lambda_{n_j}^2)\frac{0.91973}{J_1(\lambda_{n_j})}\right)J_o(\lambda_{n_j}r')}{\lambda_{n_j}^5 J_1(\lambda_{n_j})\left[(\lambda_{n_j}/\beta_i)^2 + 1\right]}exp(-\alpha'\lambda_{n_j}^2\delta)\right] \tag{11}$$

where:

$A = (a+b+c+d)$

$B = (4c+9d)$

$C = (b+2c+3d)$

λ_{nj} is found from (Eqn.6).

With (Eqns.5 and 11), it is possible to calculate temperature profiles for wall heating packed bed, with uniform or a function of inlet temperature profile respectively.

Values for the effective radial thermal conductivity (K_{er}) and wall heat transfer coefficient (h_w), for the two models, were obtained by fitting (Eqns.6 and 11) with the experimental data. The objective function minimized was a chi-square target function given according to;

$$\chi^2 = \sum_{i=1}^{N}\left(\frac{T_{\exp.} - T_{calc}(r,z,K_{er},h_w)}{\sigma_i}\right)^2 \tag{12}$$

For (Eqn.12) N is the number of measured temperatures, $T_{exp.}$ is the experimentally obtained temperature, $T_{calc}(r, z, K_{er}, h_w)$ is the calculated temperature by the model, and σ_i, is the standard deviation for the experimental data of temperature. The chi-square target- function was minimized, using Nealder-Mead method in multi-dimensions with different starting points, Press et al., (1989). Also values for the goodness-of-fit GOF, and the mean error were estimated. The GOF is the probability that the chi-square exceeds a particular value $\chi^2_{\min}$ by chance. It is worth to note that at a very small probability for some particular data set, the apparent discrepancies are unlikely to be coincidental fluctuations. Where in this case neither the model is wrong nor the measurement errors are really larger than stated, Press et al., (1989).

The mean error is calculated from:

$$Error = \frac{\sum_{i=1}^{N}\left|(T_{\exp.} - T_{calc.})\right|}{\sum_{i=1}^{N}T_{\exp.}} * 100\% \tag{13}$$

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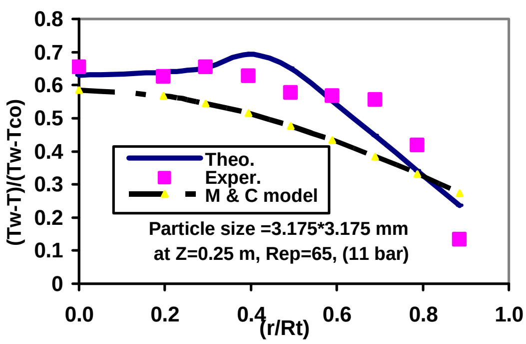
Values for the effective radial heat conductivity (K_{er}) and the wall heat transfer coefficient (h_w), can be obtained for different bed lengths using the determined radial temperature profiles. In the used experimental set-up these values are obtained throughout averaging the radial temperature profiles measured in the packed bed after repacking the bed several times; in order to minimize the effect of deviations from the intended radial positions and of irregularities in the bed surface. The calculated radial temperatures are compared to the available experimental temperature values and adjusted until the difference between two successive χ_i^2 is smaller than 0.001 (more details about experimental work see Al-Meshragi et al., 2009).

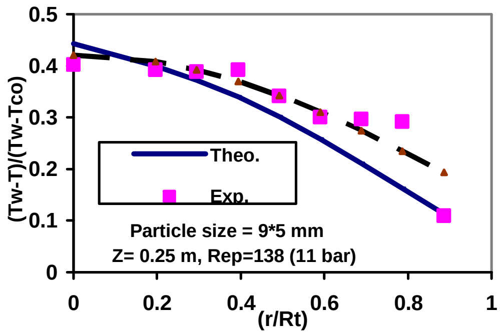
3. Results and Discussion

The modified mathematical model for prediction of heat transfer at high pressure in a fixed bed reactor in a radial direction has been derived. The values predicted from the present model equation have been compared with the experimental data (Al-Meshragi et al., 2009) and also with the values calculated using Marshall and Coberly equation (M & C model) for a given conditions. The present modified model is comprehensive equation used at different ranges of Reynolds number, high pressure and different catalyst shapes.

Figures (1-a~c) show the comparison of the radial temperature profile between the present modified model, M & C model and experimental data at 11 bars for various particle pellet sizes. A good agreement between the modified model equation and the experimental data were found, while less agreement was found by M & C model. The mean total error calculated between the modified model equation values and the experimental data show that it is not exceeds 3.3%, while the mean total error obtained from M & C model reached up to 8% at the same conditions. These indicate that the present modified model can well fitting the experimental data with great accuracy.



(a)



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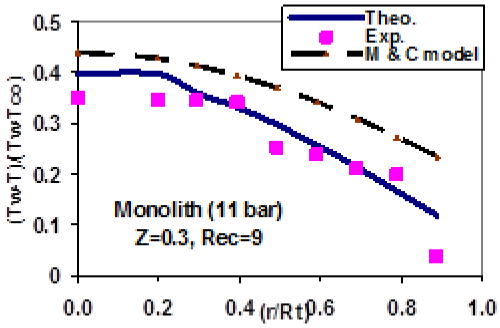
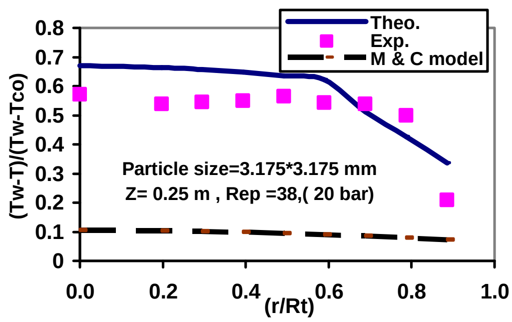


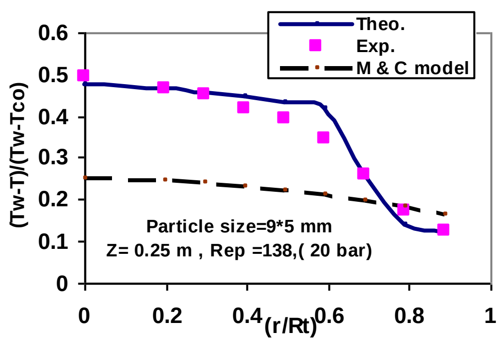
(c)

Figure 1. Comparison of the radial temperature profile of present model with M & C model and experimental data

Also, Figures (2-a~c) indicate that higher accuracy can be achieved using the modified model equation than M & C model at 20 bars. The mean total error reached up to 11% at these conditions. However, the mean total error achieved by M & C model reached up to 25 %. These results show that the present model equation provides a higher accuracy at both pressures than M & C model.



(a)



(b)

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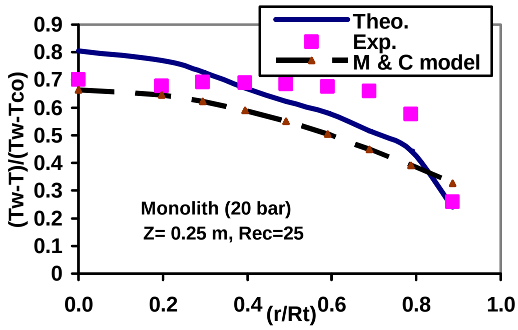

(c)

Figure 2. Comparison of the radial temperature profile of present model with M & C model and experimental data

The wall heat transfer coefficients (h_w) calculated by modified mathematical model were plotted against particle Reynolds number (Re_p) (Figures 3 and 4) at elevated pressure. It was clearly show that the wall heat transfer coefficient increased by increase of particle Reynolds number at the same pressure. Also, at the same Reynolds number the wall heat transfer decreased by increasing pressures. This is most likely attributed to the variation of gas properties with respect to the elevated pressure. While Figure (5) shows, the variation of the wall heat transfer coefficient (h_w) with channel Reynolds number (Re_c) of the monolith structure. The same trend was clearly observed as in (Figures.3 and 4) in spit of the particle size and type.

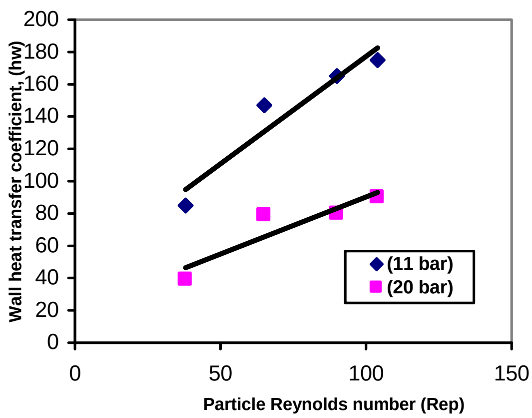

Figure 3. Wall heat transfer coefficient, for the tube with d_t= 50.88 mm and filled with cylindrical pellets of (3.175*3.175) mm.

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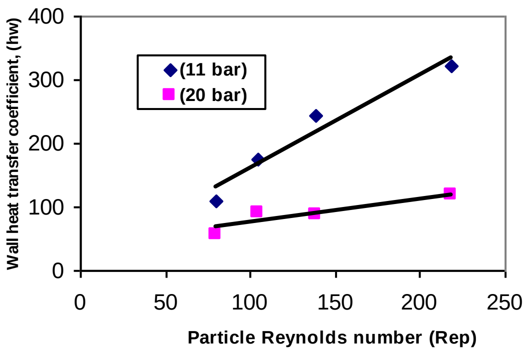

Figure 4. Wall heat transfer coefficient, for the tube with d_t= 50.88 mm and filled with cylindrical pellets of (9.0*5.0) mm.

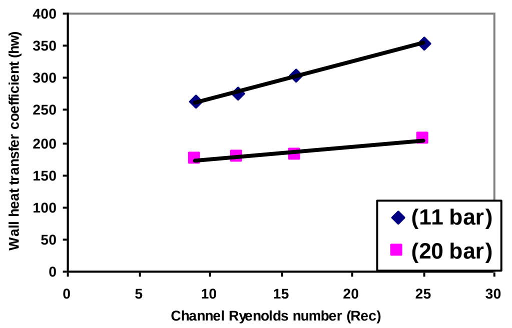

Figure 5. Wall heat transfer coefficient, for the tube with d_t= 50.88 mm and filled with Monolith catalyst.

From (Figures.3 and 4), it can be noted that the influence of the tube-particle diameter (d_t/d_p) ratio upon the wall heat transfer coefficient, at which the increase of (d_t/d_p) ratio will increase the contact area of the pellets at the wall that gives greater area of contact for heat transfer and conduction effect will be more the convection, or by other words that leads to decrease particle Reynolds number followed by decreasing in wall heat transfer coefficient (h_w).

The same trending are values of the effective radial thermal conductivities that calculated by the modified mathematical model for the different particle sizes are shown in Figures (6, 7). It was clearly that the effective radial thermal conductivity increased by the increase of particle Reynolds number at the same pressure.

Also, we can notice that the effective radial thermal conductivity increased by increasing the pressure at the same Reynolds number. Figure (8) shows also, the variation of the effective radial thermal conductivity (K_{er}) with channel Reynolds number (Re_c) of the monolith structure.

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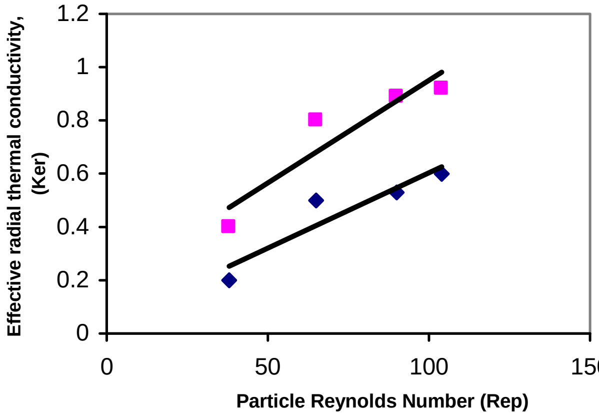

Figure 6. Effective radial thermal conductivity, for the tube with d_t= 50.88 mm and filled with cylindrical pellets of (3.175*3.175) mm.

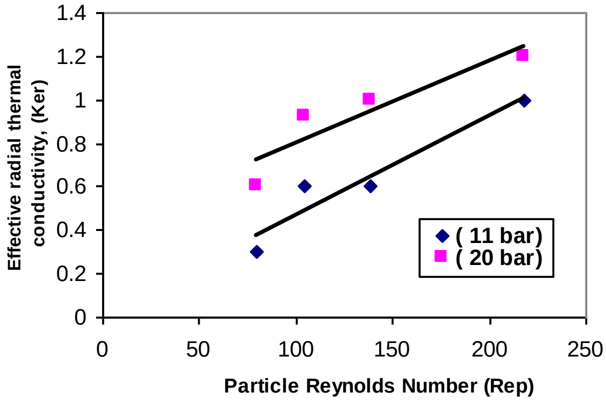

Figure 7. Effective radial thermal conductivity, for the tube with d_t= 50.88 mm and filled with cylindrical pellets of (9.0*5.0) mm.

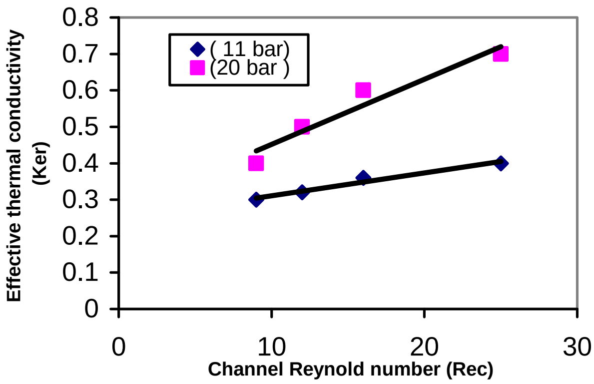

Figure 8. Effective radial thermal conductivity, for the tube with d_t= 50.88 mm and filled with Monolith Catalyst.

Figures.9~11, show the calculated radial and axial temperature profiles in fluid flowing through the packed bed which was heated from the wall, these calculations based on the modified mathematical

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model. The temperature observed theoretically at along bed depths and flow rates at high pressure. From the radial temperature profiles show in previous Figures, it was observed that them is a steep drop in the temperature in the region between the wall and a point situated away from the wall. This steep portion was more pronounced and clearly observed at the low bed heights, this is most likely due to the influence of the geometry of the pellets near the wall and to the presence of high difference in temperature between the bulk of bed and the wall. This feature probably corresponds to non-uniformity in the velocity profiles due to the packing.

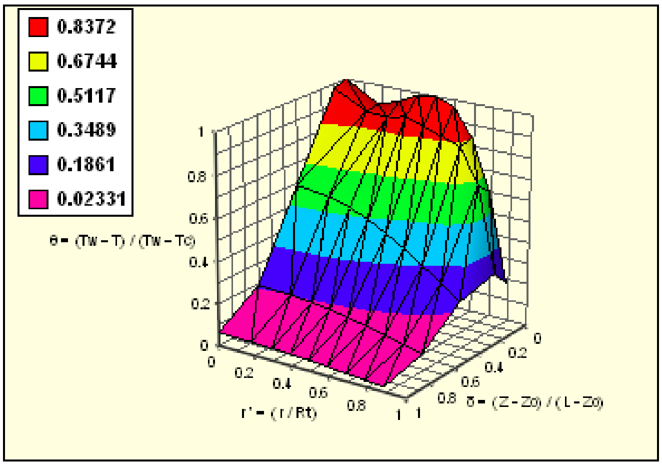

Figure 9. Calculated temperature profiles by modified mathematical model for cylindrical particle size (3.175*3.175) mm at (11 bar), Re_p=38.

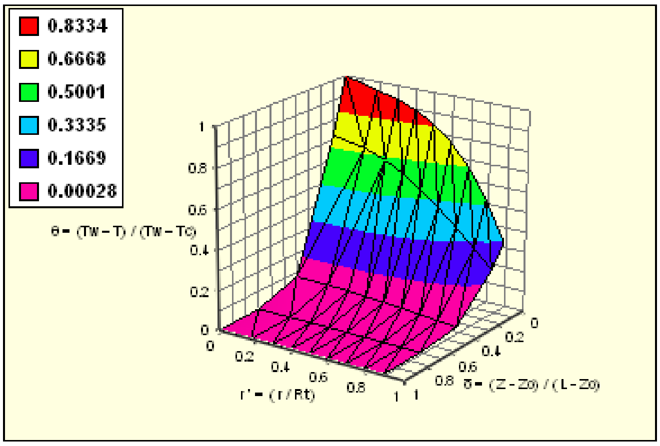

Figure 10. Calculated temperature profiles by modified mathematical model for cylindrical particle size (9.0*5.0) mm at (11 bar), Re_p=138.

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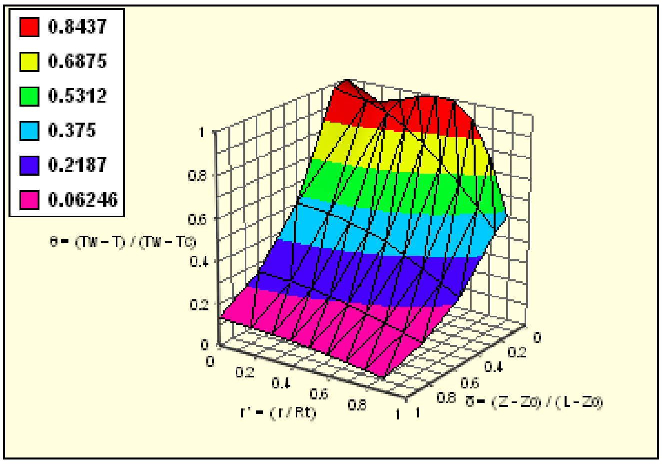

Figure 11. Calculated temperature profiles by modified mathematical model for Monolith catalyst at (11 bar), Re_C=25.

Thomeo and Friere (2000) found that the steep drop in the temperature increases near the bed wall due to the non-homogeneity of the porous medium, indicating the control of heat transfer by the boundary layer. However, this is attributed to the increase in the heat transport resistance near the wall caused by the decrease in the radial mixing due to the higher porosity and non-slip conditions at the wall.

4. Conclusion

The fit values for the effective radial heat conductivity (K_{er}) and the wall heat transfer coefficient (h_w) obtained with a polynomial inlet-temperature profile for the system were more accurate than that obtained with a flat inlet temperature. This is probably due to the fact simulate of the inlet boundary condition to the actual measured inlet temperatures at the packed bed.

In this model, the suggested assumption was clearly show good agreement between the predicted values and the experimental ones of the temperature distribution. Hence according to this finding we hope that our modified mathematical model is useful for designing correlations of heat exchanger systems.

Notation

A_p	External surface area of a pellet.	(m^2)
A,b,c and *d*	Constants of (Eqn .11) determined from the first experimentally temperature profiles.	(---)
Cp	Specific heat at constant pressure of the gas.	($J\ kg^{-1}\ K^{-1}$)
d_t	Tube diameter.	(m)
d_p	Equivalent diameter of particle, ($6V_P/A_p$).	(m)
G	Fluid mass velocity per unit area.	($kg\ m^{-2}\ sec^{-1}$)
h_w	Wall heat transfer coefficient.	($W\ m^{-2}\ K^{-1}$)
J_o ,J_1	Zeroth-order and first order Bessel functions of the first kind.	(--)
K_{er}	Effective radial thermal conductivity.	($W\ m^{-1}\ K^{-1}$)

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L	Length of the bed.	(m)
P	Pressure through packed bed.	(bar)
P_o	Atmospheric pressure.	(bar)
R_t	Tube radius.	(m)
R	Radial coordinates.	(m)
$T_{r,z}$	Temperature at any position through the bed..	(^oC)
T_w	Wall temperature tube.	(^oC)
T_o	Inlet temperature to the reactor.	(^oC)
$T_{c.}$	Center temperature of the first experimentally determined radial temperature profile.	(^oC)
V_p	Volume of particle.	(m^3)
Z	Axial coordinate (bed depth).	(m)
z_o	The axial position of the first experimentally determined radial temperature profile.	(m)
μ_g	Dynamic viscosity of the gas.	(Pa.Sec)

List of Dimensionless Group Numbers

T'	$=(T_w - T)/(T_w - T_o)$	(--)
β_i	Biot number = ($(h_w * R_t)/K_{er}$).	(--)
r'	$= r / R_t$.	(--)
ζ	$= z/L$.	(--)
α'	$= \dfrac{K_{er} * L}{GCpR_t^{\,2}}$.	(--)
θ	$= \dfrac{(T_w - T)/(T_w - T_o)}{(T_w - T_C)/(T_w - T_o)} = \dfrac{T_w - T}{T_w - T_c}$	(--)
δ	$= \dfrac{\frac{Z}{L} - \frac{Z_o}{L}}{1 - \frac{Z_o}{L}} = \dfrac{\zeta - \zeta_o}{1 - \zeta_o}$.	(--)

Re_p	Particle Reynolds number = ($\dfrac{G*d_p}{\mu_g}$).	(--)
Re_C	Channel Reynolds number = ($\dfrac{G*d_h}{\mu_g}$)	(--)

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