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HYBRID MODELING OF WELL-MIXED MODEL FOR FLUIDIZED BED REACTORS USING ARTIFICIAL NEURAL NETWORKS

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ABSTRACT

In this work, an artificial neural network approach is used to capture the reactor characteristics in terms of heat and mass transfer based on published experimental data. The developed ANN-based heat and mass transfer coefficients relations were used in a conventional FCR model and simulated under industrial operating conditions. The hybrid model predictions of the melt-flow index and the emulsion temperature were compared to industrial measurements as well as published models. The predictive quality of the hybrid model was superior to other models. This modeling approach can be used as an alternative to conventional modeling methods.

Keywords: *Fluidized bed reactor, heat transfer, mass transfer, three phase, catalytic reactor, neural networks*

INTRODUCTION

Mass and heat transfer coefficients estimation for the fluidized bed reactors has been the focus of many previous research studies. The correlations attained by these studies are used in a broad range of applications in many chemical processes involving gas–solid and solid-catalyzed gas-phase reactions. Production of linear low density polyethylene (LLDPE) through heterogeneous Ziegler–Natta catalysts is an example of such industrial applications of fluidized beds. Due to operation at lower pressures and temperatures, there is no need to use solvents and better heat removal can be attained, compared to the other polyethylene production processes. Gas-phase polymerization of ethylene in fluidized beds is now widely employed at industrial scale [Xie et al. 1994].

During the past four decades many theoretical as well as practical studies have been done in this area. Nonetheless, the diversity of operating conditions as well as reactor geometry make it necessary to give more attention to this area and study it under industrial conditions. Previous experimental investigations revealed that generally the heat transfer coefficient (h) in three-phase fluidized beds is an increasing function of the respective gas and liquid velocities, the size and density of the particles, column diameter, and the thermal conductivity and heat capacity of the liquid phase, but a decreasing function of the dynamic viscosity of the liquid, and probably of the diameter of a heating cylinder immersed inside the bed. On the other hand, the mass transfer coefficient (K_La) increases with increasing gas and liquid velocities, and the size and density of particles but it decreases with increasing surface tension and viscosity of the liquid phase in the beds (Lee et al., 1993). They conducted experimental and theoretical investigations on the heat and mass transfer coefficients of fluidized-bed catalytic reactor for two phases. Their mass transfer coefficient correlation is given in equation (1). This correlation is a function of diffusivity and bubble diameter only. Due to the lack of these important parameters, the prediction accuracy of these correlations is not high.

$$k_{be} = 0.975\, D_{AB}^{0.5} \left(\frac{g}{d_b} \right)^{0.25} \qquad (1)$$

Kunii-Levenspiel (1969) developed the bubbling-bed model for fluidized beds. They correlated experimental data of ozone to predict the mass and heat transfer coefficients. Despite the simplicity of these correlations, they proved to be of good prediction efficiency and that's why they are used (until recently) in applications involving fluidized beds. Several research efforts for production and particle growth models utilized these correlations as a basis for modeling fluidized bed reactors (Choi and Ray 1985, Hutchinson et al. 1992, McAuley et al. 1994,

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Fernandes and Lona 1999, Fernandes and Lona 2001, Alizadeh et al. 2004 , Ahmmed s ibrehem et al. 2008). The mass and heat transfer phenomena in the Kunii-Levenspiel model is described as a two steps process. The transfer is directed from the bubble to the cloud phase and then from cloud phase to emulsion phase and the overall mass transfer coefficient is composed of both components. Equations 2 to 4 represent these correlations respectively.

$$k_{bc} = 4.5\frac{u_{mf}}{d_b} + 5.85 D_{AB}^{0.5} g^{0.25} / d_b^{1.25} \quad (2)$$

$$k_{ce} = 6.77\left(\varepsilon_{mf} D_{AB} \frac{u_b}{d_b^3} \right)^{0.5} \quad (3)$$

$$k_{be} = \left(1/k_{bc} + 1/k_{ce} \right)^{-1} \quad (4)$$

Later, Sit and Grace (1981) studied the same fluidized-bed process and correlated their experimental data for mass transfer as described by equation 5. These correlations suffer from the same shortcoming of that found in Kunii-Levenspiel model as well as ignoring the effect of the cloud phase. The applicability of this relation is limited due to these two missing effects.

$$k_{be} = \frac{u_{mf}}{3} + \left(4\varepsilon_{mf} D_{AB} u_b / \left(3.14 d_b \right) \right)^{0.5} \quad (5)$$

Three phase mass transfer studies for fluidized-beds are limited in number because of the technical difficulties involved in conducting them. Peters et al. (1982) conducted their study on such systems. Similar to the two phase systems, the mass transfer occurs as a two step process. Equation 6-8 describes the mass transfer correlation obtained by their study. It can be seen that these correlation lack the effect of fluidized-bed height.

$$k_{bc} = 2\frac{u_{mf}}{d_b} \quad (6)$$

$$k_{ce} = 12\left(4\varepsilon_{mf} D_{AB} \frac{u_b}{3.14 d_b^3} \right)^{0.5} \quad (7)$$

$$k_{be} = \left(1/k_{bc} + 1/k_{ce} \right)^{-1} \quad (8)$$

Single-particle models are based on unsteady-state heat transfer between a particle and a heat transfer surface. In this approach, particles move individually and only the first row of particles is considered significant to the heat transfer process. Zabrodsky's (1966) model is one of the manifestations of the single-particle theory. He expressed the particle heat transfer coefficient as:

$$h_{pc} = C_{ps} \rho_s d_p \left[\frac{0.806(1-\varepsilon)^{2/3}}{\tau_r} \right] \left[1 - \exp\left(\frac{-9 k_f \tau_r}{C_{ps} \rho_s d_p^2} \right) \right] \quad (9)$$

The pioneering work in the context of the Cluster-Based Approach is that of Mickley and Fairbanks (1955). They assumed that particles move together as packets, which are swept to and from heat transfer surfaces by the mixing action of rising bubbles. A semi-infinite solid idealization for the emulsion is used to determine the heat transfer particle convection coefficient as:

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$$h_{pc} = (1-\delta)\sqrt{\frac{k_{mf} C_{mf} \rho_{mf} \left(1-\varepsilon_{mf}\right)}{\pi\tau}} \tag{10}$$

Baskakov (1964) and Bock *et al.* (1983) proposed a modification for the previous model, by introducing an additional thermal resistance of the gas film on the heat transfer surface. In these models heat transfer by the particle convection is modeled as the process of unstationary conduction of the particle clusters which are in contact with heat exchange surface for a definite period of time. Nearby the heat exchange surface there is a gas film which transfers heat by gas conduction. Particle convection heat transfer coefficient is defined as:

$$h_{pc} = \left(\frac{1}{h_f} + \sqrt{\frac{\pi\tau}{k_{mf} C_{mf} \rho_{mf} \left(1-\varepsilon_{mf}\right)}} \right)^{-1} \tag{11}$$

Heat transfer coefficient of the gas conduction through the gas boundary layer on the heat exchange surface is due to a thin gas layer between the packet and the wall. This coefficient is expressed as: $h_f = h_{\max}\left(1-\varepsilon_{mf}\right)/K$. Bock *et al.* evaluated the constant K to be 3 for all tested solid particles. The maximum heat transfer coefficient is given as a function of the particle mean free path to the characteristic body length (l_1) by:

$$h_{max} = \frac{4k_f}{d_p}\left[\left(1-l_1/d_p\right)\ln\left(1+d_p/l_1\right)-1\right] \tag{12}$$

Lints and Glicksman (1993), described the convective thermal exchange at the wall of a circulating fluidized bed as two processes in series leading to an overall heat transfer coefficient h defined as:

$$h_{pc} = f_h h_c + \left(1-f_h\right) h_g \tag{13}$$

The first of these processes is an exchange characterized by the coefficient h_c involving particle clusters covering the fraction f_h of the wall surface. The second has coefficient h_g and involves an emulsion around the clusters that is largely devoid of particles. Lints and Glicksman then modeled the heat transfer to clusters as two parallel processes, namely a conduction through the thin gas film of thickness δ followed by a convective exchange to the clusters with coefficient h_H,

$$1/h_c = \delta/k_f + 1/h_H \tag{14}$$

To capture h_H, Lints and Glicksman borrowed from the model of Mickley and Fairbanks (1955) for bubbling beds. In that model, the authors treated the emulsion as a semi-infinite homogeneous medium and adopted the classical heat flux expression for transient conduction into a semi-infinite slab. Then, after invoking effective thermal properties for the emulsion phase and time averaging, they derived the form of the wall heat transfer coefficient as:

$$h_H \approx \sqrt{k_e \rho_s c_p \nu_c / \tau_c} \tag{12}$$

where k_e is the effective conductivity of the emulsion phase, τ_c is its average contact time with the wall, ν_c is its solid volume fraction and ρ_s and c_p are, respectively, the material density and specific heat of the solids.

Elizabeth and Louge (2000), simplified the previous model by assuming that the effective cluster conductivity is governed by the gas conductivity k, neglecting the second term in Eq.(3), and that the convective transfer to the

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clusters dominates the conduction through the thin gas layer. They used and verified experimentally an equation of the form:

$$Nu_d = \frac{hd_s}{k} \approx f_h \sqrt{\tau_p \nu_c / \tau_c} \tag{15}$$

where $\tau_p = \rho_s c_p d_s^2 / k$ is a characteristic time for the heating of a particle of diameter d_s.

Recently, Karimipour et al. (2007), proposed to split the process of heat transfer through clusters into two periods with respect to time. Using the concept of penetration depth of temperature to model the heat transfer coefficient in the first period, they solved a heat balance equation and derived the following relation for the cluster heat transfer coefficient which is similar in form to that of Mickley and Fairbanks:

$$h_{cc} = \left(\frac{1.5t}{k_c \rho_c c_c} \right)^{-0.5} \tag{16}$$

For the second period where the wall-heating effect has reached the other side of the cluster, the penetration depth is equal to the width of the cluster. Their analysis revealed the following equation:

$$h_{cc} = \left(\frac{k_c}{d_c} \right) \Bigg/ \left(\frac{1}{2} + \left(\frac{1}{2} + \frac{1}{Bi} \right) \left[1 - \exp\left(- \frac{m\,(t - t_0)}{1 + Bi\,/\,3} \right) \right] \right) \tag{17}$$

where, Bi :the Biot number = $(h_g\, d_c / k_c)$,

m : $h_g /(\rho_c\, c_p d_c)$,

t : time (s),

t_0 : Penetration time of the applied heat flux to the other side of the cluster (s),

h_g : heat transfer coefficient of the gas in turbulent flow,

k_c, d_c, ρ_c refer to the particles cluster properties.

HYBRID MODEL SIMULATION AND TESTING

Two versions of the FCR well-mixed model were prepared as can be seen in Figure (1). The first one is a conventional version of the model were the heat and mass transfer coefficients are predicted for different polyethylene grades as can be seen in Figures (2) and (3). The second version is a hybrid model with ANN-based coefficients models. In the later model, the achieved trained ANN-based heat and mass transfer models were implemented in the well-mixed model as can be seen in Figure (4). The emulsion temperature and the MFI values for the three models as compared to the experimental data are shown in Fig (5-a). The ANN-hybrid model was superior to the well-mixed model in predicting the MFI values as indicated by the least residuals in Figure (5-b). The emulsion temperature predictions of the two models are in favor of the ANN-hybrid model with one polymer grade exception coded as HD2 in the figure where the conventional well-mixed model is better. For a better performance of the ANN-based model at this particular grade, the ANN model should be trained with reactor data resembling the properties of the fluidized bed at the same conditions. Unfortunately, these data weren't available at the time of this analysis. In general we can say the hybrid model attained the best in performance compared to the well mixed model.

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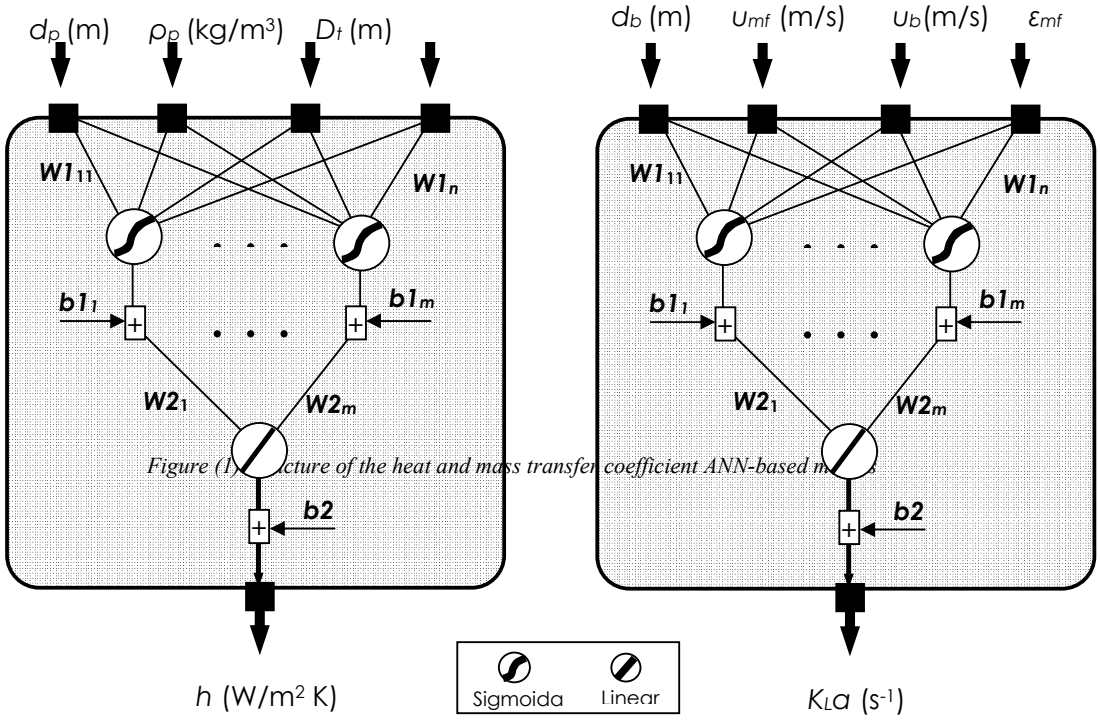

Figure (1) Structure of the heat and mass transfer coefficient ANN-based models

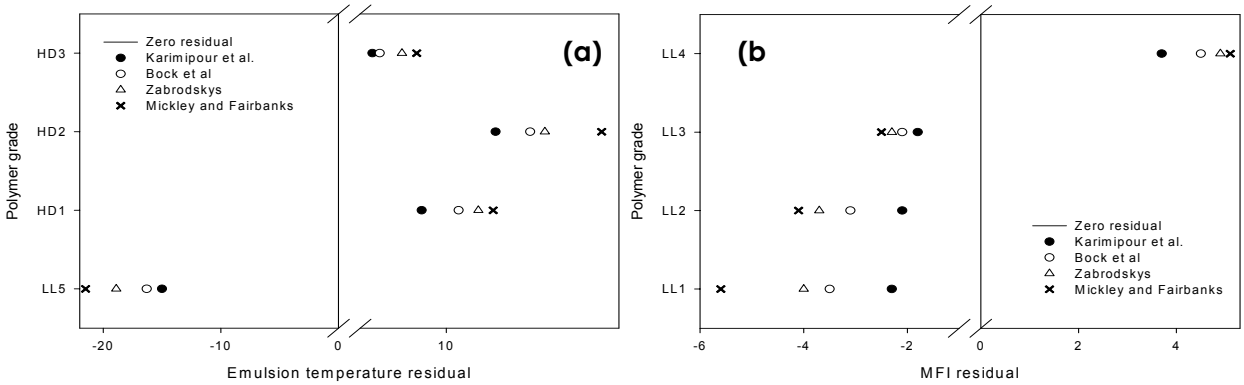

Figure (2) Effect of heat transfer coefficient correlation on the well-mixed model prediction for different polyethylene grades (a) Emulsion temperature (b) MFI values

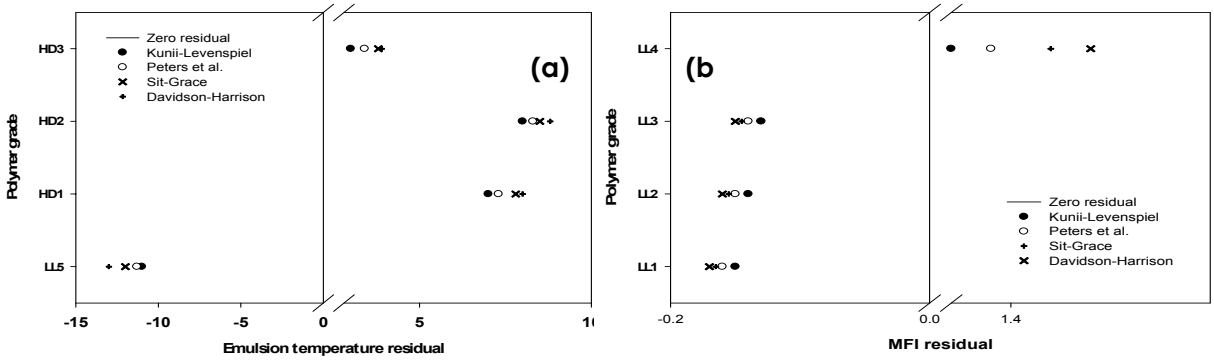

Figure (3) Effect of mass transfer coefficient correlation on the well-mixed model prediction for different polyethylene grades (a) Emulsion temperature (b) MFI values

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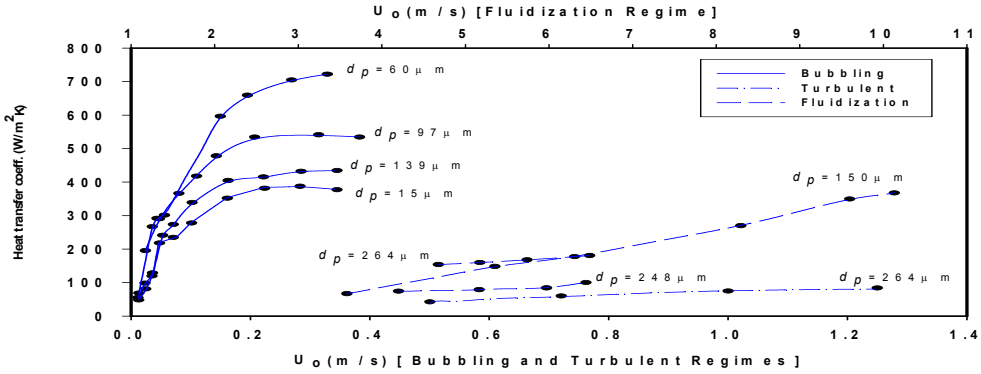

Figure (4) ANN heat transfer coefficient predictions as a function of superficial gas velocity at different hydrodynamic regimes and particles diameter.

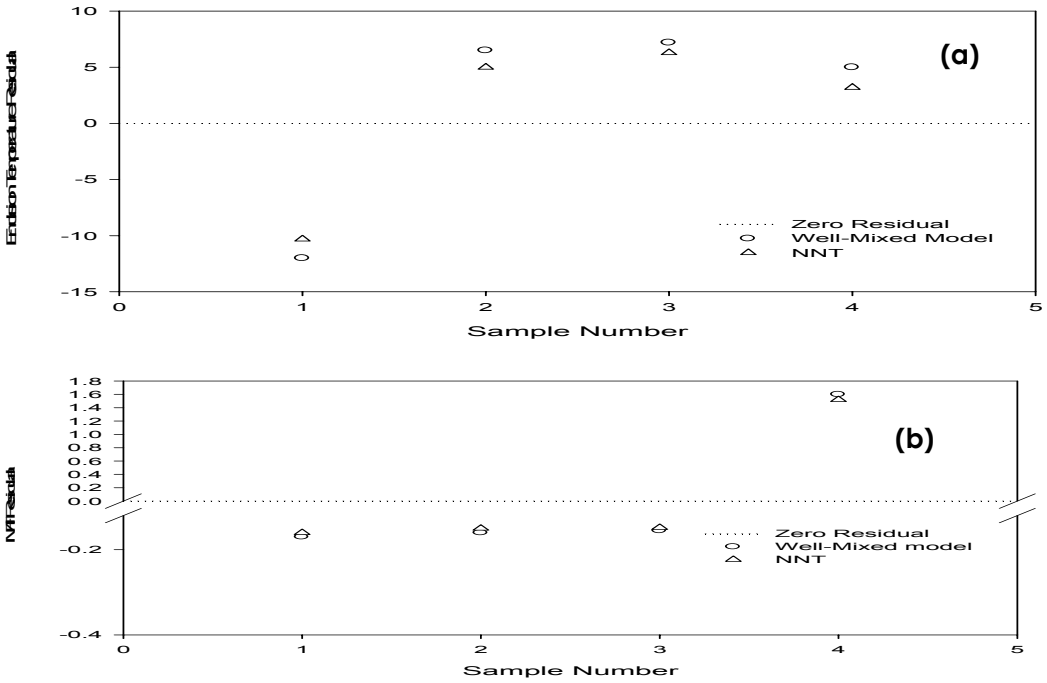

Figure (5): Actual polymer grade plant data versus FCR well-mixed model predictions and ANN modeling approaches (a) emulsion temperature (b) MFI values.

CONCLUSION

This work introduced a new technique of implementing ANN-based models of the heat and mass transfer coefficients in the mechanistic modeling of the FCRs. Experimentally reported data were used as a basis for training two ANNs to capture the relationships of heat and mass transfer coefficients. The two ANN-based models accept operational variables of the FCR as inputs and predict the coefficients within the operational ranges of the collected data. The trained ANN-based heat and mass transfer coefficient models were validated with experimental data and found to attain high prediction quality. Industrial light and heavy density polyethylene grades data were used to validate the hybrid ANN-based well-mixed FCR model. The hybrid model showed superior prediction quality as compared to conventional mechanistic models. The success of such models triggers the attention for implementing such modeling strategy for other fluidized reactor geometries and at other operating conditions.

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