Letters to the Editors

Comments on discrimination of three approaches to evaluate heat fluxes for wall-cooled fixed bed chemical reactors

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Dear Sirs,

In a recent paper, Vortmeyer and Haidegger (1991) presented a comparison of three models to evaluate heat fluxes for wall-cooled fixed-bed chemical reactors. We have the following comments about this paper.

Vortmeyer and Haidegger used the fast exothermic oxidation of ethane to test three (quasi-) homogeneous models. They concluded that the wall heat conduction model was the best because of better agreement with the experimental data and physical reality. We, however, feel that under the reaction conditions considered the homogeneous models are not applicable, and a heterogeneous model should have been used instead. In a recent article (Wijngaarden and Westerterp, 1989) we explained how improper dependences and relations are introduced when an improper model is used.

To prove this, we tested Vortmeyer and Haidegger's experimental data from their graphs 3(a)-(g), with a criterion given by Mears (1971). The interparticle heat transfer resist-

ance can be neglected if

$$\frac{-(\Delta H_r)R(T_g)d_p}{2\alpha_p T_g} < 0.15 \frac{RT_g}{E_a}$$

The results of our calculations are presented in Tables 1 and 2. However, the Nu_p relations are not given in the paper of Vortmeyer and Haidegger. In the VDI-Wärmeatlas (VDI, 1988) Nu_p numbers are given for Pe above 500. We refer to the work of Kunii and Suzuki (1967), from which it can be found that Nu_p for the given Re_p values varies between 4 and 12. This leads to $47 < \alpha_p < 141$ W/(m² K). The measured temperatures were assumed to be gas temperatures. In all experiments, particularly just before the hot spot where concentrations and temperatures are relatively high, the criterion is not satisfied. Therefore, temperature differences between catalyst surface and surrounding gas cannot be neglected.

Table 1. Calculation of Mears criterion and catalyst temperature for different experimental points and $Nu_p = 4$

Graph number	T_g (K)	y	$\frac{-(\Delta H_r)R(T_g)d_p}{2\alpha_p T_g}$	$0.15 \frac{RT_g}{E_a}$	T_k (K)	$\frac{R(T_g)}{R(T_k)}$
3(a)	670	0.0033	0.069	0.009	734	0.40
3(b)	730	0.0010	0.046	0.010	753	0.81
3(c)	700	0.0017	0.057	0.009	732	0.68
3(d)	670	0.0028	0.037	0.009	690	0.69
3(e)	660	0.0033	0.056	0.009	707	0.43
3(f)	680	0.0030	0.075	0.009	744	0.44
3(g)	700	0.0031	0.103	0.009	797	0.41

Table 2. Calculation of Mears criterion and catalyst temperature for different experimental points and $Nu_p = 12$

Graph number	T_g (K)	y	$\frac{-(\Delta H_r)R(T_g)d_p}{2\alpha_p T_g}$	$0.15 \frac{RT_g}{E_a}$	T_k (K)	$\frac{R(T_g)}{R(T_k)}$
3(a)	670	0.0033	0.023	0.009	680	0.82
3(b)	730	0.0010	0.015	0.010	737	0.93
3(c)	700	0.0017	0.019	0.009	708	0.89
3(d)	670	0.0018	0.012	0.009	675	0.90
3(e)	660	0.0033	0.019	0.009	668	0.83
3(f)	680	0.0030	0.025	0.009	691	0.83
3(g)	700	0.0031	0.034	0.009	716	0.80

Next, the steady-state heat balance for one catalyst particle,

$$\alpha_p A_p (T_k - T_g) = \frac{V_p R(T_k)(-\Delta H_r)}{(1-\varepsilon)}$$

can be solved for T_k using the same data. It was assumed that all mass transfer resistances are lumped together in the description of the reaction rate. Temperature differences between gas and catalyst varying between 5 and 97 K have been calculated (see Tables 1 and 2). These temperature differences result in a difference in reaction rate varying from 7% to 60% based on gas and catalyst temperatures. This will certainly lead to erroneous results when using the (quasi-) homogeneous model, particularly in the hot-spot region.

In conclusion, we feel that in important parts of the reactor the temperature differences between the particle and the bed are so large that a heterogeneous model should have been used, and that the experimental data collected are not adequate to draw conclusions about the applicability of certain (quasi-) homogeneous models.

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NOTATION

A_p external surface area of the catalyst pellet, m²
 d_p particle diameter equivalent ($= 6V_p/A_p$), m

E_a energy of activation, J/mol
 $(-\Delta H_r)$ reaction enthalpy, J/mol: for exothermic reactions ($-\Delta H_r > 0$)
 Nu particle Nusselt number ($= \alpha_p d_p / \lambda_g$)
 R gas constant, 8.314 J/mol K
 $R(T)$ reaction rate at temperature T , mol/m³ (packed-bed volume) s
 T_g temperature of the gas, K
 T_k temperature of the catalyst surface, K
 V_p volume of the catalyst pellet, m³
 y mass fraction of ethane

Greek letters

α_p heat transfer coefficient between the particle and the gas, W/m² K
 ε porosity, volume fraction of the gas in the bed

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Authors' reply to comments by K. R. Westerterp *et al.*

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Dear Sirs,

Westerterp *et al.* write that because of too large temperature differences between gas and catalyst particles, we should not have chosen the quasi-homogeneous model to interpret our experimental results [1]. Maximum temperature differences of 97 K are predicted in Table 1 corresponding to the experimental situation shown in Graph 3(g) of our paper [1].

Since in the course of our experimental work we have also felt that, due to chosen inlet and cooling temperatures, this experiment should exhibit the largest temperature differences between particle and gas, we have measured temperature differences [2] between gas and solid on the centerline of the reactor. Due to technical difficulties we were able to do so only in position 4 of Fig. 1 where, incidentally, the wall-cooled reactor had its hot spot. To measure the particle temperature, a specially prepared catalyst particle with a thermocouple in the center was carefully placed on the centerline of the reactor at location 4 of Fig. 1. A second thermocouple was placed close to the surface of the particle to obtain the gas temperature. We took five measurements

under steady-state conditions within a few hours and got readings between 4 and 5 K averaging $(T_k - T_g) = 4.5$ K instead of 10 K as evaluated by us according to the equations of Westerterp *et al.* At location 3 in Fig. 1, which is the point chosen by Westerterp *et al.*, naturally larger differences between the temperatures of catalyst particles and gas have to be expected. Westerterp *et al.* claim in Table 1 that the difference may be up to $(T_k - T_g) = 97$ K.

We have extended their calculations between thermocouple positions 2 and 4 and have obtained catalyst temperatures T_k as plotted in Fig. 1 of this paper. These temperatures are just unimaginable. This view is supported by the experimental work of Wicke *et al.* [3] who, even under much more severe conditions than in our work (temperature rise of 300 K under adiabatic conditions), did not observe such temperature differences for comparable flow conditions. We therefore reject the computed solid temperatures in Table 1 of the letter of Westerterp *et al.* as unrealistic due to too low particle to fluid heat transfer coefficients chosen for their calculations.

Let us follow now the theoretical arguments of Westerterp