LETTERS TO THE EDITOR

To the Editor:

Kalogerakis and Luus (*AIChE J.*, **26**, 670, 1980) studied the problem of enlarging the region of convergence when estimating the parameters in dynamic models of chemical kinetics via quasilinearization. In this letter, we would like to suggest an alternative approach to parameter estimation: the direct integral method suggested by Himmelblau, Jones and Bischoff (*Ind. Eng. Chem. Fund.*, **6**, 539, 1967) and improved by Tang (*Ind. Eng. Chem. Fund.*, **10**, 321, 1971), in which no initial estimates are required. To apply the direct integral method, the following conditions must be fulfilled:

1. All the concentrations appearing in the rate expressions must be observed.
2. The rate expressions must be linear in the parameters.
3. The time intervals between the observations must not be too large.

The first two requirements are met in the particular example studied by Kalogerakis and Luus, and the third one can be fulfilled by simply deleting the last data point. Solving the linear least squares (LSQ) problem obtained after spline interpolation yielded the following estimates, with 95% confidence intervals: $k_1 = 353.9 \pm 0.6$, $k_2 = 403.9 \pm 3.5$. These are mainly in agreement with the estimates $k_1 = 354.6$, $k_2 = 400.2$ given by Kalogerakis and Luus. A further advantage of the direct integral method over the traditional LSQ approach is that the computational effort is considerably less.

Finally, we would like to add some remarks concerning the applicability of the direct integral method. There is no evidence that the minimization of the traditional LSQ objective function gives less biased estimates than the direct integral LSQ objective function. On the contrary, computational experience with complex kinetic models (A. Yermakova, P. Valko, and A. S. Umbetov, *Hung. J. Ind. Chem.*, **8**, 205, 1980) shows that the Jacobian of the direct integral LSQ objective function may be well-conditioned when the Jacobian of the traditional LSQ objective function is ill-conditioned. This advantage of the direct integral method—at a certain level of data noise—may outweigh the inherent error due to the approximation of integrals from observed data. Also, the direct integral method can be generalized to parameter estimation problems with nonlinear dependence of the rate expressions on the parameters, as it was shown for a special class of nonlinearities (P. Valko, A. Yermakova, and S. Vajda, *Hung. J. Ind. Chem.*, **8**, 437, 1980).

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To the Editor:

We read with interest the article by Levy and Lockwood, "Laser-Doppler Measurements of Flow in Freeboard of a Fluidized Bed" (**30**, p. 174, 1983). Measurements such as these are crucial for gaining a physical understanding of the freeboard. Their major result, a nonuniform mean-velocity field contradicts our own recent results (Kale, 1984), which showed very uniform gas-velocity profiles above a bed of 50–100 μm diameter glass beads fluidized at approximately 30 times the minimum fluidization velocity.

In an attempt to duplicate their results, we loaded our bed with 600–800 μm glass beads and fluidized it at approximately 1.1 times the minimum fluidization velocity. The bed has a square cross section which was somewhat smaller than Levy and Lockwood's rectangular bed (0.15 m × 0.15 m vs. 0.3 m × 0.6 m). The bed heights were similar. Alumina seed particles, 1 μm in diameter, were added to the bed for Laser-Doppler-Anemometer measurements of the gas-phase velocity. The resulting mean velocity profile at a location 0.88 m above the 0.25 m deep bed was very uniform, except for thin boundary layers (<5 mm) near the walls. There were no velocity peaks near the walls as observed by Levy and Lockwood.

It seems likely that the difference is due to the shapes of the two beds, i.e., square or rectangular. Bubbles in the rectangular bed probably reach all the way to the wall surface, allowing gas to flow up the wall. The bubble locations appeared to be randomly distributed in the square bed. The significant differences indicate the need for further research on the basic physics governing the freeboard flow.

LITERATURE CITED

1. Kale, S. R., "An Experimental Investigation of Gas-Particle Flows through Diffusers in the Freeboard Region of Fluidized Beds," Ph.D. thesis, Dept. of Mechanical Engineering, Stanford University, May 1984.

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Reply:

This letter is in response to the contention mentioned in the letter of Christopher Rogers, S. R. Kail, and J. Eaton from Stanford University in regard to my article "Laser Doppler Measurements of Flow in the Freeboard of Fluidized Bed." I would like to point out a few facts which will probably answer some of their questions and clarify the discussed phenomena concerning the detection of a nonuniform velocity profile across the freeboard of a fluidized bed. The Stanford University group performed velocity measurements in the freeboard of a glass beads fluidized bed. Their results showed uniform velocity profiles in two experimental conditions:

1. 50–100 μm diameter glass beads at velocities up to 30 times the Umf, and
2. 600–800 μm diameter glass beads at much lower fluidizing velocities 1.1 times the Umf.

Their fluidized bed had a square 15 cm × 15 cm cross section and bed height of 25 cm.

The work mentioned in our paper concerned a rectangular (60 cm × 30 cm) bed cross section. The bed height was 30 cm to 36 cm when using 1 mm diameter sand particles and 20 cm when using 0.4 mm diameter sand particles. During our measurements a clear (and repetitive) phenomenon was observed at which the mean velocity profile of the gas flow across the bed was nonuniform and had two maximums near the walls with a dip in the center. An explanation of the phenomena given in the papers stated that with our operating conditions, toroidial gas vortices were generated during each bubble burst at the bed surface. This phenomena was confirmed by flow visualization and was recorded by an 8 mm movie camera. The rotational direction of these vortices was upwards at the outer edge and downwards on the inner side. The vortices were carried upwards in the freeboard by the fluidizing air. During our experiments the diameter of the bubbles at the bed surface was about 10 cm and one to three of them burst at the same time, ejecting the sand particles located above, just before the burst took place. These particles, when descending, dragged gas, which initiated the formation of the vortex. This phenomena was schematically illustrated in the paper.

The experimental conditions of the Stanford University group could not create conditions required for this type of vortex formation which is responsible for the special velocity profile. Their experimental cases presented two extreme flow conditions not relevant to our original application (F.B.C. – 1.5 Umf $\lesssim$ Uf $\lesssim$ 10 Umf) and therefore not analyzed. Their first experimental condition included the use of fine particles with very high fluidizing velocities. This is a situation of slug-flow, where no discrete bubble burst pattern exists at the bed surface. In slug-flow larger amounts of sand lumps are probably ejected, not necessarily generating the mentioned vortices. Their second experimental case included larger particles and much lower fluidizing velocities. The amount of the flow rate was too low and insufficient for generating the large bubbles and the consequent vortices, and, therefore, resulted in the obvious uniform velocity profile. Two other important parameters which influence the particles' motion in the freeboard during the bubble burst were:

1. The shape factor of the particles ($\phi = 1$ for sphere, ~0.6 for sand): this had an influence on the drag force and hence on the particle's trajectory;
2. The bed dimension: our bed's cross section area was selected to be large enough to minimize wall effects.

The size of the bubbles at the surface was about 10 cm in diameter. The particles ejected during the burst followed a trajectory similar to an umbrella shape (from the center upwards, radially and downwards). When descending, they occupied a larger diameter, about twice the bubble's initial diameter. The vortex generated was even larger. The fact that a much smaller bed cross section was used at Stanford (about 1/6 of our area) added to the difficulties in reproducing the results presented in the paper.

In an attempt to duplicate experimental conditions and recheck the phenomena, I would recommend performing measurements with conditions where the diameter of the bubbles is about 1/3 of the bed width, with fluidizing velocities in the range of 1.5 Umf to about 5 Umf. This would mean working with a shallower bed and relatively smaller particles ($dp \sim 200\ \mu m$). An air distributor with uniform air distribution is required (such as porosite plate). Under these conditions, I am confident that using an appropriate measuring system (a frequency shift is necessary in the Laser Doppler anemometer), the mentioned nonuniform velocity profile will be detected.

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