LETTERS TO THE EDITOR

To the Editor:

In "Dynamic and Steady-State Features of a Cooled Countercurrent Flow Reactor"(October 2000) Garg, Luss and Khinast derived balance Eqs. 2, 5, and 6, which contain model parameters different from those usually appearing in the models of fixed-bed catalytic reactors.

Nevertheless, simply multiplying these equations by the Damköhler number Da leads to a form of these parameters much more common in chemical reaction engineering. As a result of this transformation, the Peclet number of longitudinal mass dispersion is given by the relation

$$Pe_m = \frac{\phi_m^2}{Da} = \frac{LU}{\epsilon D_{ax}}.$$

When substituting the parameter values quoted in the publication, the Peclet number yields an extremely high value of 54,000, not encountered in the quantitative description of fixed-bed reactors packed with catalyst pellets.

This is caused by the assumption of the value of the mass dispersion coefficient D_{ax}, which is lower by two orders of magnitude than the values of this quantity usually occurring in packed beds. Consequently, the terms in balance (Eqs. 5 and 6) defining the mass transport by dispersion are completely negligible in comparison with the convective flow of the reaction mixture. However, above all, I am afraid that the very interesting bifurcation analysis and the dynamic behavior of the reactor considered are rather of academic significance, and provide only limited possibilities of application in the design of chemical reactors of this type.

The authors, discussing the influence of the Damköhler number on the shape of the temperature profiles in the reactor (second column page 2035) refer to Figure 4, which, however, illustrates the influence of the cooling capacity Δ at a constant Damköhler number and is, therefore, misleading for the reader.

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Reply:

(1) Burghardt's claim that common dimensionless packed-bed reactor models contain a Peclet number is correct. However, if one wants to examine the influence of a continuous change of the feed velocity (as in our article), or, equivalently, of the residence time, the formulation we use is more convenient for the construction of bifurcation diagrams. The reason is that the velocity appears only in one dimensionless group, the Damköhler number, while using a Peclet number the velocity appears also in the cooling term. This fact has been recognized by other researchers and led them (Subramanian and Balakotaiah, 1996) and us to use this formulation.

(2) The mass Peclet number in industrial packed-bed reactors is of the order of one thousand. Our base case parameters were based on the experimental data of Nieken et al. (1995), who used a honeycomb catalytic reactor in their study of a reverse flow reactor. It is well known that in packed-bed reactors the influence of the axial mass dispersion is negligible. Replacing a mass Peclet number of 1,000, even by 100,000, does not affect the predicted reactor behavior.

(3) Under adiabatic conditions the CFR ignited state is always stable. We noted that the ignited steady state of the cooled CFR may become unstable and led to a stable periodic or complex dynamic state when its temperature profile had two distinct peaks. We also noted that for the complex states the ratio between q_w, the time-averaged rate of heat removal through the wall, and q_c, the time-averaged heat convected by the effluents, was of the order of 1.0–0.1. These complex dynamic states existed within a bounded range of intermediate Damköhler numbers. Professor Burghardt is correct that Figure 4 describes the influence of the cooling capacity term Δ and not of the Damköhler number $Da(T_0)$ on the temperature profile. We referred to Figure 4 to show the different types of profiles that may occur for different Damköhler numbers. To clarify this, we enclose Figure 1 of three temperature profiles at a fixed cooling capacity of $\Delta = 600$ obtained at different Damköhler numbers by changing the feed velocity. Note that in this case decreasing the $Da(T_0)$ value increases the total heat release in the reactor at complete conversion. At high $Da(T_0)$, the stable ignited state has a single temperature peak at the reactor center and $q_c/q_w = 0.01$. At low $Da(T_0)$, a flat stable temperature profile exists in the reactor center and $q_c/q_w = 19.91$. The symmetric steady state is unstable at the intermediate $Da(T_0)$ number and its temperature profile has two peaks. A stable ignited

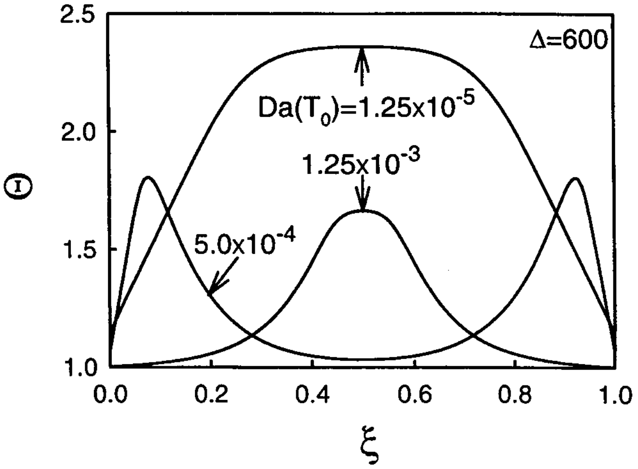

Figure 1. Dependence of the temperature profiles on $Da(T_0)$.

The calculations are based on the parameters used in the original article. $Da(T_0)$ changes reflect changes in feed velocity.

period-1 state exists in this case with $q_c/q_w = 0.26$.

(4) Professor Burghardt claims that the bifurcation analysis and the dynamic behavior are only of academic interest. We disagree. The existence of complex dynamics in practice depends very strongly on the level of cooling, the rate expression, and its temperature dependence, the heat of reaction and the feed concentration. A study based on one kinetic rate expression and one set of parameters cannot be used to make these predictions.

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To the Editor:

In "Fluidization of Solids with CO_2 at Pressures from Ambient to Supercritical" (May 2000), Marzocchella and Salatino report results of an experimental investigation into the effect of fluid pressure on the fluidization of glass beads with CO_2. At the higher pressures tested (up to 80 bar), the fluid density increases markedly—close to midway between the values for ambient air and ambient water. The tests therefore provide data under uncommon conditions which may be used to test the validity of general predictive relations for the fluidized state.

Marzocchella and Salatino's Table 2 goes some way to performing this useful function: it shows observed values of both the minimum fluidization velocity U_{mf} and the Richardson-Zaki homogeneous bed expansion parameters n and U_t to be in very reasonable agreement with independent correlations throughout the range of operation. It also reports observed values of void-fraction at the minimum bubbling condition ϵ_{mb}, but this time fails to provide comparisons with available predictions. This omission may be remedied by the following addition to their Table 2:

Material	Ballotini 88 μm				
Pressure (bar)	1	20	40	60	80
ϵ_{mb} (Obs.)	0.44	0.44	0.47	0.50	0.58
ϵ_{mb} (Pred.)*	0.40 $< \epsilon_{mf}$	0.40 $< \epsilon_{mf}$	0.40	0.43	0.60
Material	Ballotini 175 μm				
Pressure (bar)	1	20	40	60	80
ϵ_{mb} (Obs.)	0.40	0.41	0.43	0.45	0.50
ϵ_{mb} (Pred.)*	–	0.38 $< \epsilon_{mf}$	0.38 $< \epsilon_{mf}$	0.38 $< \epsilon_{mf}$	0.56

*Calculated from Foscolo and Gibilaro (1987) using experimental values for U_t and n reported by Marzocchella and Salatino (2000).

The agreement is far from perfect, but the simple fluid-dynamic criterion predicts well the effect of fluid pressure in increasing the range of homogeneous expansion—the most significant finding of the investigation, for which Marzocchella and Salatino offer no explanation whatsoever. Rather than accepting the notion of a fluid-dynamic mechanism being responsible for the state of homogeneous gas-fluidization, they repeat the view that the particles are glued together into a sustainable structure, strong enough to resist the tendency to form bubbles, by "interparticle forces." As to the nature of these forces, they are surprisingly reticent. For the bulk of the experiments, which were performed at elevated pressures, above the critical temperature, no clue whatsoever is provided as to what it is that, supposedly, binds together the dry glass particles (of up to nearly 0.2 mm diameter).

For the two experiments performed at ambient pressure and temperature, they are more forthcoming. Under these conditions, they postulate partial condensation of CO_2, leading to the formation of liquid bridges between the particles through which the structure is sustained by capillary forces. No evidence is provided in support of this esoteric hypothesis, which is hardly surprising as elementary thermodynamic considerations show the maximum capillary diameter which would lead to CO_2 condensation under these conditions to be around 10 Angstroms—a size of the order of lattice separation distances in a crystal!

If interparticle forces are indeed responsible for the homogeneous state, then the authors' own results, reported in Table 2, include an extraordinary coincidence which goes unremarked. They find that their homogeneous gas beds expand, with increasing gas velocity, in good quantitative agreement with the Richardson-Zaki correlation - which was obtained from experiments on liquid-fluidized beds, supposedly controlled by a quite different (fluid-dynamic) mechanism. Not even the most tentative of deductions is drawn from this observation.

The only "evidence" advanced in support of the role of interparticle forces is the undisputed fact that homogeneous gas-fluidized beds exhibit less random particle motion than do liquid beds: gas-fluidized suspensions are more uniformly held together than are liquid-fluidized ones. This, however, can be well explained in terms of fluid-dynamic interactions.

Consider a homogeneously fluidized bed in equilibrium. If the particles are now subjected to a small force, they will move to restore the equilibrium condition. How fast they move will depend on the specific system properties: the greater the velocity of the particles, the more uniformly held together will be the suspension, and vice versa. A possible measure of this effect could be the parameter proposed by Batchelor (1988) in one of the 60 references quoted in the article to which this letter refers: the "*bulk-mobility B* of the particles." B is defined as "the ratio of the (small additional) mean velocity, relative to zero-volume-flux axes, to the (small additional) steady force applied to each particle of a homogeneous dispersion." B may be obtained from the fluidization model that delivered the predictions for ϵ_{mb} quoted above (Foscolo and Gibilaro, 1987)

$$B = \left.\frac{\partial u_p}{\partial f}\right|_{f=0} = \frac{1.25 n u_t \epsilon^{n-1}}{\pi d_p^3 (\rho_p - \rho_f) g}.$$

This relation suggests why it is that gas-fluidized suspensions are more uniformly held together than liquid-fluidized ones. If we consider a typical example of each (air fluidization of 80 μm alumina and water fluidization of 1 mm glass), we find B to be three orders of magnitude greater for the gas system. For the examples chosen by Marzocchella and Salatino (175 μm glass particles fluidized by water, CO_2 at 80 bar, and CO_2 at 1 bar), which exhibited increasing suspension uniformities with progression through this sequence, evaluated B values are found to increase by factors of 1, 11, and 67, respectively.

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Reply:

Gibilaro and Foscolo add to the content of the article by Marzocchella and Salatino (2000) by reporting a comparison between experimental values of bed voidage on the verge of bubbling (taken from Marzocchella and Salatino, 2000) and theoretical values calculated according to a hydrodynamic criterion for the stability of uniformly fluidized beds (Foscolo and Gibilaro, 1987). The comparison is admittedly poor: only in four out of the ten experimental conditions tested does the criterion predict the existence of a range of uniform expansion of the fluidized bed, contrary to the experimental findings.

Gibilaro and Foscolo re-open the debate on the relevance of interparticle forces to the behavior of gas fluidized bed, despite M. Baerns, L. Massimilla and coworkers, O. Molerus, K. Rietema and coworkers, R. Clift and coworkers, J. P. K. Seville and coworkers, R. Jackson and coworkers, among others who have provided experimental evidence of the role of interparticle forces in gas fluidization. The subject has been recently reviewed (Clift, 1993; Jackson, 1994; Seville et al., 2000) and authoritative opinions have been expressed in support of the view that interparticle forces are relevant to the particle phase stress tensor and, in turn, to the stability of the uniform state of expansion of gas fluidized beds of A-group solids.

The authors qualify as esoteric the hypothesis expressed by Marzocchella and Salatino that capillary condensation or extensive CO_2 adsorption can give rise to interparticle forces strong enough to influence the fluidization behavior under the conditions tested. The issue of the relevance of adsorption/condensation of gases/vapors to the establishment of interparticle forces is largely open to debate. At a high relative saturation, the formation of liquid bridges and its influence on interparticle forces is fairly well understood (Schubert, 1984) and there is a broad consensus on its relevance to fluidization (Massimilla and Donsì, 1976; D'Amore et al., 1979; Seville and Clift, 1984). On the contrary, the effects of vapor adsorption at low relative saturation pressure (in the subcritical state) or that of gas adsorption in the supercritical state are far less known. Coelho and Harnby (1978) and, more recently, Wensink et al. (2000) addressed the problem of powders bonding at low relative saturation pressure in the subcritical state, highlighting that strong solid-adsorbate affinity might induce pendular particle bridging even at low saturation. Results reported by Piepers et al. (1984) are fully consistent with Marzocchella and Salatino's hypothesis. These authors investigated beds of A-type solids and correlated the elasticity modulus of the emulsion phase and the cohesion constant of a Coulomb-type equation for the shear stress with the extent of gas adsorption. The latter was varied by changing operating pressure and type of fluidizing gas, while keeping temperature well above the critical temperature of the fluidizing medium. Results provide convincing evidence of the existence of pronounced effect of gas adsorption on interparticle forces even in the supercritical state.

As to the analysis of the variance of the heat-transfer coefficients, Gibilaro and Foscolo apparently miss Marzocchella and Salatino's argument. Marzocchella and Salatino state that: "...the variance of the heat-transfer coefficient is as negligible in the homogeneous fluidization regime as it is in the fixed-bed regime...," and **not** (as Gibilaro and Foscolo state) that: "...homogeneous gas fluidized beds exhibit less random particle motion than do liquid beds...." The finding (Figure 5 in Marzocchella and Salatino, 2000) that no change of the variance of the heat-transfer coefficient is detected on the verge of fluidization (whereas abrupt change is observed on the verge of bubbling) is interpreted by the authors as related to hindered mobility of particles in the homogeneously expanded state of the bed (see also Boerefijn et al., 2000). This is consistent with the direct observation of hindered particle mobility in homogeneously gas-fluidized beds reported by Donsì and Massimilla (1973) and by Seville et al. (2000).

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To the Editor:

In the article entitled "Multiple Steady States in Distillation: Effect of VL(L)E Inaccuracies" (May 2000) Bekiaris et al., emphasize the importance of reliable thermodynamic data. They mention in the abstract, "The accuracy of the thermodynamic prediction is a key factor that determines if multiplicities can be observed in numerical simulations," and in the conclusion that "extreme care needs to be taken towards the thermodynamic model and parameters used for the simulation of heterogeneous azeotropic distillation columns, because the existence of multiplicities is very sensitive to the thermodynamic description in many, or even most, cases." These statements can be confirmed explicitly, although from studies of different articles written by other authors, one gets the feeling that often not enough care is taken about the reliability of the g^E-model parameters used.

However, reliable phase equilibrium information covering the whole composition range as a function of temperature (pressure) is most important for the development of separation processes. Buford Smith mentions in the Henley and Seader book (1981) that, "The availability of large electronic computers has made possible the rigorous solution of the equilibrium-stage model for multicomponent, multistage distillation column to an exactness limited only by the accuracy of the phase-equilibrium and enthalpy data utilized."

The UNIQUAC equation used in the article is an adequate thermodynamic model for the calculation of phase equilibria. The authors mention accurately the critical role of reliable binary UNIQUAC interaction parameters, which should be fitted to consistent experimental data.

However, the required binary parameters should be derived in no case from questionable ternary VLE data, in particular when only a limited composition range is covered. (To stop readers of our VLE Data Collection from using binary parameters derived from ternary VLE data, in the new volumes of this series no binary g^E-model parameters have been given since 1987). They should at least be derived from thermodynamically consistent binary VLE data. However, with binary VLE data problems may occur, for example, in the diluted region, since typical binary VLE data usually only cover the composition range from 5–95 mol % and provide no sufficient information about the temperature dependence. Therefore, the most reliable binary g^E-model parameters are obtained by a simultaneous fit to all reliable binary phase equilibrium data (vapor-liquid equilibria, activity coefficients at infinite dilution, azeotropic data, liquid-liquid equilibria) and excess properties (excess enthalpies) (Rarey-Nies et al., 1989). Only reliable activity coefficients at infinite dilution provide the required information about the very dilute region. Excess enthalpies are the ideal properties for describing the temperature dependence of the nonideal behavior, because of the quantitative thermodynamic relation of the activity coefficients γ_i and the partial molar excess enthalpies h_i^E following the Gibbs-Helmholtz equation

$$\left(\frac{\partial \ln \gamma_i}{\partial 1/T}\right)_{P,x} = \frac{h_i^E}{R}$$

Since the existing modern g^E-models are not able to describe excess enthalpies and VLE with the help of constant parameters, temperature-dependent binary parameters must be used. Furthermore, the pure component vapor pressures measured by the authors must be used to describe precisely the excess Gibbs energy with the help of the fitted g^E-model parameters.

In the subheading "Some easy to make mistakes" Bekiaris et al. discuss the importance of the correct units of the UNIQUAC interaction parameters. The authors warn explicitly not to mix up the different units cal/mol, J/mol, and K which are commonly used in literature. Nevertheless, they make the same mistake with the parameter set of Gmehling and Onken (1977). The parameters given by Gmehling and Onken are given in cal/mol. However, the authors use exactly the same parameters, but with the wrong unit K; that means they do not divide the parameters given by Gmehling and Onken by the general gas constant R to obtain the unit K.

With the correct unit of the UNIQUAC parameters given by Gmehling and Onken (1977) derived from the VLE data of the ternary system, the phase equilibrium behavior of the ethanol-water-benzene system can be described at least qualitatively. In Tables 1–2 the calculated azeotropic data are given using the correct as well as the wrong unit for the UNIQUAC parameters. Furthermore, the experimental values are presented in Table 3. It can be seen that using the published parameters the calculated data are not far away from the experimental data published by different authors, although the parameters were derived from ternary VLE of an unknown quality covering only a very limited composition range. Therefore, the authors conclusion is erroneous and is based on the wrong unit

Table 1. Calculated Azeotropes Using the Correct Units for UNIQUAC Interaction Parameters at 1 atm

System	Azeotrope	Type
Ethanol-Benzene	y (Ethanol) = 0.4367	homogeneous
Ethanol-Water	y (Ethanol) = 0.8633	homogeneous
Benzene-Water	y (Benzene) = 0.6981	heterogeneous
Benzene-Ethanol-Water	y (Benzene) = 0.5333 y (Ethanol) = 0.2542	heterogeneous

Table 2. Azeotropes Given by the Authors Obtained with the Wrong Units at 1 atm

System	Azeotrope	Type
Ethanol-Benzene	y (Ethanol) = 0.6229	heterogeneous
Ethanol-Water	y (Ethanol) = 0.6983	homogeneous
Benzene-Water	y (Benzene) = 0.7042	heterogeneous
Benzene-Ethanol-Water	y (Benzene) = 0.5275 y (Ethanol) = 0.2525	heterogeneous

Table 3. Experimental Values Given by Gmehling et al. (1981) at 1 atm

System	Azeotrope (Interval of Mol Fraction)	Type
Ethanol-Benzene	y (Ethanol) = 0.438 – 0.464	homogeneous
Ethanol-Water	y (Ethanol) = 0.890 – 0.905	homogeneous
Benzene-Water	y(Benzene) = 0.700 – 0.705	heterogeneous
Benzene-Ethanol-Water	y(Benzene) = 0.5387 y(Ethanol) = 0.2281	heterogeneous

used for the interaction parameters. In order to get a reliable description of the VLE and the LLE behavior of ternary systems it is recommended using binary UNIQUAC parameters fitted as suggested before or at least to consistent binary data. Since consistent binary VLE data were used to fit the group interaction parameters of the UNIFAC method, it is not surprising that with the help of the UNIFAC method reliable UNIQUAC parameters are obtained.

Perhaps, this letter to the editor can help to improve this situation in the future.

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Reply:

We would like to thank Prof. Gmehling and coworkers for adding their expert advice on the proper use of thermodynamic models to the analysis of the literature in our article (Bekiaris et al., May 2000). It is indeed true that we used the wrong units for the parameter set of Gmehling and Onken (1977).

Because we did not have access to the original source at any library close by, we adopted the set by Prokopakis and Seider (1983) as stated in our article, p. 959: "In the multiplicities-related literature, this set of parameters was first reported by Prokopakis and Seider (1983b)." Apparently, this incorrect parameter set was also used by Rovaglio and Doherty (1990), and another error in unit conversion happened to Widagdo et al. (1989) (their parameters are off by R squared). They all report the erroneous ethanol-benzene immiscibility.

Thus, we need to add another recommendation to those given in our article: when using thermodynamic models always consult the original source.

We must emphasize, however, that neither the analysis nor the conclusions in our article are affected in any way by this error. In the comparisons with published articles we had to use the parameters of the respective authors. Most importantly, the key features of our multiplicity analysis techniques (infinity/infinity analysis) are totally independent of any thermodynamic model: experimental data are sufficient. In many cases, the existence or absence of multiplicities can be predicted solely based on the composition and type of the azeotrope.

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