To the Editor:

In reference to the article "Numerical Simulation of Mass Transfer between a Single Drop and an Ambient Flow" by Piarah et al. (July, 2001), we would like to present a different point of view. The authors state that in a previous publication (Henschke and Pfennig, 1999) we use an "extended mass-transfer [model] possessing a special type of turbulent circulation in the droplet [contradicting] the fact that due to all observations internal circulation in the droplet is laminar". However, in our model we assume that, hydrodynamically, only a laminar circulation in the droplet is induced. Additionally, in some cases instabilities can occur at the interface induced by mass transfer, which we address by an instability constant. They lead to turbulences close to the interface between the droplet and ambient phase. Contrary to the above statement, those turbulences have been observed, for example, by Schombacher and Bauckhage (1997). They employed rainbow refractrometry to measure concentrations within drops and observed some random noise for high concentrations at the beginning of the mass-transfer process, which they attribute to Marangoni instabilities at the interface leading to a small-scale convection.

Just like Piarah et al. (2001) we examined if two-dimensional (2-D) axisymmetric numerical calculations can be used to reproduce the experimental results. These calculations have been performed with SEPRAN (Sepran, 1998). The outcome can be seen in Figure 1. Our experimental data are presented here together with the analytical solution of Kronig and Brink (1950) and two numerical solutions. Furthermore, the correct partition coefficient in terms of concentrations is given. When using a coarse grid with a finer subdivision at the interface between droplet and continuous phase, the experimental data can well be obtained. However, if the grid is evenly subdivided and the calculation thus becomes more precise, the results approach the analytical solution of Kronig and Brink (1950). Fitting numerical data to experimental, as was done by Piarah et al. (2001), can, therefore, only be an indication of the correctness of the numerical method used, but not a proof or validation. Eigenberger and Sokolichin (1997) experienced similar problems when numerically calculating a bubble column without modeling the turbulent behavior. The numerical errors compensated the errors of their numerical model, which was fortunately realized by the authors. Waheed (2001) used different numbers of nodal points to check the influence on the outcome of the calculations.

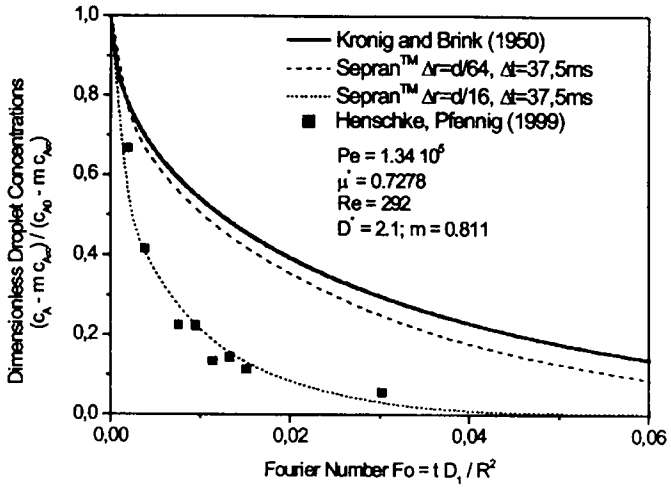

Figure 1. Average concentration in the droplet as a function of Fourier number.

Numerical diffusion is the effect that led to the above-mentioned deviations between calculations with a dense grid and calculations with a coarse grid. It occurs especially when the convective term of the equation for the conservation of mass is larger than the diffusive term, and when the used grid is coarse. Unfortunately, detailed tests showed that an appropriate grid for exactly calculating mass transfer in the droplet is very dense resulting in inappropriate computing times.

However, there are good reasons why the exact result should come very close to the analytical solution of Kronig and Brink (1950). In Figure 2 the flow pattern used by Kronig and Brink (1950) can be seen on the left hand side, and the flow pattern resulting from a simulation at a Reynolds Number of 283, which is typical for this problem, can be seen on the right hand side. A comparison shows that both almost coincide. The diffusional distances to be overcome by the transferred component are, thus, approximately identical. Since Kronig and Brink (1950) assume a constant concentration along the streamlines, the analytical result obtained should represent the maximum rate of mass transfer. We, therefore, expect that 2-D calculations with an appropriately dense grid should lead to a result very close to the analytical solution of Kronig and Brink (1950).

The introduction of a partly empirical equation, that is, the introduction of an instability constant, like in Henschke and Pfennig (1999), is one way to appropriately describe the problem. This accounts for possible 3-D effects and a concentration-dependent local interfa-

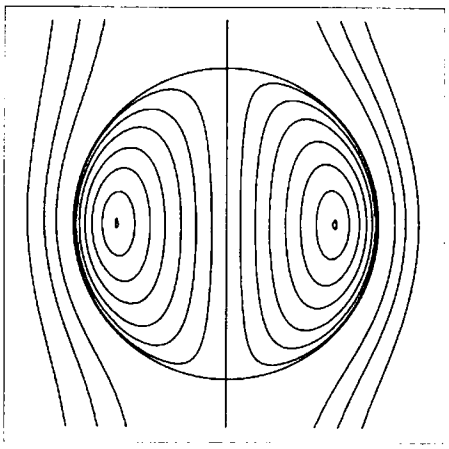

Figure 2. Comparison of streamlines of the analytical solution of Kronig and Brink (1950) (left hand side) with those computed with SEPRAN at *Re* = 292 (righthand side) for a rising drop.

cial tension leading to mass-transfer induced convection, and other interfacial instabilities as described by Sawistowski (1971).

Literature cited

Eigenberger, G., and A. Sokolichin, "Modellierung und effiziente numerische Simulation von Gas-Flüssigkeits-Reaktoren mit Blasenströmungen nach dem Euler-Euler-Konzept," DFG-Zwischenbericht Ei 131/10-2, Schwerpunktprogramm "Analyse, Modellierung und Berechnung mehrphasiger Strömungen," (http://pcvt12.icvt.uni-stuttgart.de/RESEARCH/GL/bericht_97h/bericht_97h.html) (1997).

Henschke, M., and A. Pfennig, "Mass-Transfer Enhancement in Single-Drop Extraction Experiments," *AIChE J.* 45(10), 2079 (1999).

Kronig, R., and J. C. Brink, "On the Theory of Extraction from Falling Droplets," *Appl. Sci. Res.*, A2, 142 (1950).

Piarah, W. H., A. Paschedag, and M. Kraume, "Numerical Simulation of Mass Transfer between a Single Drop and an Ambient Flow," *AIChE J.*, 47(7), 1701 (2001).

Sawistowski, H., "Interfacial Phenomena," *Recent Advances in Liquid-Liquid Extraction*, C. Hanson, ed., Pergamon Press, Oxford, pp. 293-366 (1971).

Schombacher, E. H., and K. Bauckhage, "Bestimmung von Konzentrationen und Konzentrationsgradienten an Tropfen in ternären Flüssig/Flüssig-Systemen mittels der Regenbogenrefraktometrie," *Chemische Technik*, 49, 125 (1997).

Sepran "A Finite Element Code," Ingenieurbüro Sepra, Park Nabij 3, 2267 Ax Leidschendam, The Netherlands (1998).

Waheed, M. A., "Fluiddynamik und Stoffaustausch bei freier und erzwungener Konvektion umströmter Tropfen," Diss., RWTH Aachen (2001).

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Reply:

In response to the letter from Gross-Hardt et al. concerning our article, we would like to clarify our approach including a discussion of its accuracy:

(1) As mentioned by Gross-Hardt et al., secondary flow structures appear close to the liquid/liquid interface in a couple of chemical systems within a certain concentration range. These structures, which are caused by the so-called Marangoni convection, differ from turbulence in two aspects: they are located in the vicinity of the interface, and they are defined and have no stochastic flow pattern. As a consequence, the introduction of an additional turbulent transfer coefficient (Henschke and Pfennig, 1999), that accounts for mass-transfer-induced-turbulence in analogy to the turbulent diffusivity in RANS turbulence modeling, does not seem appropriate for reasonable modeling. However, a convection phenomenon as the source of the increased mass transfer is beyond doubt for Gross-Hardt et al., as it is for us. We are thoroughly convinced that model improvement should begin with an extension in the momentum transport model and end with an improved mass transfer based on that (as, by the way, is also done in turbulence modeling).

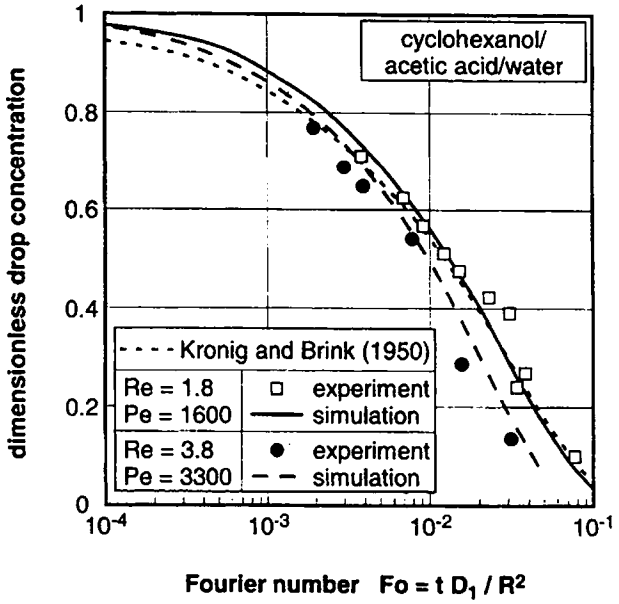

Figure 1. Dimensionless concentration in drops as a function of Fourier number.

Comparison of experimental and numerical results.

The approach of our current studies is the introduction of convective flow caused by changes of local interfacial tension due to concentration fluctuations in our numerical model. Hereby, we hope to describe Marangoni convection structures accurately and to find an improved mass transfer based on that.

(2) Contrary to Henschke and Pfennig (1999), we did no fitting of numerical data to experimental results. Our goal was basically to determine the general accuracy of numerical methods applied to mass transfer around drops. The numerical procedure that we use has been checked in regard to grid spacing as well as in regard to a discretization scheme. As can be seen from Figure 1 in the original article, grid spacing is in the range that the authors require. The special attention drawn to the subspacing at the interface is also shown there. Use of grids with a doubled number of points (two different configurations tested) has no influence on the numerical result. In contrast, application of discretization schemes of a higher order in space and time have an effect in the way that the authors mentioned (Piarah, 2001, Fig. 5.51).

Our experience shows that is generally difficult to obtain satisfying numerical results for $Pe > 10^5$, since the numerical error of nonoscillating schemes gets larger the more convection dominates diffusion. Explicit coupling between balance equations inside both phases and interfacial conditions makes things even worse.

(3) To validate our simulations, we considered among others the system water/acetic acid/cyclohexanol. For the concentration range investigated in our mass-transfer experiments, we showed the absence of Marangoni convection studying the droplet rise velocity. Reynolds numbers chosen are in the range of 2 . . . 4, and Peclet numbers vary between 1,600 . . . 3,300. Figure 1 shows good agreement between numerical and experimental results for that case. Encouraged by these results, we stress our further activities on the correct modeling of physical phenomena rather than on further improvement of our numerical method.

(4) In general we agree on the discussion regarding the solution derived by Kronig and Brink (1950). However, the experimental results that we obtained for the water/acetic acid/cyclohexanol system show a distinct devia-

tion from the curve of Kronig and Brink for $Re = 3.8$. As their approach is valid solely for creeping flow, this is not totally surprising. Figure 3 in our original article shows a weak, but significant, influence of the Reynolds number on the mass transfer for the internal and the conjugate problem. It can be seen that, for $Re < 100$, a distinct speedup of the mass transfer can be expected with increasing Re. It should also be mentioned that the velocity field derived by Hadamard on which the Kronig and Brink solution is based, as well as the derivation of Kronig and Brink, contain some additional approximations which can cause certain small differences between their solution and a numerical solution of the complete system of equations for the same case.

Literature cited

Henschke, M., and A. Pfennig, "Mass-Transfer Enhancement in Single-Drop Extraction Experiments," *AIChE J.*, **45**(10), 2079 (1999).

Kronig, R., and J. C. Brink, "On the Theory of Extraction from Falling Droplets," *Appl. Sc. Res.*, **A2**, 142 (1950).

Piarah, W. H., "Numerische Untersuchung des instationären Stofftransports an und in umströmten Einzeltropfen," Diss., Technische Universität Berlin (2001).

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To the Editor:

In "Steady-State and Decay Dynamics for Impellers of Varying Aspect Ratio in Unbaffled Tanks" (January 2002) Maynes and Butcher report on the experimental determination of the power input of a bluff body (square impeller) rotated in the cylindrical tanks without internals (such as baffles), but provided with a lid. Such equipment is important as a chemical reactor (Rousseaux et al., 2001) where absence of internals could enhance its performance and avoid an air entrapment from the liquid surface. Steady-state hydrodynamics in the above mentioned system under a turbulent regime of flow can be described as means of a simple one-dimensional (1-D) axially symmetrical flow (Medek and Fořt, 1994; Taca and Paunescu, 2001) consisting of two parts: the inner one with a constant angular velocity (a forced vortex) and the outer one with decreasing angular liquid velocity with increasing radius-free vortex. The impeller power input then is dissipated in the turbulent boundary layer at the vessel wall, bottom, and lid. In accordance with this idea the peripheral (tangential) component of the shear stress in the above mentioned boundaries of the system is proportional to the Re^{-n} where $n = 1/5$, and the same power dependence is valid for the relation between the torque coefficient C_m (or impeller power number) and Reynolds number modified for the impeller. The results of Maynes and Butcher's article correspond fairly well to the above mentioned model when the geometry of the investigated impeller (its diameter and height) corresponds to the geometry of a standard high speed impeller. The value of the exponent n at the Reynolds number is lying within the interval $n \in \langle 0.15;\ 0.30 \rangle$ in dependence on the geometry of the system. A larger deviation from this interval appears when the impeller/tank diameter ratio L^* exceeds the value 0.5 (probably due to interference between the flow around impeller and the wall) and when the impeller/tank height ratio is higher than 3.0 (probably due to interference between the flow around impeller and bottom and lid).

Maynes and Butcher mentioned some results of an investigation dealing with the velocity field in the system with a square impeller. It seems to be rather useful if they could calculate under steady-state conditions the parameters of the radial profile of the mean tangential (or peripheral) velocity in dependence on geometry of the system. Such results can help their followers (chemical engineers) in the calculation of mixing and micromixing processes in the above described newly designed system.

Literature cited

Medek, J., and I. Fořt, "Liquid Circulation in Mechanically Agitated Closed Vessels," *Proc. of 8th Eur. Conf. on Mixing*, Cambridge, U.K., 473 (1994).

Rousseaux, J.-M., H. Muhr, and E. Plasari, "Mixing and Micromixing Times in the Forced Vortex Region in Unbaffled Mixing Devices," *Can. J. Chem. Eng.*, **79**, 697 (2002).

Taca, C. D., and M. Paunescu, "Power Input in Closed Stirred Vessels," *Chem. Eng. Sci.*, **56**, 4445 (2001).

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