To the Editor:

We would like to comment on a recent article by Sikdar and Randolph [*AIChE J.*, **22**, 110, (1976)] related to interpretation of non-linear population density plots for magnesium sulfate and citric acid crystals obtained in a seeded CMSMPR crystallizer. The authors attributed deviations from straight-line CSD's on semilog plots at sizes less than 8 μm to the instability of secondary nuclei and/or a grossly lower crystal growth rate below 8 μm. However, they stated that such a dramatic decrease in growth rate is not readily explained by current growth rate theories.

This letter is intended to call attention to a model, proposed by Wey and Estrin [AIChE Symposium Series No. 121, **68**, 74 (1972)], which takes into account the surface curvature effect (Gibbs—Thomson effect) on the growth rate of secondary nuclei. Their model clearly shows a dramatic decrease in growth rate for small crystals in the stable nucleus size range. Having a higher surface curvature, crystals in this range are more soluble than larger crystals. Thus, for a given supersaturated environment, the driving force for growth of a crystal (and, hence, growth rate) decreases with a decrease in size.

Wey and Terwilliger [*AIChE J.*, **20**, 1219 (1974)] theoretically examined the potential influence of crystal surface curvature on the CSD behavior in a CMSMPR crystallizer. Incorporation of the curvature effect into a size-independent growth model (McCabe's ΔL law) yielded a nonlinear population density plot in the stable nucleus size region. Small particles were shown by the model to exhibit significantly higher population densities than expected by semilog extrapolation from the larger sizes. The concave-upward nature of the resulting distribution is essentially identical to that reported by Sikdar and Randolph (1976).

Sikdar and Randolph (1976) suggested that a large majority of initially formed nuclei never survive to populate the large sizes. This consideration seems to be supported by the large RPM dependence observed for sizes less than 8μm. One possible explanation for the nuclei instability would be that some of the nuclei generated by secondary nucleation are smaller than the stable nucleus size predicted by the Gibbs-Thomson equation. If this is the case, the smallest surviving crystals would be in a size range in which surface curvature should undoubtedly have an important influence on the growth-rate behavior (Wey and Estrin, 1972).

It is suggested that the effects of crystal surface curvature be considered in explaining the non-linear population density plots observed in the small size region for magnesium sulfate and citric acid systems. This consideration is especially important if the smallest observable and measured crystals are in the stable nucleus size range.

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Reply

It is quite logical to expect that the size dependent solubility (Gibbs-Thomson effect) of crystal nuclei would result in growth rates decreasing with the size of nuclei of secondary origin. Wey and Terwilliger's CSD model [*AIChE J.*, **20**, 1219, (1974)] indeed predicts nonlinear log-population density versus size plots that are qualitatively similar to the quasi-steady state CSD's obtained for magnesium sulfate and citric acid systems reported by Sikdar and Randolph [*AIChE J.*, **22**, 110 (1976)]. The instability of a large majority of secondary nuclei having sizes less than 8 μm as observed by us might be explained by the Gibbs-Thomson effect provided the critical size of nuclei is large, i.e., above one micron in size. This is a tempting and perhaps correct explanation of these data.

However, realistic physical-chemical properties used in the Gibbs-Thomson equation do not predict such large critical sizes. This has led others to question this explanation of the slow nuclei growth rates. For example, Garabedian and Strickland-Constable ["Collision Breeding of Crystal Nuclei: Sodium Chlorate. I," *J. Crystal Growth*, **13/14**, 506 (1972)] forwarded the theory of nuclei perfection in explaining the suppressed growth of small nuclei. This theory emanated from the failure to explain slow growth and/or nuclei instability from the Gibbs-Thomson equation. Surface curvature may be an important factor but an *a priori* quantitative explanation is still lacking. For this reason we did not advance an explanation of, or even unequivocally state the existence of, the large population of particles < 8 μm. The main thrust of the paper was to demonstrate that net effective nucleation kinetics could be obtained with fast growth systems in the particular Mini-nucleator apparatus.

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To the Editor:

The R&D Note by Valdman and Hughes [*AIChE J.*, **22**, 192 (1976)] provides an interesting demonstration of one-point orthogonal collocation as applied to catalyst effectiveness factors. The proposed method, however, was published previously in this *Journal* by Stewart and Villadsen [*AIChE J.*, **15**, 28 (1969)]. This prior article and that of Sorensen *et al.* [*AIChE J.*, **19**, 969 (1973)] describe the method fully and apply it to various reaction rate functions and particle shapes.

Stewart and Villadsen (1969) demonstrated their method graphically, for expository reasons. However, direct numerical solutions can be obtained by specifying the state at the collocation point and solving Equations (16) and (17) of that paper for the effectiveness factor and particle size. This procedure is included in the article by Sorensen, *et al.* along with higher-order collocation solutions.

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ERRATA

In "A Model for Predicting Flow Regime Transition in Horizontal and Near Horizontal Gas Liquid Flow" by Yehuda Taitel and A. E. Dukler [*AIChE J.*, **22**, 47 (1976)]:

1. The solution of Equation (7) was plotted with incorrect parameters in Figure 2. The corrected version of this figure appears below. Equation (7) is correct.
2. The correct statement of Equation (25) is

$$F^2\left[\frac{1}{C_2^2}\,\tilde{u}_G^{\,2}\,\frac{d\tilde{A}_L/d\tilde{h}_L}{\tilde{A}_G}\right] \geq 1$$

The solution of this equation which appears in Figure 4 is based on the correct equation.

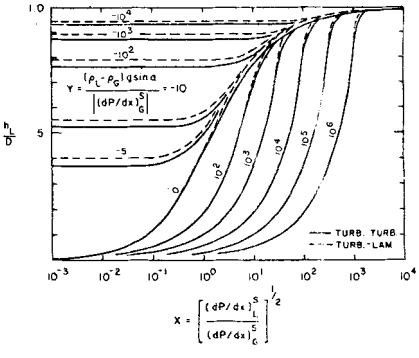

Fig. 2. Equilibrium liquid level for stratified flow (turbulent liquid, turbulent or laminar gas).