k_o = second-order Arrhenius frequency factor, cm³/gmol · s
k_l = liquid mass transfer coefficient, cm/s
M = Thiele modulus or Hatta number, defined by Eq. 10
R = universal gas constant, cal/gmol · °K
R_A = reaction rate, gmol/cm³ · s
S = reactor cooling area, cm²
T = temperature, °K
U = overall heat transfer coefficient, cal/°K cm² · s
V_l = volume of liquid phase, $\epsilon_l V_R$, cm³
V_R = reactor volume, cm³
x_l = conversion of liquid reactant, $(B_{lf} - B_l)/B_{lf}$
y_{lb}, y_{ub} = dimensionless temperature bounds, defined by Eq. 23

Greek Letters

α' = dimensionless parameter, defined by Eq. 11
θ' = dimensionless parameter, defined by Eq. 11
ρ = molar density, gmol/cm³
ϵ_l = fractional liquid holdup in the reactor
τ_l = liquid residence time, V_l/F_l, s

Subscripts

c = cooling medium
f = feed
g = gas phase
i = gas-liquid interface
l = liquid phase

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II: Discrimination among Rival Reaction Models

The discrimination of the fast second-order, pseudo-first-order and fast pseudo-first-order reaction models on the basis of the second-order reaction model justifies the use of the pseudo-first-order model in the prediction of the multiplicity regions. The use of the fast second-order reaction model leads to a certain pitfall regarding uniqueness and multiplicity. Finally, the use of the fast pseudo-first-order reaction model is justifiable not only because of the fair agreement between the multiplicity regions predicted by this model and the second-order model but also because of the analyticity of the uniqueness and multiplicity regions obtained by this model.

SCOPE

In the operation of gas-liquid CSTRs (continuous stirred tank reactors), the occurrence of multiple steady states has been observed by Ding et al. (1974) for the chlorination of *n*-decane. Hoffman et al. (1975) and Sharma et al. (1976) have shown numerically the possible existence of five and seven steady states for single and consecutive second-order reactions respectively occurring in gas-liquid CSTRs. These have prompted development of *a priori* criteria providing conditions among physico-chemical parameters which assure unique and multiple steady states.

Raghuram and Shah (1977) derived analytic criteria, which they claimed to represent conditions assuring uniqueness of the steady state, and also presented various plots to determine the number of steady states in parameter space for the case of pseudo-first-order reactions (i.e., the conversion of the liquid reactant is assumed zero) and for the case of second-order reactions in the "fast" reaction regime, respectively. However, types of multiplicity patterns were not analyzed for either case.

We have recently (Huang and Varma 1981a) given *analytic* necessary and sufficient conditions for uniqueness, multiplicity and stability of the steady state, and also full view of the multiplicity regions and patterns under various operating conditions for the specific case of pseudo-first-order reactions in the "fast" reaction regime.

When multiple steady states exist, the steady state on the high temperature branch is generally in the "fast" reaction regime and the conversion of the liquid reactant is significant. On the other hand, the steady state on the low temperature branch is usually in the "slow" reaction regime. Therefore, applying any of the reaction models with the simplifying assumptions as mentioned above cannot cover the entire range of the multiplicity phenomenon and may lead to pitfalls. This raises the need to discriminate among various reaction models.

The main goal of this communication is to compare previously reported reaction models in a hope to justify some of the information obtained from these simplified models. The results of a fully second-order reaction model from Part I of this work (Huang and Varma, 1981b) are applied for this purpose. Types of multiplicity patterns for the cases of pseudo-first-order reactions and fast second-order reactions are also analyzed.

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CONCLUSIONS AND SIGNIFICANCE

The fast second-order, pseudo-first-order and fast pseudo-first-order reaction models are analyzed and compared with the second-order reaction model, respectively. The fast second-order reaction model predicts the same multiplicity patterns as those of the second-order model with the physicochemical parameters are reported in Table 1 of Part I of this work. However, as the activation energy decreases from 29000 to 20000 cal/gmol, this model predicts uniqueness in the parameter space of Da versus τ_ℓ, while the other two simplified models and the second-order model predict multiplicity. This can be a serious pitfall in applying the fast second-order reaction model.

The pseudo-first-order reaction model leads to multiplicity *regions* that agree surprisingly well with the predictions of the second-order model. The multiplicity *patterns* predicted by these two models, however, differ significantly. If the pseudo-first-order model stands by itself, three new multiplicity patterns are possible, viz., a S-shaped plus persistent multiplicity, a "double persistent multiplicity" with a two-step extinction, and a "double persistent multiplicity" with only one-step extinction, as shown in Figures 4, 6a and 6b respectively.

The fast pseudo-first-order reaction model is shown to be better than the fast second-order reaction model in the prediction of the multiplicity *regions*. No *a priori* criteria for uniqueness and multiplicity have been derived from any reaction model other than the fast pseudo-first-order reaction model.

The level of numerical trial-and-error effort required decreases dramatically as one goes progressively from the full second-order model to the pseudo-first-order model. The simplest model with fast pseudo-first-order kinetics leads to *analytic* criteria, requiring *no* trial-and-error.

ANALYSIS

Before describing the rival reaction models, it should be mentioned that the basic assumptions are the same as those described in Part I of this work (Huang and Varma, 1980). The material balance for the gaseous reactant and the overall energy balance are the same as Eqs. 2 and 4 in Part I, respectively. However, the material balance for the liquid reactant may be the same as Eq. 3 in Part I or completely dropped out depending on the reaction model.

Second-Order Reaction Model (Model 2)

This reaction model is exactly the same as that reported in Part I of this work. The reaction factor E^*_A and the concentration of the gaseous reactant in the bulk liquid A_l are obtained from Eqs. 6-11 in Part I. Since a second-order reaction scheme has been considered without any limitation on the reaction regime, this model is taken as the basis to judge the following simplified reaction models.

Fast Second-Order Reaction Model (Model 2f)

This reaction model assumes that the second-order reaction is in the "fast" reaction regime (i.e., $M >> 1$). This model was applied by Raghuram and Shah (1977) to derive analytic criterion assuring uniqueness of the steady state in an adiabatic gas-liquid CSTR, where the pseudo first order kinetics in the "liquid film" (i.e., $B_i = B_l$) were also applied. Note that the pseudo first order kinetics is used *only* to evaluate E^*_A and A_l, and should not be confused with the pseudo-first-order reaction model (assuming $B_l = B_{lf}$) to follow next. To clarify this point, note that the film theory is applied in the evaluation of E^*_A and A_l (cf. Hoffman et al., 1975), and the "liquid film" is theoretically proposed only in the film theory (cf. Danckwerts, 1970). Further, from the van Krevelen-Hoftijzer plot (cf. Carberry, 1976), it is clear that when the ratio B_l/A_i is large, there is no difference between the values of E^*_A aplying the second order and pseudo first order kinetics. Nevertheless, while the pseudo first order kinetics may be applied to evaluate E^*_A and A_l, the conversion of the liquid reactant may not be zero in the *reactor* and second-order reaction model should then be applied. We see that the use of the second-order model is reflected by including the material balance of the liquid reactant, as Eq. 3 in Part I.

As reported in Part I, the pseudo-first-order kinetics can usually be applied to evaluate E^*_A and A_l. With $B_i = B_l$ (i.e., $b_i = 1$), Eqs. 6 and 7 in Part I become

$$E^*_A = M \frac{M^2(\alpha' - 1) + \frac{1}{\theta'} + M \tanh M}{\left[M^2(\alpha' - 1) + \frac{1}{\theta'}\right] \tanh M + M} \tag{1}$$

$$A_l = A_i \frac{M}{M \cosh M + \left[M^2(\alpha' - 1) + \frac{1}{\theta'}\right] \sinh M} \tag{2}$$

These are exactly the same as those derived directly from the pseudo-first-order kinetics (cf. Raghuram and Shah, 1977). It is believed that the use of the pseudo first order kinetics itself does not make any difference in the results presented throughout this work.

With $M >> 1$ for "fast" reaction, Eqs. 1 and 2 reduce to:

$$E^*_A = M \; ; \; A_l = 0 \tag{3}$$

respectively. These are the *only* assumptions made in this reaction model.

Pseudo-First-Order Reaction Model (Model 1)

This reaction model is a simplification of the second-order reaction model. It assumes that the conversion of the liquid reactant is zero (i.e., $B_l = B_{lf}$). Thus, the material balance for the liquid reactant is dropped and only Eqs. 2 and 4 of Part I are used in this model. E^*_A and A_l are computed from Eqs. 1 and 2, respectively; however, assuming $B_l = B_{lf}$ does not necessarily mean the applicability of the pseudo first order kinetics to evaluate E^*_A and A_l. This model has been used by Raghuram and Shah (1977) to derive analytic criterion for uniqueness of the steady state in an adiabatic gas-liquid CSTR.

Fast Pseudo-First-Order Reaction Model (Model 1f)

This reaction model assumes that the pseudo-first-order reaction is in the "fast" reaction regime. The assumptions are $B_l = B_{lf}$ plus those as in Eq. 3. This model was applied recently (Huang and Varma, 1981a) to provide analytic necessary and sufficient conditions for uniqueness and multiplicity and a complete classification of the steady state behavior.

As shown before (Huang and Varma, 1981a), the necessary condition for multiplicity is equivalent to the following condition in terms of the liquid flow rate, for the case of a fixed gas feed rate:

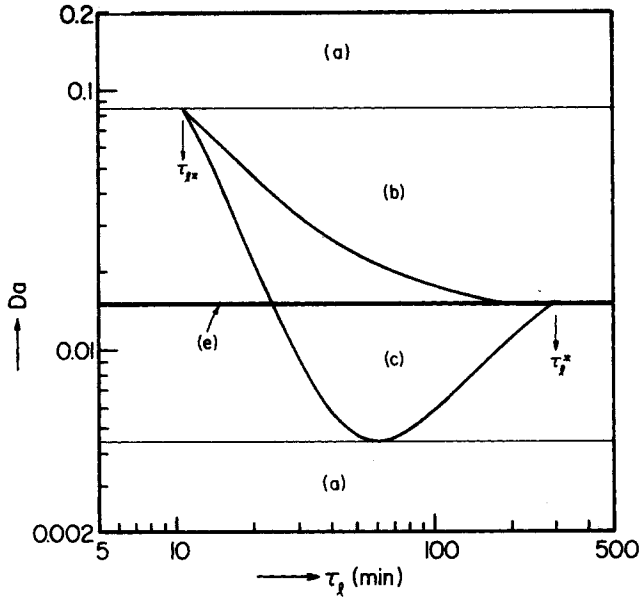

Figure 1. Effect of reactivity on the multiplicity regions and patterns: model 2f; (a) uniqueness, (b) S-shaped, (c) isola, (e) mushrooms.

$$F_{l1} < F_l < F_{l2} \tag{4}$$

where F_{l1} and F_{l2} are computed from explicit expressions. It can be easily shown that $F_{l2} = V_l/\tau_{l'}$, where $\tau_{l'}$ has the same meaning as that shown in Figure 6 of Part I. For this model, F_{l1} is usually negative. However, for model 2, F_{l1} ($\equiv V_l/\tau_{l'}$) is positive though generally very small. We see that a comparison of model 2 and this model on a plot like that of Figure 10 in Part I can be used to judge the validity of this necessary condition.

The sufficient condition for multiplicity is (Huang and Varma, 1981a):

$$Da_2 < Da < Da_1 \tag{5}$$

where Da_1 and Da_2 are computed from explicit expressions. Thus, in the parameter space of Da vs. τ_l, the multiplicity region is determined analytically. A comparison of this model and model 2 in such a parameter space, as that of Figure 6 in Part I, can be used to judge the applicability of this sufficient condition for multiplicity in a gas-liquid CSTR with second-order reactions.

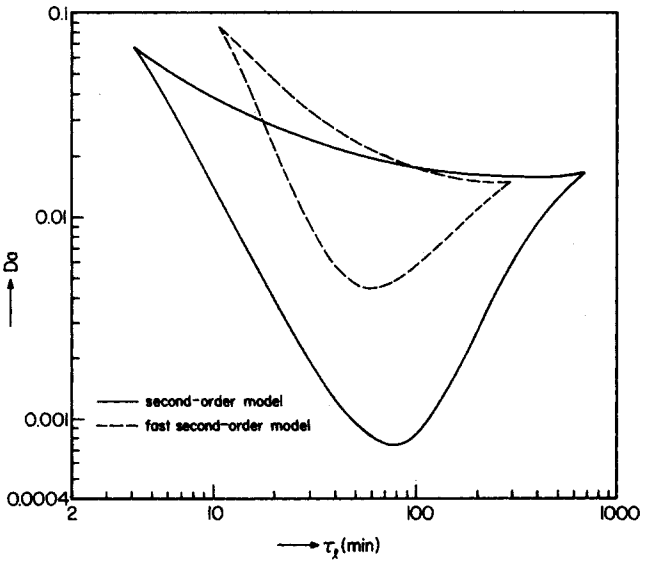

Figure 3. Comparison of the multiplicity regions of models 2 and 2f.

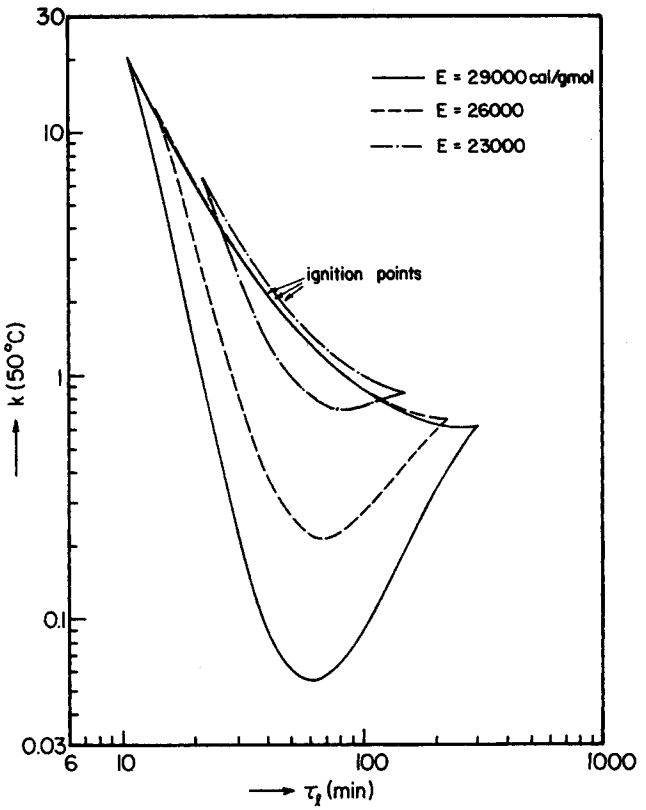

Figure 2. Effect of activation energy on the multiplicity regions: model 2f.

RESULTS AND DISCUSSION

As in Part I of this work, the case of a fixed gas feed rate is considered here. All the physico-chemical parameters are as reported in Table 1 of Part I except when otherwise specified. These parameters are believed capable of describing some real systems and can thus be used to make the following comparisons.

Some results of Part I are used to make the comparisons. As shown in Figure 9 of Part I, when the activation energy decreases, the multiplicity region shrinks with respect to *both* the reactivity and the liquid residence time; also note that at E = 20 000 cal/gmol, a sizable multiplicity region exists. These features will be examined in the following comparisons. Also, Figures 6 and 10 of Part I will be reproduced to make various comparisons.

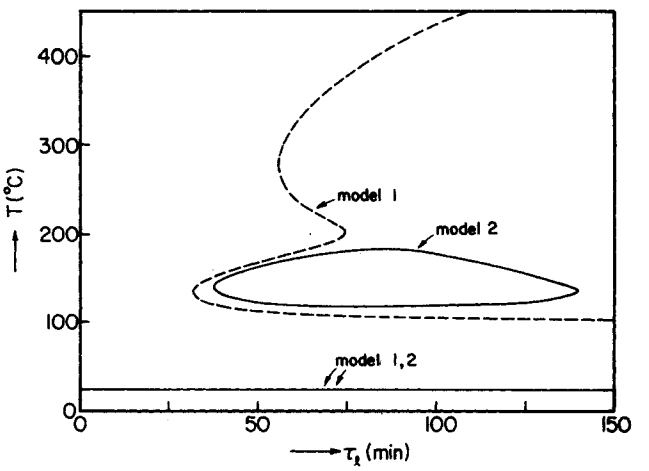

Figure 4. Comparison of the predictions of models 1 and 2.

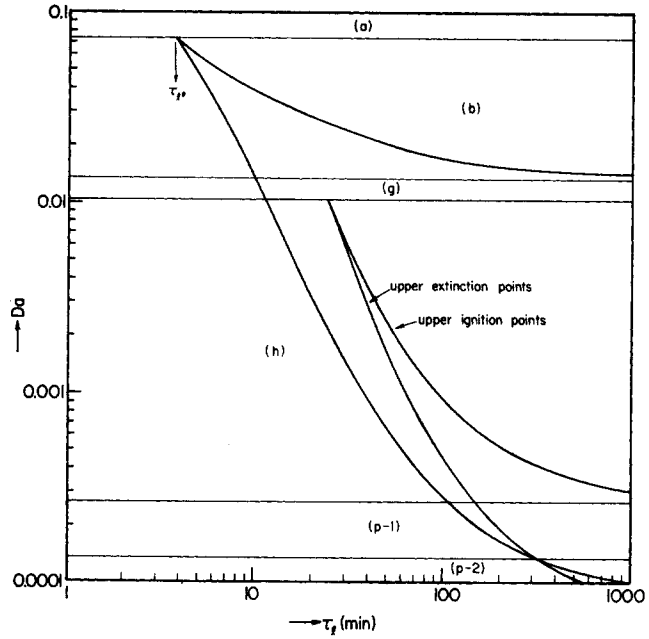

Figure 5a. Effect of reactivity on the multiplicity regions and patterns: model 1; (a) uniqueness, (b) S-shaped, (g) persistent multiplicity, (h) S-shaped + persistent multiplicity, (p-1) and (p-2) double persistent multiplicity.

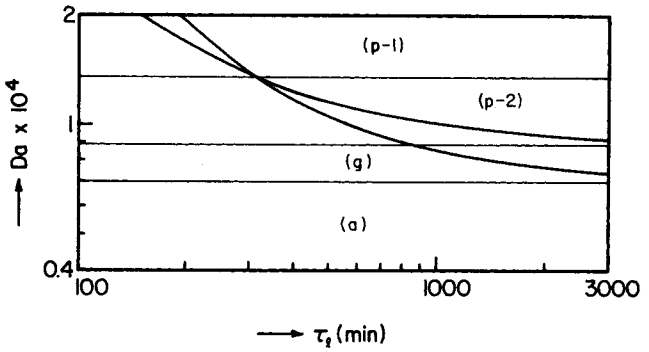

Figure 5b. The regions of "double persistent multiplicity."

Comparison of Models 2 and 2f

In Figure 1 of Part I, experimental results showing multiple steady states were presented and compared with prediction of model 2. For exactly the same parameters, model 2f predicts uniqueness. As shown in Figure 1, the multiplicity *patterns* predicted by model 2f, however, are the same as those predicted by model 2.

Figure 2 shows the effect of activation energy E on the multiplicity regions for model 2f, where the region for E = 29 000 cal/gmol is the same as that in Figure 1 but with k (50°C), in $cm^3/gmol \cdot s$, explicitly shown. The general features, e.g., the multiplicity region shrinks as E decreases, are the same as those of model 2 except that the ignition points go to higher Da and τ_l values, in contradiction to model 2. Further, no multiplicity region is found at E = 20 000 cal/gmol for model 2f, although a sizable multiplicity region exists for model 2 for this activation energy. This can be a serious pitfall in applying model 2f.

Figure 3 compares model 2f with model 2 in the parameter space of Da vs. τ_l. It is seen that the multiplicity region predicted by model 2f is smaller than that by model 2.

Comparison of Models 1 and 2

The predictions of models 1 and 2 for the simulation of the experimental results are presented in Figure 4. Good agreement is seen except that model 1 predicts two additional higher temperature branches.

Since the multiplicity patterns for model 1 have not been analyzed before, they are now identified. As shown in Figures 5a and 5b, five different multiplicity patterns are predicted by model 1, where (a) denotes uniqueness for all values of τ_l, (b) denotes S-shaped multiplicity, (g) persistent multiplicity as described before (Huang and Varma, 1981a), (h) S-shaped plus persistent multiplicity as shown in Figure 4 (model 2), and (p-1) and (p-2) denote two different types of "double persistent multiplicity". For $\tau_l < \tau_{l'}$, uniqueness is guaranteed for all values of Da. Figure 6a presents an example of "double persistent multiplicity" in region (p-1), where the upper extinction point is at higher τ_l value than the lower one, and so the extinction may occur in two steps. Figure 6b presents an example of "double persistent multiplicity" in region (p-2), where the upper extinction point is at lower τ_l value than the lower one, and so extinction occurs in one step only.

From Figure 7, as E decreases, the multiplicity region of model 1 shrinks in the same manner as that of model 2f. However, a sizable multiplicity region remains at E = 20 000 cal/

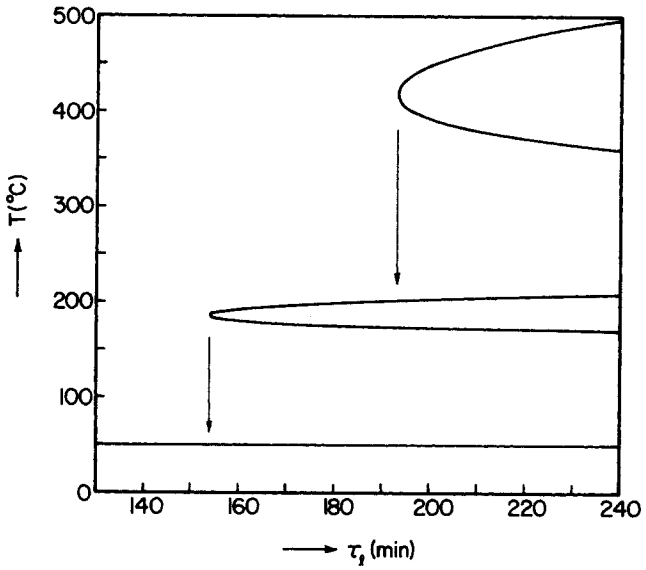

Figure 6a. An example of "double persistent multiplicity" in region (p-1); $k = 1.1 \times 10^{-4}$ ($Da = 2 \times 10^{-4}$).

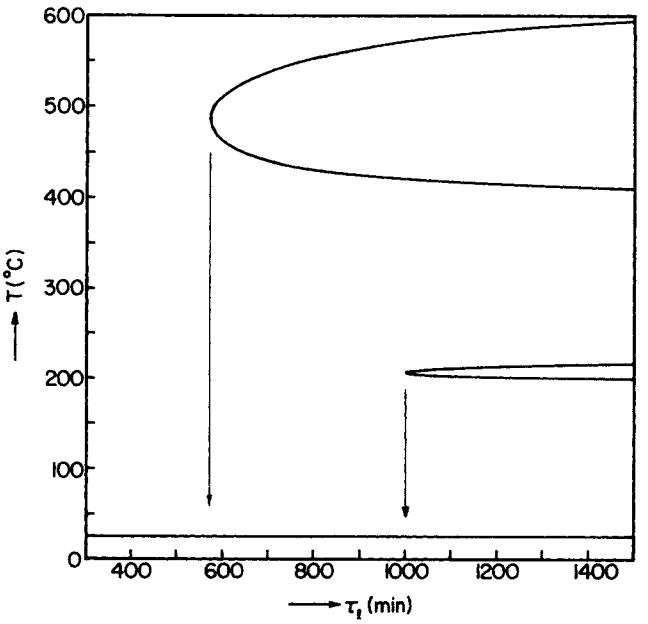

Figure 6b. An example of "double persistent multiplicity" in region (p-2); $k = 2.8 \times 10^{-5}$ ($Da = 10^{-4}$).

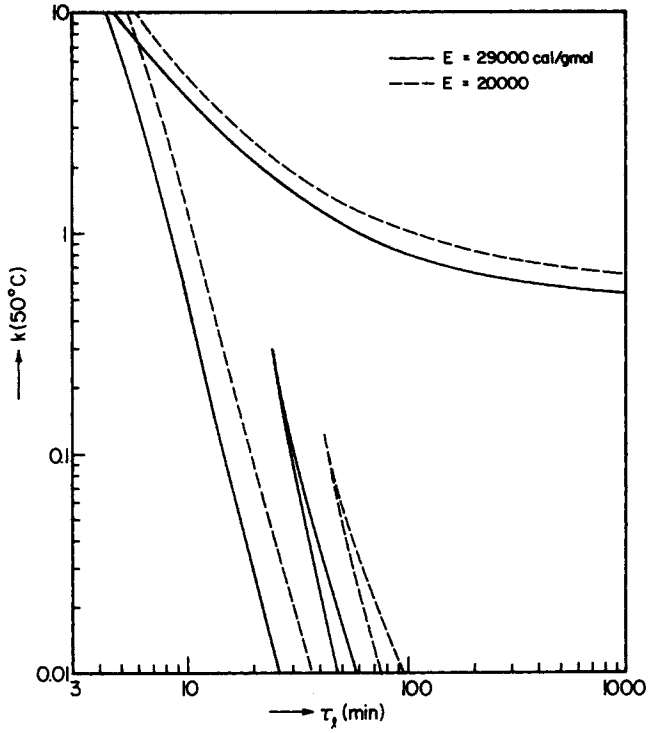

Figure 7. Effect of activation energy on the multiplicity regions: model 1.

gmol for model 1. Figure 8a shows that the multiplicity region predicted by model 1 agrees quite well with that of model 2. This point is made clearer in Figure 8b where it can be seen that the low-F_l (i.e., high-τ_l) extinction points of model 2 correspond to very small liquid flow rates and may be overlooked in practice. Also note that the multiplicity *patterns* are not considered in comparing the multiplicity *regions*. Thus for model 1, the additional region classified inside the entire multiplicity region does not affect this comparison.

Comparison of Models 2 and 1f

While model 2f predicts uniqueness for the simulation of the experimental results, model 1f predicts multiplicity, as can be realized from Figure 9 or Figure 10. The predicted multiplicity patterns of models 2 and 1f are different except the S-shaped multiplicity as shown in Figure 9, where the multiplicity regions and patterns are classified for model 1f. Note that Da_1 and Da_2 are as in Eq. 5 and are computed by explicit expressions reported earlier (Huang and Varma, 1981a). From Figure 10, as E decreases, the multiplicity region for model 1f shrinks in the same way as that of model 1 but the shrinkage is somewhat faster. However, at E = 20 000 cal/gmol, the multiplicity regions still exists and this is an indication of the superiority of model 1f to model 2f in the prediction of the multiplicity regions.

From Figure 11, it is seen that model 1f predicts multiplicity for all the specific Da values were multiplicity is predicted by model 2. Since reactivities of most gas-liquid reactions are not in the upper range of Da in Figure 11, the agreement of the multiplicity regions predicted by models 2 and 1f is seen to be practically fair. This justifies the use of criterion Eq. 5, i.e., the sufficient condition for multiplicity, at least in providing some insight into the uniqueness and multiplicity features of the system.

Critical Liquid Flow Rate

Figure 12 presents the values of the critical liquid flow rate F_{l2} at different values of the heat of reaction for all four reaction models. The values of F_{l1} for models 2 and 2f are positive but very small and are not shown in Figure 12, and those for models 1 and 1f are negative and thus should be defined as zero. The region above the F_{l2} curve for each model is a region assuring uniqueness for all values of Da, and multiple steady states are possible under that curve for some values of Da except that for models 2 and 2f, uniqueness is again assured for all values of Da at $F_l < F_{l1}$, which is not shown.

From Figure 12, it is seen that the F_{l2} curves of models 1 and 2 are in very good agreement and the F_{l2} values of model 1f, which are analytic as shown before, are closer to the model 2 values than those of model 2f. Note that the F_{l2} values of models 2, 2f and 1 are obtained by trial-and-error. Thus, the use of the explicit expression for F_{l2} reported by us before (Huang and Varma, 1981a) is justifiable at least as a preliminary step to determine the possibility of the occurrence of multiple steady states, and so is our previously reported necessary condition for multiplicity.

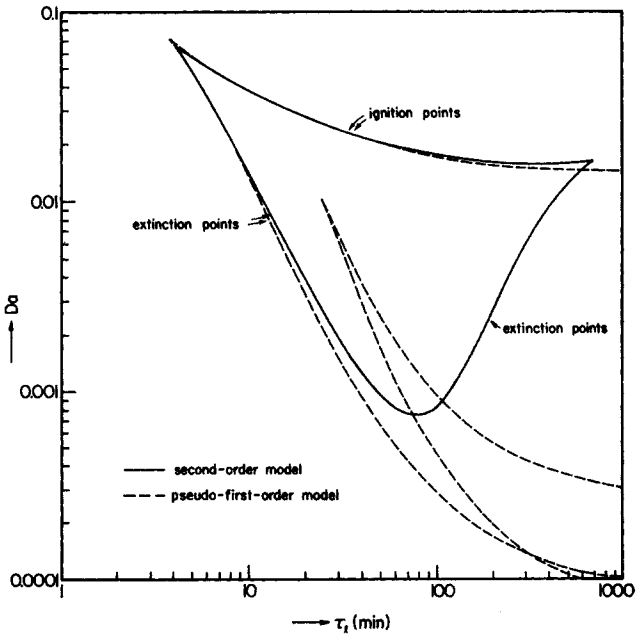

Figure 8a. Comparison of the multiplicity regions of models 1 and 2.

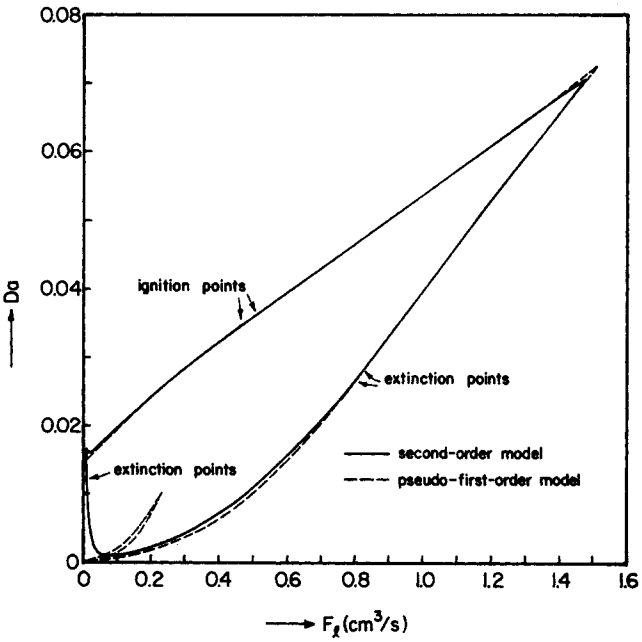

Figure 8b. Comparison of the multiplicity regions of models 1 and 2.

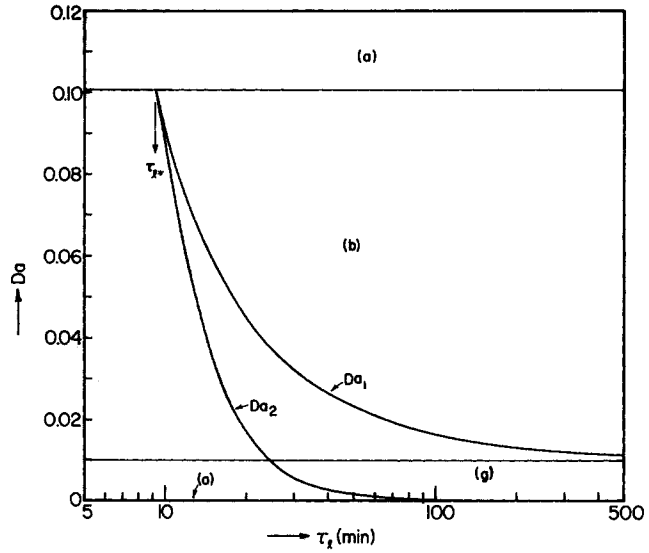

Figure 9. Effect of reactivity on the multiplicity regions and patterns: model 1f. (a), (b) and (g) as in Figure 5a.

CONCLUDING REMARKS

It should be pointed out that the multiplicity *regions* have been emphasized in the above comparisons over the multiplicity *patterns*. This is because the information obtained from the multiplicity patterns is only qualitative in making those comparisons, i.e., the parameter values for which a multiplicity pattern is predicted by the simplified reaction models cannot match those of the second-order reaction model. On the other hand, information from the multiplicity regions is quantitative since only uniqueness and multiplicity are considered instead of the details within the multiplicity regions. Also, because the total uniqueness predicted by model 2f at $E = 20\,000$ cal/gmol is in contrast with the other models, the use of the multiplicity region for discrimination among these reaction models is further justified.

Analytic criteria were derived by Raghuram and Shah (1977) for an adiabatic gas-liquid CSTR applying the pseudo-first-order reaction model (model 1), and the fast second-order reaction model (model 2f). These criteria were claimed to represent conditions that assure uniqueness of the steady state. However,

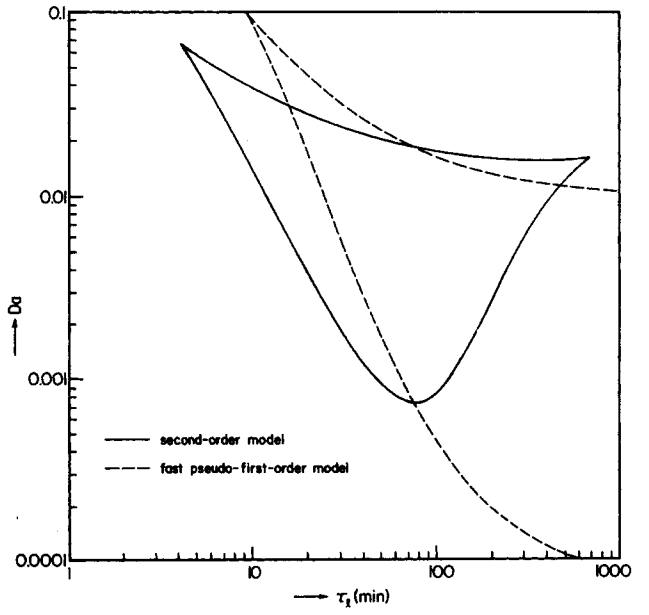

Figure 11. Comparison of the multiplicity regions of models 2 and 1f.

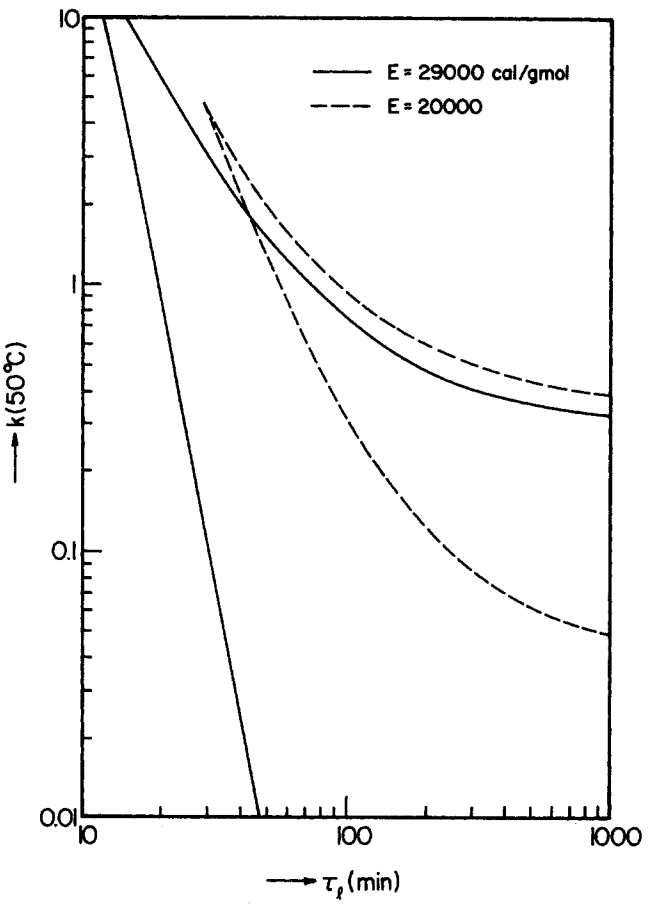

Figure 10. Effect of activation energy on the multiplicity regions: model 1f.

these criteria are *not a priori*, because to apply them, a trial-and-error procedure is necessary for the steady state temperature which is included in these criteria. Thus, these criteria would seem of little practical use.

It should be mentioned that the necessary condition for stability reported by Hoffman et al. (1975) in their Eq. 30, which was applied by Raghuram and Shah (1977) to derive the analytic criteria mentioned above, is actually just a sufficient condition for instability. Thus this condition can be used only to prove the instability of the saddle point (i.e., the steady state on the middle branch if there are three steady states, etc.). This has been stated by Hoffman et al. (1975) in a different way, and is restated here just for clarification.

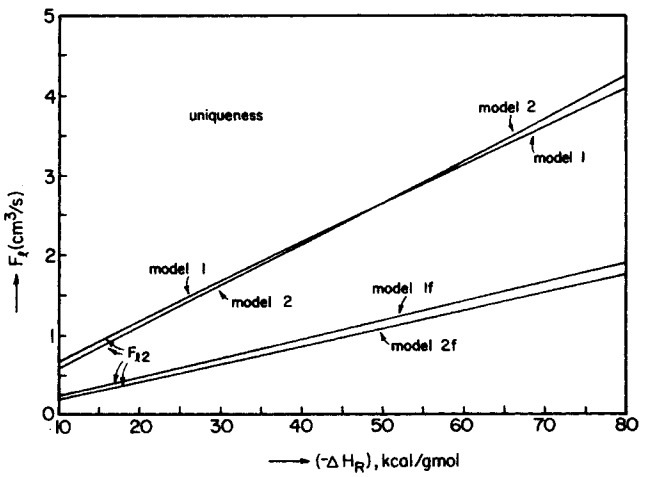

Figure 12. The influence of the reaction models on the critical liquid flow rate (F_{f2}).

On the other hand, *a priori* criteria have been reported recently (Huang and Varma 1981a) applying the fast pseudo-first-order reaction model (model 1f) in a nonadiabatic gas-liquid CSTR to provide necessary and sufficient conditions among physico-chemical parameters which assure unique and multiple steady states, and also the asymptotic stability of the steady states. It is believed that these *a priori* criteria can be derived only by assuming the pseudo-first-order reaction in the "fast" reaction regime (i.e., the fast pseudo-first-order reaction model). Therefore, the use of the fast pseudo-first-order reaction model is justifiable not only because of the fair agreement between the multiplicity regions predicted by this model and the second-order reaction model, but also because of its capability to lead to *a priori* criteria, as shown by the analyticity of the uniqueness and multiplicity regions presented in this work.

Thus, it is recommended that the fast pseudo-first-order reaction model (model 1f) be always applied first to give an idea of the multiplicity regions of second-order reactions; if specific operational conditions are close to regions of multiplicity predicted by this model, then a fine tuning can be done to identify the possibility precisely by using the pseudo-first-order reaction model (model 1) and the full second-order reaction model (model 2)—in that order. Although both models 1 and 2 use trial-and-error, the computational effort is significantly lower for model 1.

ACKNOWLEDGMENT

The Union Oil Fellowship in Reaction Engineering and a Reilly tuition scholarship for D. T.-J. Huang is gratefully acknowledged.

NOTATION

Only those symbols not reported in Part I of this work (Huang and Varma, 1981b) are listed below.

F_{l1}, F_{l2} = critical liquid flow rates, defined by Eq. 4

Da_1, Da_2 = critical values of Da, defined by Eq. 5

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Liquid-Liquid Dispersion in Turbulent Couette Flow

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A dimensionless dispersion number was developed to characterize liquid-liquid dispersions created in turbulent Couette flow with respect to their subsequent coalescence in a settling zone. For a given solvent pair and continuous phase, the dispersion number is relatively constant over a wide range of operating and dimensional parameters. Because of this, the dispersion number can be used for the design and scale-up of liquid extraction equipment with discrete stages.

SCOPE

Couette flow is the name applied to flow in the annular region between two cylinders when one cylinder is rotating. When the rotational speed is sufficient, the flow becomes turbulent and can be used to disperse one liquid in another. This turbulent Couette mixing is an integral part of a centrifugal contactor for solvent extraction developed by Bernstein et al. (1973). In this work, our objective was to develop an understanding of the Couette mixing zone so that (1) the design of the Couette mixing zone can be put on a firm basis and (2) the liquid-liquid dispersion so formed could be related to its subsequent rate of coalescence in the separating zone.

Only the work of Clay (1940) reports on the use of a turbulent Couette mixer to produce a liquid-liquid dispersion. In that study, there was no flow through the mixer. Previous characterizations of liquid-liquid dispersions were made on the basis of: (1) the drop size or interfacial area or (2) the time for the emulsion to break. The use of drop size or interfacial area was avoided in the present work because of the time required to make such measurements and the limited usefulness of such results when mass transfer rates are high. On the other hand, the time for the emulsion to break is easy to measure and has been reported by many persons including Rodger et al. (1956), Ryon et al. (1959), Groenier (1968), and Barnea and Mizrahi (1975). The limitation of this measurement is that the time for an emulsion to break is very specific to the liquids used, the type of mixer, and the type of settling zone. In addition, it has not been clear how the results of this batch test apply to a continuous-flow system.

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