Analysis of Nonstationary Heat and Mass Transfer in a Porous Catalyst Particle II*

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I. INTRODUCTION

The purpose of this paper, which forms a continuation of the previous paper (13), is to discuss the accuracy of the approximations given before (8) by means of comparing them with correct results. Further, problems connected with a stability of steady states and with an occurrence of limit cycles will be investigated.[†]

II. NUMERICAL SOLUTION OF PARTIAL DIFFERENTIAL EQUATIONS

The partial differential equations describing the process form a system of nonlinear parabolic equations.

$$Lw \frac{\partial y}{\partial \tau} = \frac{\partial^2 y}{\partial x^2} + \frac{a}{x} \frac{\partial y}{\partial x} - \frac{\delta}{\gamma \beta} y^n \\ \times \exp \frac{\theta}{1 + \theta/\gamma} \quad (1)$$

$$\frac{\partial \sigma}{\partial \tau} = \frac{\partial}{\partial x^2} + \frac{\alpha}{x} \frac{\partial}{\partial x} + \delta y^n \\ \times \exp \frac{\theta}{1 + \theta/\gamma} \quad (2)$$

$$\tau = 0: \theta = \theta_i(x), \ y = y_i(x)$$

$$\tau > 0: x = 1: y = 1, \ \theta = 0$$
 (3)

$$x = 0: \partial y / \partial x = 0, \frac{\partial \theta}{\partial x} = 0$$
 (4)

An implicit-explicit difference schema of the Crank-Nicolson type with an automatic control of the both step sizes was used for numerical solution. Memory capacity of the digital computer E 503, used for computation, enabled division to 360 parts in radial

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[†] The symbols used in this article are the same as in part I.

direction, but for most calculated cases division to 40-160 parts was sufficient. An average computational time was between 1-5minute. The calculations performed have shown, that the proposed difference schema was always stable. No difficulties during its employment were observed.

III. AN ANALYSIS OF STABILITY OF STATIONARY SOLUTIONS OF Eqs. (1)-(4)

A. Approximation by a System of Ordinary Differential Equations

When we substitute differences for partial derivatives with respect to space coordinate $x(\text{differential-difference} \mod (1))$ we obtain the set of ordinary differential equations:

$$\frac{d\theta^{j}}{d\tau} = \frac{1}{h^{2}} \left[\left(1 - \frac{a}{2_{j}} \right) \theta^{j-1} - 2\theta^{j} + \left(1 + \frac{a}{2_{j}} \right) \theta^{j+1} \right] + R^{j} \quad (5)$$

$$Lw \frac{dy^{j}}{d\tau} = \frac{1}{h^{2}} \left[\left(1 - \frac{a}{2_{j}} \right) y^{j-1} - 2y^{j} + \left(1 + \frac{a}{2_{j}} \right) y^{j+1} \right] - \frac{R^{j}}{\gamma \beta} \quad (6)$$
for $i = 1, 2, \qquad M - 2$

$$\frac{d\theta^0}{d\tau} = \frac{2(a+1)}{h^2} \left(\theta^1 - \theta^0\right) + R^0 \quad (7)$$

$$Lw \frac{dy^{0}}{d\tau} = \frac{2(a+1)}{h^{2}} (y^{1} - y^{0}) - \frac{R^{0}}{\gamma\beta}$$
(8)
for $j = 0$

and

$$\frac{d\theta^{M-1}}{d\tau} = \frac{1}{h^2} \left[\left(1 - \frac{a}{2 (M-1)} \right) \theta^{M-2} - 2\theta^{M-1} \right] + R^{M-1} \quad (9)$$

$$Lw \frac{dy^{M-1}}{d\tau} = \frac{1}{h^2} \left[\left(1 - \frac{a}{2(M-1)} \right) y^{M-2} - 2y^{M-1} + 1 + \frac{a}{2(M-1)} \right] - \frac{R^{M-1}}{\gamma\beta} \quad (10)$$
for $i = M - 1$

where

$$R^{j} = \delta(y^{j})^{n} \exp\left(\frac{\theta^{j}}{1+\theta^{j}/\gamma}\right)$$

and where θ^j and y^j are values of θ and y in the node point x = jh (hence are only dependent on independent variable τ and on M = 1/h).

B. First Method of Ljapunov

For investigation of stability of a stationary solution we shall use the first method of Ljapunov (5). For application of this method it is necessary to linearize Eqs. (5)–(10) in the neighborhood of the stationary state:

$$\frac{d\vartheta^{j}}{d\tau} = \frac{1}{h^{2}} \left(1 - \frac{a}{2_{j}} \right) \vartheta^{j-1} \\
+ \left[\frac{R_{0}^{j}}{(1 + \theta_{0}/\gamma)^{2}} - \frac{2}{h^{2}} \right] \vartheta^{j} \\
+ \frac{1}{h^{2}} \left(1 + \frac{a}{2_{j}} \right) \vartheta^{j+1} + \frac{nR_{0}^{j}}{y_{0}^{j}} \eta^{j} \quad (12)$$

$$Lw \frac{d\eta^{j}}{d\tau} = \frac{1}{h^{2}} \left(1 - \frac{a}{2_{j}} \right) \eta^{j-1} - \left[\frac{nR_{0}^{j}}{\gamma\beta y_{0}^{j}} + \frac{2}{h^{2}} \right] \eta^{j} \\
+ \frac{1}{h^{2}} \left(1 - \frac{a}{2_{j}} \right) \eta^{j-1} - \left[\frac{nR_{0}^{j}}{\gamma\beta y_{0}^{j}} + \frac{2}{h^{2}} \right] \eta^{j} \quad (12)$$

$$+\frac{1}{h^2}\left(1+\frac{a}{2_j}\right)\eta^{j+1} - \frac{h_0^j}{\gamma\beta(1+\theta_0^j/\gamma)^2}\vartheta^j (13)$$

for $j = 1, 2, \ldots, M-2$

$$\begin{aligned} \frac{d\vartheta^{0}}{d\tau} &= \frac{2(a+1)}{h^{2}} \vartheta^{1} \\ &+ \left[\frac{R_{0}^{0}}{(1+\theta_{0}^{0}/\gamma)^{2}} - \frac{2(a+1)}{h^{2}} \right] \vartheta^{0} + \frac{nR_{0}^{0}}{y_{0}^{0}} \eta^{0} \end{aligned}$$
(14)

$$Lw \frac{d\eta^{0}}{d\tau} = \frac{2(a+1)}{h^{2}} \eta^{1} \\ -\left[\frac{nR_{0}^{0}}{\gamma\beta y_{0}^{0}} + \frac{2(a+1)}{h^{2}}\right] \eta^{0} \\ -\frac{R_{0}^{0}}{\gamma\beta(1+\theta_{0}^{0}/\gamma)^{2}} \vartheta^{0} \quad (15) \\ \frac{d\vartheta^{M-1}}{d\tau} = \frac{1}{h^{2}} \left[1 - \frac{a}{2(M-1)}\right] \vartheta^{M-2}$$

$$+\left[\frac{R_{0}^{M-1}}{(1+\theta_{0}^{M-1}/\gamma)^{2}}-\frac{2}{h^{2}}\right]\vartheta^{M-1} + \frac{nR_{0}^{M-1}}{y_{0}^{M-1}}\eta^{M-1} \quad (16)$$

$$Lw \frac{d\eta^{M-1}}{d\tau} = \frac{1}{h^2} \left[1 - \frac{a}{2(M-1)} \right] \eta^{M-2} \\ - \left(\frac{nR_0^{M-1}}{\gamma\beta y_0^{M-1}} + \frac{2}{h^2} \right) \eta^{M-1} \\ - \frac{R_0^{M-1}}{\gamma\beta(1+\theta_0^{M-1}/\gamma)^2} \cdot \vartheta^{M-1} \quad (17)$$

Equations (12)–(17) form a system of linear differential equations with constant coefficients. From eigenvalues of this system it is possible to conclude on a stability of steady state solutions θ_0^{j} , y_0^{j} of Eqs. (5)–(10), and hence also on a stability of steady state solutions of the original system (1)–(4).

Let us write Eqs. (12)-(17) in the form

$$\frac{d\mathbf{w}}{d\tau} = \mathbf{A}\mathbf{w} \tag{18}$$

where the vector function \mathbf{w} is defined as

 $\mathbf{w} = (\vartheta^0, \eta^0, \vartheta^1, \eta^1, \ldots, \vartheta^{M-1}, \eta^{M-1})^T \quad (19)$

The matrix \mathbf{A} is in this case a five-diagonal (there are even zero elements in these five main diagonals). The elements of matrix \mathbf{A} can be evaluated on the basis of a knowledge of the steady state solution (2).

A sufficient condition for stability of steady states is, that the real parts of all eigenvalues of the real matrix \mathbf{A} are negative. A single positive real part of an arbitrary eigenvalue will cause instability.

C. Calculation of Eigenvalues. Routh–Hurwitz Criterion

To be able to make conclusions on stability it is necessary to know or all eigenvalues of the matrix \mathbf{A} , or to use some procedure that enables us to avoid the search for eigenvalues (3, 4, 5) (in this work the Routh-Hurwitz criterion (3) was used).

For determination of all eigenvalues it is necessary to know the characteristic polynomial of the matrix **A**. The simple methods of Krylov and Leverrier (4) were used for the calculation of the coefficients of this polynomial. The whole procedure is relatively laborious, and was, therefore, used only for comparison with the above mentioned



FIG. 1. Comparison of trajectories from the onedimensional and the two-dimensional model (phase plane). n = 1, $\gamma = 20$, $\beta = 0.2$, $\delta = 2.56$; a = 0; $\rho_1 = \pi/2$; Lw = 1. - - - - one-dimensional approximation. — two-dimensional approximation.

Routh-Hurwitz criterion. When applicating this criterion we calculate successively Nmain determinants of the matrix constructed from the coefficients of the characteristic





polynominal. If all the determinants are positive then the system under consideration is stable.

The purpose of the following chapters is to verify the validity of the one-dimensional model. We shall follow mainly an applicability of the approximation in the neighborhood of steady states, a validity of conclusions on stability of steady states made on the basis of the model and a degree of a coincidence between trajectories in the phase plane.

When deriving the simplified model we may suppose, that we work with mean values of temperature and concentration within a particle. We then can compare these mean values of the variables with the integral mean values obtained from the two-dimensional model

$$\bar{\theta}(\tau) = (a+1) \int_0^1 x^a \theta(x,\tau) \, dx \quad (20)$$

First we shall deal with a comparison of the two models in the case where only single steady state (stable or unstable) exists. The steady state classified on the basis of the onedimensional model as stable (β) is in Figs. 1-3 denoted A_1 . The mean value of the two-



dimensional model (in Figs. 1–3 is denoted A_2) can be obtained either from the solution of the steady state problem or as the limit profile of the transient problem. As can be seen from the figures both steady states A_1 and A_2 are stable. The initial profiles of temperature and concentration within a particle were chosen constant. As is shown in the figures, the character of solution in the neighborhood of steady state is well approximated by the one-dimensional model, but the steady state A_1 is somewhat displaced, when



FIG. 5. Trajectories from the two-dimensional model in the phase plane. n = 1; $\gamma = 20$; $\beta = 0.4$; $\delta = 3.38$; a = 2; Lw = 1. ---- constant initial profiles. ______ parabolic initial profiles.



FIG. 6. Comparison of trajectories from the onedimensional and the two-dimensional model (phase plane). n = 1, $\gamma = 20$, $\beta = 0.2$, $\delta = 9$; a = 2; $\rho_1 = 3.83$; Lw = 2.5.

compared with the two-dimensional case. In Fig. 4 the case is shown, where the onedimensional model forecasts a single unstable steady state for given values of parameters. The steady state is encircled by a limit cycle. The steady state A_2 was obtained on the basis of steady state balance equations. The solution of the two-dimensional model (also demonstrated in the figure) has shown, that an asymptotic periodical solution exists which correspond in the phase plane also to a limit cycle. As for the steady states A_1, A_2 so the corresponding limit cycles are shifted in this case. It seems, (when taking into consideration a number of solved examples) that the steady state A_1 is usually shifted in the phase plane with regards to A_2 along the straight line $\theta = \gamma \beta$ \times (1 - y) towards the higher values of θ . The corresponding limit cycle, if it exists at all, is usually shifted in the same direction and its diameter is greater. The character of solution in the neighborhood of the limit cycles is usually conserved.

The case where three stationary states exist $(\gamma\beta > (\gamma\beta)^*, \phi_1 < \phi < \phi_2)$ (9) is for two-dimensional model shown in Fig. 5 (the corresponding figure for one-dimensional



FIG. 7. The influence of Thiele modulus ϕ upon the critical Lewis number Lw^* . n = 1; $\gamma = 20$; $\beta = 0.2$; a = 0. (1) relation (33'), part I; (2) relation (33'), part I, where \bar{y}_0 instead of y_0 was used; (3) Routh-Hurwitz criterion (M = 5); (4) the influence of ϕ upon y_0 ; (5) the influence of ϕ upon \bar{y}_0 .



FIG. 8. The influence of Thiele modulus ϕ upon the critical Lewis number Lw^* . n = 1; $\gamma = 20$; $\beta = 0.2$; a = 1. (1) relation (33'), part I; (2) relation (33'), part I, where \bar{y}_0 instead of y_0 was used; (3) Routh-Hurwitz criterion (M = 5); (4) the influence of ϕ upon y_0 ; (5) the influence of ϕ upon \bar{y}_0 .



FIG. 9. The influence of Thiele modulus ϕ upon the critical Lewis number Lw^* . n = 1; $\gamma = 20$; $\beta = 0.2$; a = 2. (1) relation (33'), part I; (2) relation (33'), part I, where \bar{y}_0 instead of y_0 was used; (3) Routh-Hurwitz criterion (M = 5); (4) the influence of ϕ upon y_0 ; (5) the influence of ϕ upon \bar{y}_0 .



FIG. 10. The influence of Lw upon limit cycles. $n = 1; \gamma = 20; \beta = 0.2; \delta = 9; a = 2.$

model is represented by Fig. 2, part I). Similarly as in the one-dimensional case in the phase plane exist two spheres of influence when considering an appurtenance to the upper (C_2) or the lower (A_2) steady state (the middle one is of a saddle type). The boundary between the two spheres of influence is not fixed and depends on the chosen initial conditions $\theta_i(x)$ and $y_i(x)$. In Fig. 5 the full lines denote the trajectories that have parabolical initial concentration and temperature profiles $y_i(x)$, $\theta_i(x)$. The dashed lines (in the same figure) then denote the trajectories with constant initial profiles of temperature and concentration. In the vicinity of the steady state solution the character of trajectories is similar to that of the one-dimensional case and is independent on the choice of form of initial conditions.

By the proper change of parameter ρ_1 we can achieve an agreement between the onedimensional and the two-dimensional steady states. The agreement between the corresponding trajectories $\theta - y$ is then also better. Such a case is shown in Fig. 6.

STABILITY OF SOLUTION. EFFECT OF LEWIS NUMBER

The purpose of this chapter is to verify the results obtained for the approximate model and to link up them with the results obtained by other authors (10-12).

Let us firstly take into consideration the case, where a single steady state exists. No attention has been devoted to the problem of its instability heretofore. As the analysis of the one-dimensional model (8, 13) has shown and as was verified by the results based on the two-dimensional model, in this case there can exist an asymptotic periodical solution. This can be attained when the Lewis number exceeds certain critical value Lw^* . The problem of determination of this critical value was for the approximate model solved in the first part of this paper (13). In the two-dimensional model we can determine the value of Lw^* or by the method described in chapter III, or by repeated numerical simulation of Eqs. (1)-(4). In Figs. 7-9 dependencies $Lw^* = f(\phi)$ obtained by means of three different methods are given. The line denoted 1 corresponds to the values of Lw^* obtained from the one-dimensional model by using Eq. (33') of the part I; dashed line 4 represents the steady state values of concentration y_0 obtained from the transcendental equation

$$\rho_{1}^{2}(1-y_{0}) - \phi^{2}y_{0}^{n} \\ \times \exp\left[\frac{\gamma\beta(1-y_{0})}{1+\beta(1-y_{0})}\right] = 0. \quad (21)$$

The line 3 represents the dependence $Lw^* =$ $f(\phi)$ obtained from the difference-differential method by means of the Routh-Hurwitz criterion and 5 then corresponds to the values of \bar{y}_0 obtained as mean values of the steady state concentration profiles within a particle. The line 2 was obtained when in the relation (33') (part I) the values of \bar{y}_0 instead of y_0 were used. The line 3 can be taken as a very good approximation of actual values of Lw^* , for the numerical simulation of Eqs. (1)-(4) gives practically the same value of Lw^* . As can be seen from the figures, the agreement between the values of Lw^* obtained from the one-dimensional model and the two-dimensional one is very good for lower values of Thiele modulus ϕ . For higher values of ϕ the agreement between the lines 3 and 2 is, on the contrary, better (this can be explained by great relative differences between y_0 and \bar{y}_0). Limit cycles for different values of Lewis



FIG. 11. Periodical changes of concentration profiles within the catalyst particle. $n = 1; \gamma = 20; \beta = 0.2;$ $\frac{\delta = 9; a = 2; Lw = 5;}{Curve 1 2 3 4 5 6 7 8}$

10.8

10.9

10.7

F1G. 12. Per	iodical chan	ges of te	emperat	ture pr	ofiles w	ithin tl	he cata	alyst pa	rticle. n	$= 1; \gamma$	$= 20; \beta$	= 0.2;
$\delta = 9; a = 2;$	Lw = 5;											_
Curve	1	2	3	4	5	6	7	8	9	10	11	12

Curve	1	2	3	4	5	6	7	8	9	10	11	12
Time	10.45	10.55	10.7	10.8	10.9	10.95	11	11.05	11.15	11.25	11.35	11.5

number are shown in Fig. 10. With increasing Lw the limit cycle also increases. These limit cycles are, similarly as it was shown by Gurel and Lapidus for CSTR (14), asymptotically stable from both sides. The limit cycles in the two-dimensional model have the same course of rotation as in the one-dimensional case.

10.45

10.55

The form of radial concentration and temperature profiles in a particle when autooscillations take place is given in dependence on time in Figs. 11 and 12. On the temperature profile we can sometimes meet with periodical maxima between the center and the surface of the particle (15).

Now we shall deal with the case, where three steady states may exist. As number of authors have shown (2, 10, 11) the middle steady state is always unstable. In the onedimensional approximation, discussed in the part I, this steady state is of a saddle type which is a reason for its instability. A study of instability of the both boundary steady states was still not performed. In the onedimensional analysis it was shown, that for the upper steady state critical value of Lewis number, Lw^* exists always and for the lower one in certain cases. These conclusions hold in an analogous way in the two-dimensional model. If we consider the one-dimensional case, then in a certain range of Lewis numbers there can exist around the upper steady state a limit cycle (13), that disappears when the Lewis number is further increased.

11.05

11.35

11.5

Time



FIG. 13. Unstable upper steady state (phase plane). n = 1; $\gamma = 20$; $\beta = 0.4$; $\delta = 3.38$; a = 2; (a) Lw = 2.5; (b) Lw = 2.25; (c) Lw = 2.12.



FIG. 14. Unstable upper steady state temperature profile in dependence on time. n = 1; $\gamma = 20$; $\beta = 0.4$; $\delta = 3.38$; a = 2, Lw = 2.12.

To verify its existence for the two-dimensional case seems to be a difficult task. The case shown in Fig. 13 was followed. On the basis of the values of Lw^* , obtained by use of the method described in chapter IIIA, the values of Lw were chosen so that the upper steady state was unstable. The slightly perturbated steady state profiles were chosen for the initial profiles $\theta_i(x)$ and $y_i(x)$. An analogous dynamical analysis was performed also for the case where Lw = 2. Mildly perturbated steady state profile changed imperceptible with time, so that it was impossible to decide whether the change was caused by the error of the difference approximation or not. The perturbations were then increased and the numerical solution has shown, that in this case the steady state is either stable or it is surrounded by a small limit cycle. The rate of unfolding of a trajectory in the phase plane can be well followed in Fig. 14, where the dependence $\bar{\theta} = \bar{\theta}(\tau)$ is given for the case shown in Fig. 13 (Lw = 2.12). An unstable lower steady state exists for very high values of Lewis number.

From the case where three steady states can exist it is possible by a small change in the value of Thieles modulus to obtain the case where only single steady state exists; similar case is presented in Fig. 15 (value of ϕ is higher than ϕ_2^9). The dashed line denotes trajectory for Lw = 1, the full line for the case where Lw = 5. Here the dimensionless temperature $\bar{\theta}$ attains extremal values and probably the single stationary state is in the





FIG. 16. Trajectories from the two-dimensional model in the phase plane. n = 1; $\gamma = 30$; $\beta = 0.15$; $\delta = 5.621$; a = 2; Lw = 3.

form of a great limit cycle (but the necessary computation time for evaluation of this limit cycle would be extraordinarily high).

CONCLUSION

The study here presented is only an attempt to make a systematic analysis of nonstationary heat and mass transfer within a porous catalyst particle. The analytical criteria are given which enables one to forecast a priori dynamic properties of the system under consideration. It shows, that for values of the parameter $\gamma\beta \leq 1$ the system is always stable. Wei (16) obtained earlier the same conclusion by applying Ljapunov method in the functional space and recently was this condition given by Luss and Amundson (17). The condition $\gamma\beta \leq 1$ is necessary and also sufficient. As is shown in Table 1a (in the first part of this study) parameter $\gamma\beta$ for many important reactions fulfills this condition. The stationary point has in this case a character of node and the maximum temperature inside the particle is determined during the course of whole transient process by Prater relation with except of the cases where the initial values of temperature and concentration are high.

This transient process can be easily followed in the phase diagram, which can be constructed on the basis of simple lumped parameter model (this here well approximate the complicated "distributed parameter model"), so that it is no need for solution of the system of partial differential equations. As follows from the Table 1a mentioned before, only few reactions have the values of parameter $\gamma\beta > 1$; but all up to this time experimentally studied reactions have, as far as the authors know, the value of the parameter $\gamma\beta$ less than $(\gamma\beta)^*$. The three stationary profiles of concentration and temperature were not experimentally realized up to date on the contrary with other processes where simultaneous heat and mass transfer with chemical reaction takes place, as are, for instance, heat and mass transfer to the surface of catalytic particle (18) or axial mixing in the tubular reactor (19).

From the theoretical point of view it seems interesting that here we meet with a possibility of existence of undamped periodical variations of the temperature and concentration profiles within the particle. So, for example, when $\gamma\beta = 4$ and $\delta = 9$ (see Fig. 3 in the first paper) the oscillations can exist for $Lw \geq 3.25$. Other case is presented in Fig. 16 where for $\gamma\beta = 4.5$ ($\beta = 0.15$) and Lw = 3 also exists a limit cycle. But in all the cases which were here studied the values of Lw, necessary for occurrence of autooscillations are so high, that probably cannot be realized experimentally. In other words, the occurrence of periodical phenomena in the catalyst particle where an exothermic chemical reaction with the power-law kinetics is going on has small probability in practice. The method, given in this paper, which makes use of the fact that the distributed parameter models where the differential operator is of the form $(1/r^m)(d/dr)[r^m(d/dr)] m = 0$, 1, 2) can be substituted by the lumped parameter models is applicable generally. The possibility of mutual confrontation of the approximate model with the original one always exists.

SUMMARY

A method of solution of partial differential equations describing the problem is presented. A discussion of the stability of steady state solutions, based on the analysis of the system of partial differential equations for transient process is given. The procedure used consist of the transformation to the system of ordinary differential equations which are then analyzed by the first method of Ljapunov. In the vicinity of a steady state solution the validity of the one-dimensional model is tested by comparing with the exact model. It is shown, that the forecasts obtained on the basis of the simple model are satisfactory. The effect of Lewis number on the stability of steady states is discussed by making use of the two-dimensional model. The conclusions are illustrated by a number of numerically solved examples.

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