

Mathematical Foundations of Modeling of Catalytic Processes: A Review

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Abstract—The mathematical modeling of catalytic processes is considered, and the qualitative results obtained at the Sobolev Institute of Mathematics and the Boreskov Institute of Catalysis, Siberian Division, Russian Academy of Sciences in recent decades are presented.

General mathematical models of catalytic processes are rather complicated. They often involve nonlinear systems of differential equations, including not only ordinary equations but also parabolic- and hyperbolic-type partial differential equations. These systems are so sophisticated that their comprehensive analysis now seems to be impossible. To make a qualitative or numerical analysis possible, one first has to simplify the model.

When constructing, selecting, and simplifying models, one should take into account the requirements imposed on these models. The solutions of the equations involved must have a physical meaning; in particular, reagent concentrations, temperature, etc. must be positive at any time. Obviously, the mass-, energy-, and momentum-conservation laws must be obeyed. The solutions must exist in the “whole” with respect to time, and in a closed system, the processes must come to a stable equilibrium state. The requirements for the regularity of the initial data of the corresponding boundary problems must not be too rigorous. The norms in which the deviations of solutions from each other are measured are dictated by the nature of the phenomena being examined.

The list of necessary conditions can be continued, but it is already clear that proper modeling of the dynamics of nonlinear catalytic systems is possible only if the qualitative theory of differential equations is well developed, and the theory of partial differential equations in particular. However, despite the relative perfection of the classical theory of ordinary differential equations, it had turned out by the mid-1960s that there was no satisfactory theory of systems with distributed parameters. Only a few particular results were

obtained, and these were insufficient for the mathematical modeling of catalytic processes. This is the reason why researchers began a systematic development of the theory of evolutionary equations in partial derivatives. This work has been intensively performed until now, so we can say that some of the divisions of the theory have now been developed almost completely, and marked progress has been made in the others. Below, we consider the principal branches that are being developed at present.

First of all, it was necessary to fill the gaps in the theory of initial- and boundary-condition problems for linear evolutionary equations. These problems have been thoroughly studied for the simplest equations, such as the heat-conduction and wave equations. As to the systems in the general form, exhaustive information on the classical solvability of parabolic systems has only been obtained for relatively smooth data and only when a large number of so-called fitting conditions have been satisfied. In the hyperbolic case, the search for the correctness conditions for general initial-boundary problems have been started only recently.

The above-noted restrictions on the parabolic systems have been circumvented with the use of a special technique that involves weighted Holder spaces. The principally new result obtained with this approach is that it has become possible to consider not only fractional, but also integer Holder indexes. This approach is currently used to analyze even more general quasi-elliptic equations in special spaces of functions with qualified continuity moduli. Unexpected results were obtained for hyperbolic systems: the class of initial-boundary problems was specified for which the

smoothness of solutions grows indefinitely with time. This behavior is typical of parabolic equations.

The existence of a good linear theory enables one to easily analyze the local solvability (with respect to both time and initial deviations) of nonlinear boundary problems. Nonlocal results are much more difficult to obtain. Progress in this field is due to the works of S.N. Bernshtein, E. De Giorgi, O.A. Ladyzhenskaya, and S.N. Kruzhkov. Another new method for analyzing nonlinear problems employs the so-called generalized Lyapunov functionals. This method gave the most important nonlocal results to date for second-order parabolic equations in one spatial variable. It was possible not only to derive the sufficient (nearly necessary) conditions for the solvability of basic boundary problems as a whole but also to obtain accurate *a priori* estimates of solutions and to prove the theorem on the unconditional stabilization of bounded solutions near the stationary ones. This theorem served as the basis for the complete qualitative theory of quasi-linear one-dimensional equations. The structure of the set of stationary solutions was examined; efficient criteria of solution stability that cover various critical cases were suggested; the attraction regions of stable stationary solutions were determined. The method of generalized Lyapunov functionals proved to be useful in the analyzing systems of parabolic equations relevant to the modeling of chemical processes. Some of its modifications were used to analyze nonclassical equations with alternating coefficients at the highest derivatives.

The passage from second-order equations to equations and systems of higher orders was nontrivial. In the latter case, the behavior of solutions is much more diversified. Even in the case of autonomous systems of ordinary differential equations, the bounded solutions do not necessarily tend to the stationary solutions. It can be demonstrated that almost any closed connected set may be the bounding set for the trajectories of these systems, even when the trajectories allow for the existence of a generalized Lyapunov functional. Therefore, we have to describe the asymptotic behavior of the solutions of parabolic systems in relation to the structure of these systems and, in particular, to set off the classes of problems whose solutions allow for stabilization. This problem was partly solved by elaborating the theory of integral manifolds for nonlinear parabolic systems.

The stability of stationary solutions of dynamic problems is one of the most important items of the qualitative theory. A classical method for solving this type of problems is the linearization method, which is often applied to a wide range of general parabolic and hyperbolic equations without being substantiated. It turned out that the applicability of linearization depends on the norm selected to measure the deviation between the perturbed and stationary solutions. If the norm is appropriately selected, one can analyze the stability of the linearized, rather than the nonlinear, system. The stabil-

ity of solutions of linear problems is also of great importance. It is related to the arrangement of spectral points of the corresponding stationary differential operators with respect to the imaginary axis. For arbitrary elliptic operators, there are no easily checkable stability criteria. However, for second-order scalar operators and for systems of any order with one variable, such criteria have been found. In the latter case, the criterion was obtained by extending Morse's theory of indexes (which was initially intended for self-conjugate systems) to nonsymmetric systems.

Unstable stationary solutions are also of considerable interest. Sometimes, they are very attractive from one or another standpoint, but their instability prevents them from being realized. However, if a system contains enough free parameters that can be arbitrarily varied within certain ranges, one may attempt to organize a controlled process that would occur in the vicinity of a given unstable regime for an arbitrarily long time. Wide classes of problems that really allow such a controlled process were set off. These results, in combination with experimental data, prove the feasibility of unstable processes in chemical reactors. The first steps were made to develop the theory of the optimum control of processes with distributed parameters by generalizing the classical Pontryagin's maximum principle.

The above-listed results made it possible to proceed to a qualitatively new level in modeling chemical processes. A mathematical model was constructed for multicomponent chemical reactions accompanied by diffusion and convective heat and mass transfer. That model was proved to possess all the properties dictated by the physicochemical problem formulation. After a more concrete definition of the expressions for fluxes and the right-hand sides of the relevant equations, which are written in terms of the Marcelin-De Donder law, the Lyapunov functional method was used to formulate the theorem on the stabilization of bounded and strictly positive solutions to a problem that allows for the existence of positive points of the detailed and complex equilibria, and also the theorem on the Lyapunov stability of these equilibria. These results substantiate Prigogine's local potential method. A wide variety of classes of problems were considered. With appropriately interpreted coefficients, they include problems in nonisothermal systems; the Fick and Fourier laws, which describe heat and mass transfer; and the law of mass action and the Arrhenius law, which describe the concentration and temperature variations of a reaction rate.

NONLINEAR DYNAMICS OF CATALYTIC REACTIONS AND PROCESSES

A catalytic reactor is an open system, where chemical reactions and physical processes occur far from their equilibria states. The reaction medium may be multicomponent, heterogeneous, and multiphase. The reaction model is chosen from some hierarchic struc-

ture, depending on the problem being solved and the purpose of the analysis. The data obtained with smaller scale models are introduced in larger scale models.

The geometric similarity theory of catalytic processes is inapplicable to complex reaction systems. The effect of mass, heat, and momentum transfer on the catalytic-reaction rate may depend strongly on the process scale. Accordingly, the process scale may influence the way and selectivity of the reaction. Note that processes occur in wide temperature and pressure ranges, characterized by widely varying relaxation times of reaction steps.

The features that define the type of a mathematical model are the following: the properties of the model, the way of describing the thermodynamics of reactions, the number of phases present in the course of the process, the properties of the reaction medium, fluid dynamics of the reaction medium, heat and mass transfer in the system, and the time variation of these features.

The wealth of experience in modeling industrial catalytic processes and in solving the relevant theoretical problems allows us to classify the mathematical models of catalytic processes into two groups, namely, models with lumped and distributed parameters. The latter are subdivided into models described by parabolic and hyperbolic equations.

In the theory of catalytic reactors, an important place is occupied by the diffusion model, which is a constituent of all of the more complex problems. This model and its limiting variants—the mixing and replacement models—proved to be very fruitful and capable of describing a wide variety of phenomena.

The development and analysis of a mathematical model enables one to significantly simplify the experimentation. However, experiments on the diffusion in a catalyst pellet, heat and mass transfer between a catalyst pellet and the flowing medium, axial blurring of a mark, pellet packing in the reactor, the effect of the reactor size on the gas-velocity field, etc. remain very important. Also critical are studies on the kinetics under the conditions of unrestricted transfer of the reagents, products, and heat. Kinetic studies in special-purpose small gradientless reactors reduce the time it takes to develop a new process or to improve an existing one.

Lumped-Parameter Models

Some problems in the chemical kinetics in well-stirred and plug-flow reactors, heat and mass transfer in porous plates, and some other processes can be solved with the use of lumped-parameter models. These lead to nonstationary systems of first- and second-order ordinary differential equations. The coefficients in rate equations are temperature-dependent. Therefore, the ordinary differential equations modeling chemical reactions in the form of dynamic relationships are nonlinear.

We now list some problems that arise in the qualitative analysis of model equations:

- analysis of the mathematical correctness of the problem (a solution must exist, be unique, and depend continuously on the initial data);

- determination of the number of stationary solutions in relation to the distinctive features of the model, the conditions for the uniqueness and multiplicity of the solution in particular;

- analysis of the stability of stationary solutions;

- finding the bifurcation points for the stationary and periodic solutions;

- mathematical substantiation of the passage to the limits of time and problem parameters;

- model simplification with the retention of its basic qualitative properties and predicting power.

The problem of the determination of the number of stationary solutions arises, for example, in searching for the stationary solution that is the most advantageous from the standpoint of the formation rate of the desired product and in describing the relevant experimental data. The number of stationary solutions can be estimated by topological methods. For the case of one or two stationary solutions, the corresponding phase diagrams were studied and solution stability was analyzed.

The problem of stationary solutions was considered to account for the hysteresis behavior of the stationary rates of H_2 oxidation on nickel, palladium, and platinum and CO hydrogenation. The necessary condition for the stationary-state multiplicity was deduced, which is illustrated by examples of simple catalytic reactions.

A system of rate equations (written as the mass-action equation) and conservation equations has only one positive stationary solution at a given temperature and volume (or at a given temperature and pressure). This statement is valid for the points of chemical (thermodynamic) equilibrium (so-called positive points of detailed balancing) that are the stationary points of rate equations for reversible reactions.

Systems that allow irreversible reactions and are characterized by positive points of the complex equilibrium were introduced and described. It was proved that there is a single complex-equilibrium point in any positive reaction simplex assuming that at least one such point exists. Apparently, these inferences on the uniqueness of the detailed-balancing and complex-equilibrium points were extended to more general closed systems. For this reason, the multiplicity of the stationary solutions of the rate equations written in accordance with the law of mass action for a closed system was discovered only recently.

In practice, one needs not only information on the local stability of stationary solutions, but also knowledge of the dynamic behavior of nonstationary solutions (dynamic trajectories) at long times. Sometimes, of special importance are dynamic regimes that are unbounded at long times. The importance of studying

the stability of stationary solutions and stabilization of nonstationary solutions (existence of a limit at $t \rightarrow \infty$) is commonly recognized.

If the number of stationary solutions is finite, the Poincaré–Bendixon theory for a plane enables one to solve the problem of stabilization of the solutions of ordinary differential equations that are reducible to a system of two equations. For example, in the case that $n = 2$ or 3 and the conservation law is obeyed, no periodic solutions are possible for systems without autocatalysis (a system where reaction products do not accelerate the reaction). For divergent systems of higher dimensionalities, solution stabilization was proven for the case of a finite number of stationary solutions. If the number of stationary solutions is infinite, stabilization will not take place even in infinitely smooth systems.

Modeling of the stationary heat and mass transfer that accompanies a chemical reaction at a catalyst pellet or plate leads to two-point boundary problems for ordinary differential equations. The number of solutions of such a problem depends on the kind of linearity and the dimensionality of the system. For $n = 1$ and 2 (nonisothermic reaction of the type $lA \rightarrow mB$), the number of stationary solutions was found by solving a single specially constructed functional equation. The qualitative and numerical analysis of that equation demonstrated the possibility of the existence of less than three solutions to the corresponding boundary problem. The consideration of spherically symmetric solutions (solutions depending on a radius only) for a sphere with regard for diffusion showed that five or more stationary solutions are possible. The conditions were formulated under which the problems considered have a unique solution.

The unique stationary solution can be numerically found by the “stationarization” of the dynamic problem. However, this method only allows one to obtain stable stationary solutions only. In the case of multiple stationary solutions, the method of solution continuation was used with respect to a parameter. In the case of a single spatial variable, it is appropriate to reduce the two-point problem to the Cauchy problem and then apply the shoot method.

When the stationary solutions are stable, it is possible to use the quasi-steady-state approximation, reduce the dimensionality of the base system of equations, separate the “fast” and “slow” variables, and, finally, reduce the problem to the system of algebraic and differential equations. This method has been widely applied to problems in chemical engineering. In particular, the quasi-steady-state treatment demonstrates that, in the finite interval $t \in [0, T]$, chemical systems may show rather sophisticated behavior.

Periodic (nearly periodic) behavior of reactant concentrations was first observed many years ago, but it has not been the subject of modeling, as it was considered thermodynamically implausible. However, this

observation has stimulated constructing models that reflect the self-oscillating properties of the solutions.

The simplest plane chemical models for non-autocatalytic reactions, formulated in accordance with the law of mass action, have no limit cycles. Therefore, it is appropriate to consider the models that imply an exponential dependence of the reaction rate on the surface coverage of the catalyst. When the dimensionality of the model is increased because of the nonisothermicity of the reaction, the Hopf bifurcation method enables one to fit the model parameters so that the corresponding ordinary differential equations may have either stable or unstable periodic solutions.

Note that the bifurcational values of the parameters at which oscillations occur can be determined quite precisely from experimental data. Thus, the conditions for the existence of periodic solutions (bifurcations) impose certain restrictions on the model parameters and, finally, allow the behavior of the solutions at other parameter values to be refined.

As a rule, chemical processes proceed under nonisothermal conditions. Therefore, the rate equations, which describe the time variation of concentrations, should be supplemented by an equation reflecting the heat balance in the system. The temperature dependence of the rates of the reaction stages is described by the Arrhenius equation.

An isolated chemical system that exchanges neither matter nor heat with the environment must possess the properties of a closed chemical system. If there exist positive points of detailed balancing, the following statements are valid:

solutions of the system tend to stabilization (have a limit at $t \rightarrow \infty$);

there are no periodic solutions;

the equilibrium points are stable.

In some catalytic reactions of partial hydrocarbon oxidation and combustion, one can observe the phenomenon of the motion of the reaction or heat front. A mathematical description of this phenomenon can be obtained by separating out the self-similar solutions of sets of partial differential equations. In terms of the self-similar variable ξ , one arrives at a system of nonlinear ordinary differential equations for which the data are set at $\xi \rightarrow \pm\infty$.

The requirement of the existence of bounded solutions with specified limits at infinity imposes some conditions on the parameters of the problem (including the front velocity). When these conditions are simultaneously satisfied, there exist self-similar solutions (i.e., moving reaction fronts). In the simplest cases (such as a plug-flow reactor with catalyst poisoning), the self-similar solutions can be found analytically. In a more general case, it is possible to prove the existence of self-similar solutions for various front velocities when the problem is appropriately formulated and only physicochemical constraints are imposed.

In view of this, it is necessary to select a realizable self-similar solution from the set of the theoretically feasible ones and to interpret it. One possible realizability criterion is the stability of the solutions in the corresponding functional spaces, because only stable solutions can be realized in experiments.

Distributed-Parameter Models

In describing chemical reactions with regard for diffusion, one has to solve boundary problems for systems of parabolic equations. The accuracy of such problems was investigated in a large number of works. Here, we note only that the classical solution for continuous initial data that satisfy the fitting conditions exists in the small. The solution exists in the large, when certain constraints are imposed on the growth of nonlinearity (constraints that ensure the applicability of the maximum principle and allow one to obtain an *a priori* estimate of the solution gradient).

From both chemical-engineering and theoretical standpoints, it is interesting to analyze the qualitative behavior of solutions at $t \rightarrow \infty$ and the existence, number, and stability of periodic and stationary solutions.

The most comprehensive analysis was performed for a scalar case (one equation). The use of the linearization principle in the first-approximation analysis of the stability of the stationary solution was substantiated. A convenient criterion for the asymptotic stability of the trivial solution of the linear problem was suggested. It was demonstrated how important it is to select an appropriate norm of the deviation of the initial data from the stationary solution. From the standpoint of the general theory of nonlinear parabolic equations, the norm of the difference between solutions is to be measured in spaces that contain derivatives, for example, in the space of continuously differentiable functions C^1 or the Sobolev space W_2^1 . However, in practice, the deviations between functions are most conveniently measured in the space C . This fact is critically important for examining the stability and attraction regions of stationary solutions.

The Lyapunov functional method was used to prove the theorem of the stabilization of all solutions bounded within some norm. It was also used to prove the stabilization of the solutions of variational problems in the case of a finite number of stationary solutions and for reaction-diffusion systems.

Let us consider in greater detail the stability and the way of description of the attraction regions of stationary solutions. The structure of the attraction regions of a stable stationary solution of a nonlinear problem is very complex. By using the maximum principle and the theorem of stabilization, it is possible to demonstrate that the natural subsets of the attraction regions are the regions defined by the neighboring stationary solutions; that is, the stable and unstable solutions alternate.

The number of stationary solutions is an important point in analyzing the stabilization of nonstationary solutions and in describing the attraction regions of stable stationary solutions. In the scalar case, owing to the possibility of natural parametrization, this number can be found by solving the Cauchy problem for the corresponding stationary equation. Examples show that the number of stationary solutions is precisely estimated in terms of the trivial solutions of the Cauchy problem.

Multicomponent chemical reactions accompanied by diffusion and convective mass and heat transfer are described by systems of nonlinear parabolic equations. In their most general form, these equations have long been known as the laws of conservation in the differential form. Without explicitly expressed nonlinearity, only little can be said of the properties of the solutions of these equations. Therefore, it is first required to specify the minimum system of constraints on the nonlinearity that would ensure the satisfaction of the following requirements:

all of the components of the concentration and temperature vectors must be positive;

the total weight of the system must be invariable in time if there is now mass transfer between the system and the environment through an interface;

solutions must exist at any $t > 0$;

in the closed state, when there is no mass transfer between the system and the environment, the system must come to a stable equilibrium (solutions must stabilize).

It is necessary to impose such constraints on nonlinearity that would allow for the existence of a wide variety of models but would not affect the required properties of solutions. This makes the problem in the qualitative analysis of the resulting equations. Note the importance of the problem of deriving the equations describing fluxes as a function of concentrations and their gradients or diffusion coefficients on the concentrations and temperature. The constraints on the diffusion fluxes and nonlinearities that describe the reaction kinetics have been formulated in the axiomatic form. By using these constraints and the theory of the solvability of linear parabolic problems in weighted Holder spaces, it was possible to prove the solvability of the corresponding nonlinear system in the large.

Note that the solutions obtained possess all of the properties dictated by the physicochemical formulation of the problem. After a more concrete definition of the expressions for fluxes and the right-hand sides of the relevant equations, which are written in terms of the Marcelin-De Donder law, the Lyapunov functional method was used to prove the theorem on the stabilization of bounded and strictly positive solutions to a problem that allows for the existence of positive points of the detailed and complex equilibria, and also the theorem on the Lyapunov stability of these equilibria. These results substantiate the local potential method.

The axioms formulated are quite general: with appropriately interpreted coefficients, they are valid for problems in nonisothermal systems; the Fick and Fourier laws, which describe heat and mass transfer; and the law of mass action and the Arrhenius law, which describe the concentration and temperature variations of a reaction rate. Furthermore, weakening the constraints formulated (or abandoning some of them) will lead to the loss of one or more required properties of solutions.

To perform a mathematical analysis of the periodic solutions arising from stationary ones (analogs of those derived from the Andronov–Hopf theorem for parabolic problems), substantiate the linearization principle, and control a process in the vicinity of stationary solutions, one needs the knowledge of the localization of the eigenvalues of the spectral problems obtained by the linearization of the initial problem in the vicinity of stationary solutions. In particular, one needs to know how many eigenvalues are situated in the imaginary right-hand semisphere and how the eigenvalues “creep” to the right-hand semiplane as the problem parameters are varied.

The answers to these questions are given by extending the Lyapunov methods to boundary problems for parabolic systems. For self-conjugate problems with one spatial variable, formulas were derived that give the number of positive eigenvalues. These formulas generalize the familiar Morse’s formulas. Finding the instability indexes of non-self-conjugate problems is reduced to determining the indexes for auxiliary problems that are self-conjugate and conveniently solvable. These results were used to substantiate the process controllability in the vicinity of unstable stationary regimes.

Lyapunov’s stability theory, suggested for parabolic problems, can be used to simplify models and to reduce their dimensionality by passing the limits of parameters. This may be needed, for example, in order to substantiate the quasi-steady-state approach. The situation when the diffusion and kinetic parameters are markedly different is known to be described by singularly perturbed parabolic systems of equations. Tikhonov’s theorems on singularly perturbed ordinary differential equations with small parameters at the time derivatives were extended to parabolic problems. This substantiates the application of the quasi-steady-state treatment to chemical distributed-parameter models described by parabolic equations.

The mass transfer due to only convective diffusion and chemical reaction is commonly described by systems of hyperbolic equations. These equations may arise from the passage to the limits of low diffusion coefficients. The model suggested by A.V. Lykov for matter fluxes that depend not only on the concentration gradients with respect to the spatial variables, but also on the time derivative also leads to highly nonlinear hyperbolic problems. All of the items of the qualitative theory of partial differential equations are valid for

these problems. Note that there are no comprehensive theories of the Lyapunov stability, stabilization, and solvability for hyperbolic problems, due to computational difficulties and large differences between hyperbolic and parabolic problems. However, practical problems in chemical engineering have stimulated the mathematicians. As a result, a theory of correctness and stability was elaborated for a certain class of hyperbolic problems with one spatial variable. The necessary and sufficient conditions were formulated for the increase of the smoothness of the solutions of linear problems with time. These results find application in the qualitative analysis of hyperbolic problems relevant to counterflow chemical reactors. In particular, the cycle-birth bifurcation method (analog of the Andronov–Hopf theorem) was proven to be applicable to the search for periodic solutions to the hyperbolic problem in the vicinity of a stationary solution that changes its stability when system parameters are varied.

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