

## Industrial Reactions Kinetics

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## Abstract

In the paper is presented a theoretical analysis of the role of the reaction kinetics for the solution of the main problems in the chemical industry (biotechnology, heat energy), i.e. the optimal design of new devices and the optimal control of active processes. The thermodynamic and hydrodynamic approximations for the modeling of the industrial process rates are presented and analyzed. The relation between the Onsager's linearity coefficient and mass transfer coefficient is presented.

**Keywords:** Reaction Kinetics; Optimal Design; Optimal Control; Thermodynamic Approximation; Hydrodynamic Approximation

## Introduction

The main problems in the chemical industry (biotechnology, heat energy) are the optimal design of new devices and the optimal control of active processes, i.e. minimization of the investment and operating costs. In the paper will be presented a theoretical analysis of the role of the reaction kinetics for the solution of these main problems. They are solved by chemical engineering with modeling and simulation methods.

Mathematical models of processes in the chemical industry [1] contain equations involving variables and parameters. Depending on the problem solved, variables can become parameters and vice versa. They are input mode variables ( $X$ ), output mode variables ( $Y$ ), and construction parameters ( $A$ ):

$$F(X, Y, A) = 0 \quad (1)$$

where is a "vector", containing all the equations in the model,  $X(Y)$  are the vectors of the input (output) variables, that contain the flow rates and temperatures of the input (output) phase flows and the concentrations of their components,  $A$  is the vector of the constructive parameters, which contains the constructive parameters of the apparatuses.

The solutions to the optimal design problems of new apparatuses use algorithms, where input mode variables ( $X$ ) and output mode variables ( $Y$ ) are set (as parameters) and optimal construction parameters must be obtained (as variables):

$$A = F_1(X, Y) \quad (2)$$

The problems of optimal control of current processes use algorithms, where the output mode variables and constructive parameters are set (as parameters) and the optimal input mode variables are searched:

$$X = F_2(Y, A) \quad (3)$$

In the case of renovation (optimal reconstruction), part of the input mode variables and construction parameters are set.

Optimal design and control in the chemical industry is uniquely related to process rates, so all mathematical descriptions of processes are linked to algorithms to determine these rates.

## The processes rates in the chemical engineering

The processes in the chemical industry (biotechnology, heat energy) are the result of the deviation of the systems from their thermodynamic equilibrium [2]. One system is not in a thermodynamic equilibrium when the concentrations of the components (substances) and the temperature at the individual points in the phase volumes are different. These differences are the result of reactions, i.e. of processes that create or consume substance and (or) heat.

The presented analysis shows that processes in the chemical industry are result of reactions that occur in the phase volume (homogeneous) or on the boundary between two phases (heterogeneous). Homogeneous reactions are generally chemical, while heterogeneous reactions are chemical, catalytic, physical and chemical adsorption, interphase mass transfer in gas-liquid and liquid-liquid systems (on the interphase surface the substance disappears from

one phase and occurs in the other phase). The rates of these processes are determined by the reaction kinetics, which lies at the basis of modeling and simulation in chemical engineering, and solving the basic problems in the chemical industry (biotechnology, heat energy).

### Modeling and simulation

The basics of modeling and simulation in chemical engineering, as part of human knowledge and science, are related to the combination of intuition and logic that has different forms in individual sciences [3]. In the mathematics the intuition is the axiom (unconditional statements that cannot be proven), while the logic is the theorem (the logical consequences of the axiom), but logic prevails over intuition. In the natural sciences (physics, chemistry, biology), the "axioms" (principles, postulates, laws) are not always unconditional, but logic prevails over intuition too.

The processes in chemical engineering take place in the industrial apparatuses, where gas, liquid and solid phases move together or alone. They are described by variables, which are extensive or intensive. In the case of merging of two identical systems, the extensive variables are doubled, but the intensive variables are retained.

The processes in the chemical engineering are the result of a deviation from the thermodynamic equilibrium between two-phase volumes or the volume and phase boundaries of one phase and represent the pursuit of the systems to achieve the thermodynamic equilibrium. They are irreversible processes and their kinetics use mathematical structures derived from Onsager's principle of linearity [2]. According to him, the average values of the derivatives at the time of the extensive variables depend linearly on the mean deviations of the conjugated intensive variables from their equilibrium states. The principle is valid near equilibrium and the proportionality factors are the kinetic constants. When the process is done away from equilibrium (high intensity processes) kinetic constants become kinetic complexes, depending on the corresponding intensive variables.

### Mechanism of influence of reaction kinetics

In the chemical industry (biotechnology, heat energy), processes take place in moving phases (gas, liquid, solid). Reactions (reaction processes) lead to different concentrations (and temperatures) in the phase volumes and the phase boundaries. As a result, hydrodynamic processes, diffusion mass transfer and heat conduction are joined to the reaction processes. Under these conditions there are various forms of mass transfer (heat transfer) that are convective (as a result of phase movements) and diffusion (as a result of concentration (temperature) gradients in the phases).

Convective mass transfer (heat transfer) can be laminar or turbulent (as a result of large-scale turbulent pulsations). Diffusion mass transfer (heat transfer) can be molecular or turbulent (as a result of small-scale turbulent pulsations).

Mathematical models of industrial processes aim at determining the concentration of substances (flow temperatures) in the phases. Mathematical models represent a material (thermal) balance in an elementary (small) phase volume that is equivalent to a mathematical point. Components in this balance are convective mass transfer (heat transfer), diffusion mass transfer (heat transfer) and homogeneous reactions (heat effect of reactions). Heterogeneous reactions take part in the boundary conditions of the equations in mass transfer (heat transfer) models. On this basis models of classical theory of mass transfer were created.

### Theory of mass transfer

The modern theory of mass transfer is based on the theory of the diffusion boundary layer [4,5] (Landau, Levich). This approach replaces (physically justified) elliptic partial differential equations with parabolic partial differential equations, which facilitates their mathematical solution and offers a mathematical description of physical processes with free (not predetermined) ends. The theory of the diffusion boundary layer develops in the case of drops and bubbles [5] (Levich, Krylov), liquid film flows [6] (Levich, Krylov, Boyadjiev, Beschkov), non-linear mass transfer and hydrodynamic stability [7,8] (Krylov, Boyadjiev, Babak).

The modeling of chemical engineering processes has two levels of detail - thermodynamic and hydrodynamic approximation.

### Thermodynamic approximation

The processes in chemical engineering are the result of a deviation from the thermodynamic equilibrium between two-phase volumes or the volume and phase boundaries of one phase and represent the pursuit of systems to achieve thermodynamic equilibrium. They are irreversible processes and their kinetics use mathematical structures derived from Onsager's principle of linearity. According to him, the average values of the derivatives at the time of the extensive variables depend linearly on the mean deviations of the conjugated intensive variables from their equilibrium states. The principle is valid close to equilibrium, and the Onsager's linearity coefficients are kinetic constants. When the process is done away from equilibrium (high intensity processes) kinetic constants become kinetic complexes, depending on the corresponding intensive variables. The thermodynamic approximation models cover the entire volume of the phase or part of it.

### Hydrodynamic approximations

The hydrodynamic level uses the approximations of the mechanics of continua, where the mathematical point is equivalent to an elementary physical volume, which is sufficiently small with respect to the apparatus volume, but at the same time sufficiently large with respect to the intermolecular volumes in the medium. In this level the molecules are not visible, as is done in the next level of detail of Boltzmann.

The models of the hydrodynamic approximations are possible to be created on the basis of the mass (heat) transfer theory, whose models are created by the models of the hydrodynamics, diffusion, thermal diffusion and reaction kinetics, using the logical structures of three main "axioms", related with the impulse, mass and heat transfer:

1. The postulate of Stokes for the linear relationship between the stress and deformation rate, which is the basis of the Newtonian fluid dynamics models;
2. The first law of Fick for the linear relationship between the mass flow and the concentration gradient, which is the basis of the linear theory of the mass transfer;
3. The first law of Fourier for the linear relationship between the heat flux and the temperature gradient, which is the basis of the linear theories of the heat transfer.

These are the laws of the impulse, mass and energy transfer.

In Boltzmann's kinetic theory of the ideal gas, these are three "theorems" that derive from the axiom of the "elastic shock" (in a shock between two molecules the direction and the velocity of the movement change, but the sum of their kinetic energies is retained, i.e. there is no loss of kinetic energy) and the rate coefficients are theoretically determined by the average velocity and the average free run of the molecules.

### Rate of thermodynamic processes

In the chemical engineering, the Onsager's linearity principle is used to determine the mass transfer rate in one phase or between two phases, where the mass of the transferred substance is an extensive quantity, but its concentration is an intensive quantity. The mass  $m$  [kg-mol] of a substance, dissolved in the phase volume, and its derivative at the time (its rate of change over time)  $J_0 = dm/dt$  [kg-mol.s<sup>-1</sup>] depends linearly on the mean deviation from the thermodynamic equilibrium  $(c_0 - c^*)/\delta > 0$  [kg-mol.m<sup>-4</sup>], where  $c_0$  [kg-mol.m<sup>-3</sup>] is the concentration of the dissolved substance in a

point in the phase,  $c^*$  is its equilibrium concentration on the phase boundary,  $\delta$  [m] is the distance between this point and the phase boundary, i.e.

$$J_0 = k_0 \frac{c - c^*}{\delta} \quad (4)$$

is the mass flow through the surface  $s$  [m<sup>2</sup>], where [m<sup>4</sup>.s<sup>-1</sup>] is the Onsager's linearity coefficient.

The mass flow per unit surface  $J = \frac{J_0}{s}$  [kg-mol.m<sup>-2</sup>.s<sup>-1</sup>] is obtained directly

$$J = k (c_0 - c^*) \quad (5)$$

where  $k = \frac{k_0}{s\delta}$  [m.s<sup>-1</sup>] is the mass transfer rate coefficient.

This is a fundamental result, representing the relationship between the thermodynamic kinetics (the Onsager's linearity principle) and the mass transfer kinetics, where the mass transfer coefficient is proportional to the Onsager's linearity coefficient.

In a two-phase system (gas-liquid, liquid-liquid), the concentrations in phase volumes are  $c_i$ ,  $i=1,2$ , and at the phase boundary there is always a thermodynamic equilibrium of the dissolved substance in the two phases  $c_1^* = \chi c_2^*$ , where  $c_i^*$ ,  $i=1,2$ , are the equilibrium concentrations of the dissolved substance. This is the law of Henry and  $\chi$  is the number of Henry (in liquid-liquid systems this is the distribution coefficient). In this case the reaching rate of the thermodynamic equilibrium (Onsager's linearity principle) is

$$J_0 = k_{01} \frac{c_{10} - c_1^*}{\delta_1} = k_{02} \frac{c_2^* - c_{20}}{\delta_2} > 0 \quad (6)$$

The mass flow in this case is

$$J = k_1 (c_{10} - c_1^*) = k_2 (c_2^* - c_{20}) \quad (7)$$

where  $k_i = \frac{k_{0i}}{s\delta_i}$ ,  $i=1,2$  [m.s<sup>-1</sup>] are the mass transfer rate coefficients.

Similarly, the velocity of the interphase mass transfer can be expressed:

$$J = K_1 (c_{10} - \chi c_{20}) = K_2 \left( \frac{c_{10}}{\chi} - c_{20} \right) \quad (8)$$

where  $K_i$ ,  $i=1,2$  [m.s<sup>-1</sup>] are the interphase mass transfer rate coefficients  $c$ :

$$K_1^{-1} = k_1^{-1} + \chi k_2^{-1}, \quad K_2^{-1} = \chi^{-1} k_1^{-1} + k_2^{-1} \quad (9)$$

An analogous result can also be obtained in the case of heat conduction in the presence of temperature differences.

The Onsager's principle of linearity is the thermodynamic approximation of the mathematical description of the kinetics of the complex irreversible processes, but it does not show the way to achieve the equilibrium (the mechanism of the process) as a result the velocity coefficients are unknown. Obviously, this "thermodynamic level" does not allow for a real quantitative description of the reaction kinetics of irreversible processes in chemical engineering, and the use of the next level of description detail, the so-called "hydrodynamic level", is required.

### Relationship between thermodynamic and hydrodynamic approximation

The presence of concentration differences in phase volumes (as a result of the reactions) leads to molecular diffusion and the diffusion mass transfer flow [kg·mol·m<sup>-2</sup>·s<sup>-1</sup>] is determined by the first law of Fick:

$$I = -D \text{grad}c \quad (10)$$

where  $D$  [m<sup>2</sup>·s<sup>-1</sup>] is the molecular diffusion coefficient.

In the case of one-dimensional diffusion

$$I = -D \frac{dc}{dx} \quad (11)$$

where  $c$  is the solution of the equation:

$$\frac{d^2c}{dx^2} = 0; \quad x = 0, \quad c = c^*; \quad x = \delta, \quad c = c_0. \quad (12)$$

As a result is obtained

$$(13)$$

The condition  $I = -J$  makes it possible to determine the diffusion mass transfer coefficient  $k = D/\delta$ . This is a fundamental result that links the thermodynamic and hydrodynamic approximation in determining the read of the industrial processes, i.e. the relationship between Onsager's linearity principle and Fick's first law, where Fick's first law is a consequence of Onsager's principle of linearity in diffusion processes. An analogous connection can also be obtained with the first Fourier law.

This approach will be illustrated in the cases of mass transfer in flowing liquid films and one-phase and two-phase diffusion boundary layers.

### Liquid film flows

Let's consider the absorption of slightly soluble gas in a laminar liquid film flow on a vertical flat surface ( $y=0$ ). The equation of convection-diffusion has the form [1]:

$$(14)$$

There is a thermodynamic equilibrium at the surface ( $y = h_0$ ) of the film and  $c^*$  is the equilibrium concentration. The solid surface ( $y = h_0$ ) is impermeable to the diffusion substance, whose concentration at the inlet is  $c_0 < c^*$  (absorption). A film with length  $l$  will be considered. The thickness of the diffusion boundary layer  $\delta$  is less than the thickness of the liquid film  $h_0$ , which allows the diffusion boundary layer approximation to be used. As a consequence of this approach, the following generalized variables can be introduced:

$$x = lX, \quad y = h_0 - \delta Y, \quad c = c_0 + (c^* - c_0)C, \quad (15)$$

where  $\delta \ll h_0$  and  $h_0 \ll l$ .

The introduction of the generalized variables leads to

$$\left(1 + \frac{\delta^2}{h_0^2} Y^2\right) \frac{\partial C}{\partial X} = \frac{Dl}{u_m \delta^2} \left(\frac{\delta^2}{l^2} \frac{\partial^2 C}{\partial X^2} + \frac{\partial^2 C}{\partial Y^2}\right), \quad (16)$$

where

$$(17)$$

In these equations  $u_{av}$  is the film average velocity,  $Fo$  - Fourier number),  $Pe$  - Peclet number. In these conditions the problem must be solved in diffusion boundary layer approximation ( $10^{-2} > Pe^{-1} = 0$ ), i.e.

$$(18)$$

The mass transfer rate in a film of length  $l$  is the average value of the local mass flux through the film surface ( $y = h_0$ ). It can also be represented by the mass transfer coefficient  $k$ , i.e.

$$J = \frac{D}{l} \int_0^l \left(\frac{\partial c}{\partial y}\right)_{y=h_0} dx = k(c^* - c_0). \quad (19)$$

The resulting expression allows the determination of the Onsager's linearity coefficient after solving the convection-diffusion equation (18).

The introduction of the generalized variables leads to:

$$Sh = \frac{kl}{D} = -\sqrt{Pe} \int_0^1 \left( \frac{\partial C}{\partial Y} \right)_{Y=0} dX. \quad (20)$$

In (18) is a small parameter and the perturbation method must be used. As a result is obtained:

$$Sh = \sqrt{\frac{6Pe}{\pi}} \left( 1 - \frac{Fo}{6} - \frac{19Fo^2}{120} \right), \quad (21)$$

where  $Sh$  is the Sherwood number and represent the dimensionless form of the mass transfer coefficient.

The thickness of the diffusion boundary layer varies along the film length, and as a result, the Onsager's linearity principle has the form:

$$J_0(x) = k_0 \frac{(c_0 - c^*)}{\delta_0(x)}, \quad \delta_0(x) = \sqrt{\frac{Dx}{u_{av}}}, \quad J(x) = \frac{k_0}{s} (c_0 - c^*) \sqrt{\frac{u_{av}}{Dx}}. \quad (22)$$

As a result, the average value of the mass flux through the film surface is:

$$\bar{J}(x) = \frac{1}{l} \int_0^l J(x) dx = \frac{2k_0}{s} (c_0 - c^*) \sqrt{\frac{u_{av}}{Dl}} = k(c_0 - c^*), \quad (23)$$

From where the relation between of the Onsager's linearity coefficient and the mass transfer coefficient is obtained:

$$k_0 = \frac{ks}{2} \sqrt{\frac{Dl}{u_{av}}}. \quad (24)$$

### One-phase diffusion boundary layers

The interphase mass transfer in the gas (liquid) – solid systems is realized at a fixed phase boundary. The flow on a smooth semi-infinite plate of a potential flow with a constant velocity, will be considered. The substance of the solid phase proceeds with a concentration, and on the solid surface its equilibrium concentration is. Depending on the sign of the concentration difference there is a process of deposition (crystallization) or dissolution. In this case, the velocity and the concentration satisfy the equations of the laminar boundary layer and the diffusion boundary layer [1]:

(25)

The mass transfer rate in a diffusion boundary layer of length  $l$  is the average value of the local mass flux through the solid surface ( $y = 0$ ). It can also be represented by the mass transfer coefficient  $k$ , i.e.

$$J = \frac{D}{l} \int_0^l \left( \frac{\partial c}{\partial y} \right)_{y=0} dx = k(c^* - c_0). \quad (26)$$

The solution of (25) permits to be obtained the Sherwood number:

$$Sh = \frac{kl}{D} = -Pe^{0.5} \psi'(0) \approx \frac{2}{3} \sqrt{Re} \sqrt[3]{Sc}, \quad (27)$$

where  $\psi'(0)$  is a solution of:

$$\begin{aligned} \varphi''' + \varepsilon^{-1} \varphi \varphi'' = 0, \quad \psi'' + \varepsilon \varphi \psi' = 0; \\ \varphi(0) = 0, \quad \varphi'(0) = 0, \quad \psi(0) = 1, \quad \varphi'(\infty) = 2\varepsilon^{-1}, \quad \psi(\infty) = 0. \end{aligned} \quad (28)$$

The thickness of the diffusion boundary layer varies along the solid surface, and as a result, the Onsager's linearity principle has the form:

$$J_0(x) = k_0 \frac{(c_0 - c^*)}{\delta_0(x)}, \quad \delta_0(x) = \sqrt{\frac{Dx}{u_0}}, \quad J(x) = \frac{k_0}{s} (c_0 - c^*) \sqrt{\frac{u_0}{Dx}}. \quad (29)$$

As a result, the average value of the mass flux through the solid surface is:

$$\bar{J}(x) = \frac{1}{l} \int_0^l J(x) dx = \frac{2k_0}{s} (c_0 - c^*) \sqrt{\frac{u_0}{Dl}} = k(c_0 - c^*), \quad (30)$$

from where the relation between of the Onsager's linearity coefficient and the mass transfer coefficient is obtained:

$$k_0 = \frac{ks}{2} \sqrt{\frac{Dl}{u_0}}. \quad (31)$$

### Two-phase diffusion boundary layers

The interphase mass transfer into the gas-liquid and liquid-liquid systems is realized at a moving interphase boundary. In the approximations of the boundary layer theory the processes are described by the following set of equation [1]:

$$\begin{aligned}
 \varphi_j(0) = 0, \quad \varphi_j'(\infty) = \frac{2}{j\varepsilon_j}, \quad \psi_j(\infty) = 0, \quad j = 1, 2; \\
 \varphi_1'(0) = 2\theta_1 \frac{\varepsilon_2}{\varepsilon_1} \varphi_2'(0), \quad \varphi_2''(0) = -0,5\theta_2 \left(\frac{\varepsilon_1}{\varepsilon_2}\right)^2 \varphi_1''(0), \\
 \psi_1'(0) = \frac{\chi}{\varepsilon_0} \psi_2'(0), \quad \psi_1(0) + \psi_2(0) = 1.
 \end{aligned}
 \tag{32}$$

The index for the first phase ( $j = 1$ ) is for gas or liquid, and for the second phase ( $j = 2$ ) is for liquid. At the phase boundary there is thermodynamic equilibrium and  $\chi$  is the Henry's number (in liquid-liquid systems is the distribution coefficient).

The average rate of the interphase mass transfer through a phase boundary with a length  $l$  is similarly determined:

$$\begin{aligned}
 J &= K_1 (c_{10} - \chi c_{20}) = \frac{1}{l} \int_0^l I_1 dx = k_1 (c_{10} - c_1^*) = \\
 &= K_2 \left( \frac{c_{10}}{\chi} - c_{20} \right) = \frac{1}{l} \int_0^l I_2 dx = k_2 (c_2^* - c_{20}), \quad c_1^* = \chi c_2^*,
 \end{aligned}
 \tag{33}$$

where  $K_j, j = 1, 2$  are the interphase mass transfer coefficient and  $k_j, j = 1, 2$  are mass transfer coefficient in the phases.

The local mass fluxes are

$$I_j = -D_j \left( \frac{\partial c_j}{\partial y} \right)_{y=0}, \quad j = 1, 2 \tag{34}$$

and the Sherwood numbers are:

$$Sh_j = \frac{K_j l}{D_j} = \frac{\chi^{j-1}}{c_{10} - \chi c_{20}} \int_0^l \left( \frac{\partial c_j}{\partial y} \right)_{y=0} dx, \quad j = 1, 2. \tag{35}$$

From (33) and (35) is possible to be obtained:

$$\begin{aligned}
 K_1^{-1} &= k_1^{-1} + \chi k_2^{-1}; \quad \chi = 0, \quad K_1 = k_1; \\
 K_2^{-1} &= (\chi k_1)^{-1} + k_2^{-1}; \quad \chi \rightarrow \infty, \quad K_2 = k_2.
 \end{aligned}
 \tag{36}$$

At the end for the Sherwood numbers is obtained:

$$Sh_j = -\sqrt{Pe_j} \psi_j'(0), \quad Pe_j = \frac{u_{j0} l}{D_j}, \quad j = 1, 2,$$

where  $\psi_j'(0), j = 1, 2$  is the solution of the equations set:

$$\varphi_j'' + j\varepsilon_j^{-1} \varphi_j \varphi_j'' = 0, \quad \psi_j'' + j\varepsilon_j \varphi_j \psi_j' = 0;$$

The thickness of the diffusion boundary layers changes along the interphase surface, and as a result, the Onsager's linearity principle has the form:

$$\tag{39}$$

As a result, the average values of the mass fluxes through the interphase surfaces are:

$$\bar{J}(x) = \frac{1}{l} \int_0^l J(x) dx = \frac{2k_{0j}}{s} (c_{j0} - c_j^*) \sqrt{\frac{u_{0j}}{D_j l}} = k_j (c_{j0} - c_j^*), \quad j = 1, 2, \tag{40}$$

From where the relation between of the Onsager's linearity coefficients and the mass transfer coefficients are obtained:

$$k_{0j} = \frac{k_j s}{2} \sqrt{\frac{D_j l}{u_{0j}}}, \quad j = 1, 2. \tag{41}$$

## Conclusions

In the paper is presented the role of the reaction kinetics for solution the main problems in the chemical industry (biotechnology, heat energy), i.e. the optimal design and optimal control of the industrial processes.

Industrial processes are the result of reactions, i.e. creation or disappearance of a substance and (or) heat as a result of chemical and (or) physical processes and their rate is determined by the reaction kinetics.

The reactions deviate the systems from the thermodynamic equilibrium and as a result processes arise, who are trying to restore that equilibrium. The rate of these processes can be determined by Onsager's "linearity principle", where the rate of the process depends linearly on the deviation from the thermodynamic equilibrium.

The Onsager's linearity coefficient can be determined after solving the hydrodynamics, mass transfer and heat transfer equations, where it is proportional to the mass transfer (heat transfer) coefficient.

The dimensionless form of the mass transfer coefficient is the Sherwood number.

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