UDC 517.9

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EXISTENCE AND UNIQUENESS THEOREM TO A MODEL OF BIMOLECULAR SURFACE REACTIONS

ТЕОРЕМА ІСНУВАННЯ ТА ЄДИНОСТІ ДЛЯ МОДЕЛІ БІМОЛЕКУЛЯРНИХ ПОВЕРХНЕВИХ РЕАКЦІЙ

We prove the existence and uniqueness of classical solutions to a coupled system of parabolic and ordinary differential equations in which the latter are determined on the boundary. This system describes the model of bimolecular surface reaction between carbon monoxide and nitrous oxide occurring on supported rhodium in the case of slow desorption of the products.

Доведено існування та єдиність класичних розв'язків зв'язаних систем параболічних та звичайних диференціальних рівнянь, останні з яких визначені на межі. Система описує модель бімолекулярної поверхневої реакції між монооксидом вуглецю та закисом азоту, що відбувається на нанесеному родії у випадку повільної десорбції продуктів.

1. Introduction. Heterogeneous catalytic reactions are modeled by a coupled system of parabolic and ordinary differential equations. Some of these equations are considered in the domain, while the other equations have to be solved on a part of the boundary. The unimolecular reaction model taking into account the reactant adsorption and desorption and both fast and slow product desorption is considered in [1] and [2] and the existence and uniqueness of a classical solution are proved. In [9] and [10] the same problems are solved numerically. A model of unimolecular surface reactions involving adsorbate diffusion and rapid product desorption is studied in [3], where the existence and uniqueness of classical solutions are proved. The model is described by a system of parabolic differential equations, with one of them defined on a part of the boundary. In [11], the same problem is solved numerically. A bimolecular surface reaction model, where the concentration of the reactant on the surface is given and the product desorption is fast, is studied in [7] and [12] by using Monte Carlo simulations.

In [4] we proved the existence and uniqueness theorem of the classical solution to the model of bimolecular surface reactions between the carbon monoxide and nitrous oxide, $CO + N_2O = N_2 + CO_2$, occurring on supported rhodium, Rh in the case of the rapid products desorption. In the present paper we consider the same reaction but with a slow products desorption and prove the existence and uniqueness theorem of the classical solution. This reaction proceeds via the following elementary steps:

$$\begin{gathered} CO + K \underset{\kappa_{11}}{\overset{\kappa_1}{\rightleftarrows}} COK, \qquad N_2O + K \underset{\kappa_{22}}{\overset{\kappa_2}{\rightleftarrows}} N_2OK, \qquad N_2OK \overset{\kappa_{22}^*}{\to} \hat{N}_2 + OK, \\ COK + OK \overset{\kappa_{13}}{\to} \widehat{CO}_2 + 2K, \qquad \hat{N}_2 \overset{\kappa_4}{\to} N_2, \qquad \widehat{CO}_2 \overset{\kappa_5}{\to} CO_2, \end{gathered} \tag{1}$$

where K is a free adsorption site of the catalyst surface S, CO and N_2O are reactants, COK and N_2OK are adsorbates of CO and N_2O, OK is the intermediate product, $\hat{N}_2$ and $\widehat{CO}_2$ are reaction products before the desorption, N_2 and CO_2 are reaction products after desorption from the catalyst surface, κ_i and κ_{ii}, $i = 1, 2$, are the adsorption and desorption rate constants, κ_{13} and κ_{22}^* are the forward reaction rate constants, κ_4 and κ_5 are the products $\hat{N}_2$ and $\widehat{CO}_2$ desorption rate constants. Two first steps in this reaction are reversible, while the other four ones are irreversible.

The paper is organized as follows. In Section 2 we describe the model. In Section 3, we formulate main results. A priori estimates are given in Section 4. Sections 5 and 6 are devoted to the uniqueness and existence of the classical solution to problems (2)–(4).

2. The slow product desorption model. Set $A_1 = CO,\ A_2 = N_2O,\ B_1 = N_2,\ B_2 = CO_2,$ $\hat{B}_1 = \hat{N}_2,\ \hat{B}_2 = \widehat{CO}_2,\ A_1K = COK,\ A_2K = N_2OK,\ QK = OK$. Then we have a mathematical model of a bimolecular heterogeneous catalytic reaction of type $A_1 + A_2 = B_1 + B_2$, which proceeds via scheme (1). In what follows we consider the case where the desorption of reaction products B_1 and B_2 is slow.

Suppose that the reactants $A_1,\ A_2$ and reactions products $B_1,\ B_2$ occupy a bounded domain $\Omega \subset \mathbb{R}^n$, $n \geq 3$; $a_1 = a_1(x,t)$, $a_2 = a_2(x,t)$ and $b_1 = b_1(x,t)$, $b_2 = b_2(x,t)$ are their concentration at point $x \in \Omega$ at time t, respectively. Let $S := \partial\Omega \subset C^{1+\alpha}$, $\alpha \in (0,1)$, be a surface of dimension $n-1$, and let S_2 be not empty closed part of S of the same dimension, and $S_1 = S \setminus S_2$. We suppose that $\rho = \rho(\xi)$ is the concentration of the adsorption sites of surface S at point $\xi \in S$, $\rho \in C(S)$, $\rho(\xi) \geq 0$ for $\xi \in S$ and $\rho(\xi) = 0$ for $\xi \in S_1$; $\rho\theta_i = \rho(\xi)\theta_i(\xi,t)$ is the concentration of A_iK, $i = 1,2$, at point $\xi \in S_2$ at time t; $\rho\theta_3 = \rho(\xi)\theta_3(\xi,t)$ is the concentration of the intermediate product QK at point $\xi \in S_2$ at time t; $\rho\theta_4 = \rho(\xi)\theta_4(\xi,t)$ and $\rho\theta_5 = \rho(\xi)\theta_5(\xi,t)$ are the concentrations of products $\hat{B}_1,\ \hat{B}_2$ at point $\xi \in S_2$ at time t before their desorption; $\rho(1-\theta)$ is the concentration of the free adsorption sites of S_2; $\theta = \sum_{i=1}^{5} \theta_i$.

Applying the mass action law and assumption that the desorption of reaction products $\hat{B}_1$ and $\hat{B}_2$ is slow we get the Cauchy problem for $\theta_i = \theta_i(\xi,t)$, $i = 1,2,\ldots,5$, $\xi \in S_2$,

$$
\begin{aligned}
&\theta_1' = \kappa_1 a_1(1-\theta) - \kappa_{11}\theta_1 - \kappa_{13}\rho\theta_1\theta_3, && \theta_1|_{t=0} = \theta_{10},\\
&\theta_2' = \kappa_2 a_2(1-\theta) - \kappa_{22}\theta_2 - \kappa_{22}^*\theta_2, && \theta_2|_{t=0} = \theta_{20},\\
&\theta_3' = \kappa_{22}^*\theta_2 - \kappa_{13}\rho\theta_1\theta_3, && \theta_3|_{t=0} = \theta_{30},\\
&\theta_4' = \kappa_{22}^*\theta_2 - \kappa_4\theta_4, && \theta_4|_{t=0} = \theta_{40},\\
&\theta_5' = \kappa_{13}\rho\theta_1\theta_3 - \kappa_5\theta_5, && \theta_5|_{t=0} = \theta_{50}.
\end{aligned}
\tag{2}
$$

System (2) involves the unknown values of a_1 and a_2 on the boundary S_2. To close this system we add equations for diffusion of reactants A_1 and A_2,

$$
\begin{aligned}
&\frac{\partial a_i}{\partial t} - k_i \Delta a_i = 0 && \text{in} \quad \Omega \times (0,T),\\
&k_i \frac{\partial a_i}{\partial \mathbf{n}} = 0 && \text{on} \quad S_1 \times (0,T),\\
&k_i \frac{\partial a_i}{\partial \mathbf{n}} + \kappa_i \rho a_i(1-\theta) = \kappa_{ii}\rho\theta_i && \text{on} \quad S_2 \times (0,T),\\
&a_i|_{t=0} = a_{i0} && \text{in} \quad \overline{\Omega}
\end{aligned}
\tag{3}
$$

for $i = 1,2$.

The diffusion of products B_1 and B_2 can be described by the equations

$$
\begin{aligned}
&\frac{\partial b_i}{\partial t} - \hat{k}_i \Delta b_i = 0 && \text{in} \quad \Omega \times (0,T),\\
&\hat{k}_i \frac{\partial b_i}{\partial \mathbf{n}} = 0 && \text{on} \quad S_1 \times (0,T),\\
&\hat{k}_i \frac{\partial b_i}{\partial \mathbf{n}} = \kappa_{3+i}\rho\theta_{3+i} && \text{on} \quad S_2 \times (0,T),\\
&b_i|_{t=0} = b_{i0} && \text{in} \quad \overline{\Omega}
\end{aligned}
\tag{4}
$$

for $i = 1, 2$. Here $\theta_i' = \partial\theta_i/\partial t$, $\theta_{i0} = \theta_{i0}(\xi)$, $\xi \in S_2$, is the initial value of θ_i, $i = 1, 2, \ldots, 5$; Δ is the n-dimensional Laplace operator; $\partial/\partial\mathbf{n}$ is the outward normal derivative to S; $a_{i0} = a_{i0}(x)$ and $b_{i0} = b_{i0}(x)$ are the initial concentrations of A_i and B_i at point $x \in \overline{\Omega}$; k_i and $\hat{k}_i$ are the diffusivities of the reactant A_i and product B_i, $i = 1, 2$. All constants κ_1, κ_{11}, κ_{13}, κ_2, κ_{22}, κ_{22}^*, κ_4, κ_5, k_i, $\hat{k}_i$ are assumed to be positive.

Model (2) – (4) possesses the following three mass conservation laws:

$$
\int\limits_{\Omega} \big(a_1(x,s) + b_2(x,s)\big)\,dx\Big|_{s=0}^{s=t} + \int\limits_{S_2} \rho(\xi)\big(\theta_1(\xi,s) + \theta_5(\xi,s)\big)\,dS_\xi\Big|_{s=0}^{s=t} = 0,
$$

$$
\int\limits_{\Omega} \big(a_2(x,s) + b_1(x,s)\big)\,dx\Big|_{s=0}^{s=t} + \int\limits_{S_2} \rho(\xi)\big(\theta_2(\xi,s) + \theta_4(\xi,s)\big)\,dS_\xi\Big|_{s=0}^{s=t} = 0,
$$

$$
\int\limits_{\Omega} \big(a_1(x,s) + 2b_2(x,s) + a_2(x,s)\big)dx\Big|_{s=0}^{s=t} +
$$

$$
+ \int\limits_{S_2} \rho(\xi)\Big(2\theta_5(\xi,s) + \theta_2(\xi,s) + \theta_3(\xi,s)\Big)dS_\xi\Big|_{s=0}^{s=t} = 0.
$$

To prove these laws, it is sufficient to integrate eqs. (3), (4) over the cylinder $\Omega \times (0,t)$, apply the formula of integration-by-parts, and use eqs. (2) with the boundary and initial conditions.

Thus, the bimolecular catalytic reactions can be described by system (2) – (4). Our aim is to prove, for this system, the existence and uniqueness theorem. For every collection of continuous functions θ_4, θ_5, problem (4) has a unique classic solution. Therefore, it is sufficient to prove the solvability of problem (2), (3).

3. Main results.

Assumption 3.1. *The initial functions θ_{i0}, $i = 1, 2, \ldots, 5$, a_{i0}, b_{i0}, $i = 1, 2$, and given function ρ satisfy the following conditions*:

1. *The functions θ_{i0}, $i = 1, 2, \ldots, 5$, are continuous and nonnegative on S_2, and $\theta_0(\xi) = \sum_{i=1}^{5} \theta_{i0}(\xi) < 1$ for all $\xi \in S_2$.*
2. *The functions a_{i0}, b_{i0}, $i = 1, 2$, are continuous and nonnegative in a closed domain $\overline{\Omega}$.*
3. *$\rho \in C(S)$, $\rho(\xi) \geq 0$ for all $\xi \in S$, and $\rho(\xi) = 0$ for all $\xi \in S_1$.*

Assumption 3.2. *The functions a_{i0}, b_{i0}, $i = 1, 2$, are continuously differentiable on a neighbourhood of the surface S.*

Definition 3.1. *Functions θ_i, $i = 1, 2, \ldots, 5$, and a_i, b_i, $i = 1, 2$, form a classical solution to problem* (2) – (4) *if $\theta_i \in C(S_2 \times [0,T])$, $\theta_i' \in C(S_2 \times (0,T))$, the derivatives $\partial a_i/\partial\mathbf{n}$ and $\partial b_i/\partial\mathbf{n}$ are continuous on $S \times [0,T]$, and $a_i, b_i \in C^{2,1}(\Omega \times (0,T]) \cap C(\overline{\Omega} \times [0,T])$, $i = 1, 2$, and they satisfy system* (2) – (4).

The main result is the following theorem.

Theorem 3.1. *Let* Ω *be a bounded domain in* $\mathbb{R}^n$ *and* $S = \partial\Omega$ *be a surface of class* $C^{1+\alpha}$, $\alpha \in (0,1)$. *Let the known functions* θ_{i0}, a_{i0}, b_{i0}, *and* ρ *satisfy Assumptions* 3.1, 3.2, *and* $\kappa_{22} \geq \kappa_{22}^*$. *Then problems* (2)–(4) *has a unique classical solution.*

The proof of this theorem is based on the a priori estimates formulated in the following propositions.

Lemma 3.1. *Let* a_i, $i = 1,2$, *be a given continuous and nonnegative on* $S_2 \times [0,T]$ *functions,* θ_{i0}, $i = 1,\ldots,5$, *and* ρ *satisfy Assumption* 3.1, *and* $\kappa_{22} \geq \kappa_{22}^*$. *Let* θ_i, $i = 1,\ldots,5$, *be a solution*[1] *of Cauchy problem* (2). *Then* $\theta_i(\xi,t) \geq 0$, $i = 1,\ldots,5$, *and* $\theta(\xi,t) = \sum_{i=1}^{5} \theta_i(\xi,t) < 1$ *for all* $\xi \in S_2$, $t \in [0,T]$.

Lemma 3.2. *Let* $\theta_i = \theta_i(\xi,t)$, $i = 1,2,\ldots,5$, *be given continuous and nonnegative on* $S_2 \times [0,T]$ *functions such that* $\theta(x,t) = \sum_{i=1}^{5} \theta_i(\xi,t) \leq 1$ *for all* $\xi \in S_2$, $t \in [0,T]$. *Let* a_{i0}, $i = 1,2$, *and* ρ *satisfy Assumption* 3.1, *and* a_i, $i = 1,2$, *be a classical solution to problem* (3). *Then for all* $x \in \overline{\Omega}$, $t \in [0,T]$ *we have the inequalities*

$$0 \leq a_i(x,t) \leq \beta_i, \quad i = 1,2, \tag{5}$$

where the constants β_i *do not depend on concrete functions* $\theta_1,\ldots,\theta_5$ *satisfying the above conditions.*

4. Proof of a priori estimates. ***Proof of Lemma*** **3.1.** Let

$$\gamma = \Big\{(\theta_1,\theta_2,\theta_3,\theta_4,\theta_5) \in \mathbb{R}^5 : \theta_i = \theta_i(\xi,t),\ i = 1,2,\ldots,5,\ t \in [0,T]\Big\}$$

be a trajectory of system (2), which begins at the point $(\theta_{10}(\xi),\ \theta_{20}(\xi),\ \theta_{30}(\xi),\ \theta_{40}(\xi),\ \theta_{50}(\xi))$, $\xi \in S_2$. We prove that γ does not leave domain D, which is bounded by planes $\sum_{j=1}^{5} \theta_j = 1$ and $\theta_i = 0$, $i = 1,2,\ldots,5$.

Integrating equations (2) with respect to variable t, we get the integral equations

$$\begin{aligned}
&\theta_1 e^{\int_0^t (\kappa_{11}+\kappa_{13}\rho\theta_3)\,d\tau} = \theta_{10} + \int_0^t \kappa_1 a_1 (1-\theta) e^{\int_0^\tau (\kappa_{11}+\kappa_{13}\rho\theta_3)\,ds}\,d\tau,\\
&\theta_2 e^{(\kappa_{22}+\kappa_{22}^*)t} = \theta_{20} + \int_0^t \kappa_2 a_2 (1-\theta) e^{(\kappa_{22}+\kappa_{22}^*)\tau}\,d\tau,\\
&\theta_3 e^{\int_0^t \kappa_{13}\rho\theta_1 d\tau} = \theta_{30} + \int_0^t \kappa_{22}^* \theta_2 e^{\int_0^\tau \kappa_{13}\rho\theta_1 ds} d\tau,\\
&\theta_4 e^{\kappa_4 t} = \theta_{40} + \int_0^t \kappa_{22}^* \theta_2 e^{\kappa_4\tau}\,d\tau,\\
&\theta_5 e^{\kappa_5 t} = \theta_{50} + \int_0^t \kappa_{13}\rho\theta_1\theta_3 e^{\kappa_5\tau}\,d\tau.
\end{aligned} \tag{2*}$$

[1]For given continuous on $S_2 \times [0,T]$ functions a_i, $i = 1,2$, we say that functions θ_i, $i = 1,2,\ldots,5$, form a classical solution to Cauchy problem (2), if $\theta_i \in C^1(S_2 \times (0,T)) \cap C(S_2 \times [0,T])$ and they satisfy system (2).

Suppose that the trajectory γ leaves the domain D by crossing (or by touching) the plane $\theta_i = 0$ for any $i = 1, 2, \ldots, 5$ at the moment t^*, and does not cross the other planes, that is $\theta_i(\xi, t^*) = 0$, $\theta_j(\xi, t^*) > 0$ for $j \neq i$, and $\theta(\xi, t^*) < 1$. Then there exists $\varepsilon > 0$ such that $\theta_i(\xi, t) < 0$ for $t \in (t^*, t^* + \varepsilon]$ and $\theta_j(\xi, t^*) \geq 0$, $j \neq i$, $\theta(\xi, t) \leq 1$ for $t \in [t^*, t^* + \varepsilon]$. But for these t from the ith equation of system (2*) we get $\theta_i(\xi, t) \geq 0$. The contradiction shows that γ does not leaves this domain D through the plane $\theta_i = 0$. Similarly it can be shown that the trajectory γ does not leave the domain D through the intersection of several planes $\theta_i = 0$. For example, if the trajectory γ leaves the domain D through the intersection of planes $\theta_1 = 0$ and $\theta_2 = 0$ at the moment t^*, then there exists $\varepsilon > 0$ such that $\theta_1(\xi, t) < 0$ or $\theta_2(\xi, t) < 0$ and $\theta_j(\xi, t) > 0$, $j \neq 1, 2$, for $t \in (t^*, t^* + \varepsilon]$, $\theta(\xi, t) \leq 1$ for $t \in [t^*, t^* + \varepsilon]$. But from the first two equations of the system (2*) for these t we get $\theta_1(\xi, t) \geq 0$ and $\theta_2(\xi, t) \geq 0$. The contradiction shows that γ does not leaves this domain D through the intersection of planes $\theta_1 = 0$ and $\theta_2 = 0$. Thus $\theta_i(\xi, t) \geq 0$, $i = 1, 2, \ldots, 5$, for $\xi \in S_2$, $t \in [0, T]$.

Suppose that γ leaves the domain D by crosses or touching of the plane $\theta = 1$. By assumption, $\theta_0(\xi) < 1$. Then there exists the moment $t^* > 0$ such that $\theta_i(\xi, t) \geq 0$, $i = 1, 2, \ldots, 5$, $\theta(\xi, t) \leq 1$ for $t \in [0, t^*]$. Summing all equations of system (2) we get

$$-(1-\theta)' = (\kappa_1 a_1 + \kappa_2 a_2)(1-\theta) - \kappa_{11}\theta_1 - \kappa_{22}\theta_2 + \kappa_{22}^*\theta_2 - \kappa_4\theta_4 - \kappa_5\theta_5 - \kappa_{13}\rho\theta_1\theta_3. \qquad (6)$$

Multiplying both sides of this equation by $e^{\int_0^t (\kappa_1 a_1 + \kappa_2 a_2)\, ds}$ and integrating with respect to t we get the equation

$$(1-\theta)e^{\int_0^t (\kappa_1 a_1 + \kappa_2 a_2) ds} = 1 - \theta_0 + \int_0^t (\kappa_{11}\theta_1 + (\kappa_{22} - \kappa_{22}^*)\theta_2 +$$

$$+ \kappa_4\theta_4 + \kappa_5\theta_5 + \kappa_{13}\rho\theta_1\theta_3)e^{\int_0^\tau (\kappa_1 a_1 + \kappa_2 a_2) ds}\, d\tau.$$

By assumption of Lemma 3.1, $\theta_0(\xi) < 1$ and $\kappa_{22} \geq \kappa_{22}^*$. Then for $t = t^*$ the right-hand side of this equation is positive while the left one is equal to zero. The contradiction shows that γ does not leave domain D through the plane $\theta = 1$. Hence, $\theta(\xi, t) < 1$ for $\xi \in S_2$, $t \in [0, T]$.

Lemma 3.1 is proved.

***Proof of Lemma* 3.2.** According to the positivity lemma (see [8, p. 19], Chapter 1, Lemma 4.1), the functions a_i, $i = 1, 2$, in $\overline{\Omega} \times [0, T]$ cannot have a negative minimum. Therefore $a_i(x, t) \geq 0$ for all $x \in \overline{\Omega}$ and $t \in [0, T]$.

Let $\hat{a}_i$, $i = 1, 2$, be the solution to the problem

$$\frac{\partial \hat{a}_i}{\partial t} - k_i \Delta \hat{a}_i = 0 \qquad \text{in} \quad \Omega \times (0, T),$$

$$k_i \frac{\partial \hat{a}_i}{\partial \mathbf{n}} = 0 \qquad \text{on} \quad S_1 \times (0, T),$$

$$k_i \frac{\partial \hat{a}_i}{\partial \mathbf{n}} = \kappa_{ii}\rho \qquad \text{on} \quad S_2 \times (0, T),$$

$$\hat{a}_i|_{t=0} = a_{i0}, \quad i = 1, 2, \qquad \text{in} \quad \overline{\Omega}.$$

According to the positivity lemma function $\hat{a}_i - a_i$, $i = 1, 2$, in $\overline{\Omega} \times [0, T]$ cannot have a negative minimum. Therefore $a_i(x, t) \leq \hat{a}_i(x, t)$ for all $x \in \overline{\Omega}$ and $t \in [0, T]$ and $a_i(x, t) \leq \beta_i$ for all $x \in \overline{\Omega}$ and $t \in [0, T]$, where $\beta_i = \max_{x \in \overline{\Omega},\, t \in [0, T]} \hat{a}_i(x, t)$.

Lemma 3.2 is proved.

5. Uniqueness of classical solution.

Theorem 5.1. *Problem* (2), (3) *cannot have two different classical solutions.*

***Proof*.** Let $\hat{\theta}_i,\ i = 1, \ldots, 5, \hat{a}_i,\ i = 1, 2,$ and $\tilde{\theta}_i,\ i = 1, \ldots, 5,\ \tilde{a}_i,\ i = 1, 2,$ form two classical solutions to problem (2), (3). Set $\theta_i = \hat{\theta}_i - \tilde{\theta}_i,\ i = 1, \ldots, 5,\ a_i = \hat{a}_i - \tilde{a}_i,\ i = 1, 2,$ and $\theta = \sum_{i=1}^{5} \theta_i$. Then for $\theta_i,\ i = 1, \ldots, 5,$ we get Cauchy problem

$$\theta_1' = \kappa_1 a_1 (1 - \hat{\theta}) - \kappa_1 \tilde{a}_1 \theta - \kappa_{11} \theta_1 - \kappa_{13} \rho (\theta_1 \hat{\theta}_3 + \tilde{\theta}_1 \theta_3), \quad t \in (0, T),$$

$$\theta_2' = \kappa_2 a_2 (1 - \hat{\theta}) - \kappa_2 \tilde{a}_2 \theta - (\kappa_{22} + \kappa_{22}^*) \theta_2, \quad t \in (0, T),$$

$$\theta_3' = \kappa_{22}^* \theta_2 - \kappa_{13} \rho (\theta_1 \hat{\theta}_3 + \tilde{\theta}_1 \theta_3), \quad t \in (0, T),$$

$$\theta_4' = \kappa_{22}^* \theta_2 - \kappa_4 \theta_4, \quad t \in (0, T),$$

$$\theta_5' = \kappa_{13} \rho (\theta_1 \hat{\theta}_3 + \tilde{\theta}_1 \theta_3) - \kappa_5 \theta_5, \quad t \in (0, T),$$

$\theta_i|_{t=0} = 0,\ i = 1, 2, \ldots, 5,$ for $\xi \in S_2$. Integrating these equations with respect to variable t, we get the integral equations

$$\theta_1 = \int_0^t \kappa_1 a_1 (1 - \hat{\theta}) - \kappa_1 \tilde{a}_1 \theta - \kappa_{11} \theta_1 - \kappa_{13} \rho (\theta_1 \hat{\theta}_3 + \tilde{\theta}_1 \theta_3)\, ds, \quad t \in (0, T),$$

$$\theta_2 = \int_0^t \kappa_2 a_2 (1 - \hat{\theta}) - \kappa_2 \tilde{a}_2 \theta - (\kappa_{22} + \kappa_{22}^*) \theta_2\, ds, \quad t \in (0, T),$$

$$\theta_3 = \int_0^t \kappa_{22}^* \theta_2 - \kappa_{13} \rho (\theta_1 \hat{\theta}_3 + \tilde{\theta}_1 \theta_3)\, ds, \quad t \in (0, T),$$

$$\theta_4 = \int_0^t \kappa_{22}^* \theta_2 - \kappa_4 \theta_4\, ds, \quad t \in (0, T),$$

$$\theta_5 = \int_0^t \kappa_{13} \rho (\theta_1 \hat{\theta}_3 + \tilde{\theta}_1 \theta_3) - \kappa_5 \theta_5\, ds, \quad t \in (0, T).$$

Let $\|\theta\| = \sum_{i=1}^{5} |\theta_i|$. Then

$$\|\theta\| \leq \int_0^t (\kappa_1 |a_1| + \kappa_2 |a_2|)\, ds + C \int_0^t \|\theta\|\, ds,$$

where

$$C = \sum_{i=1}^{2} \kappa_i \tilde{m}_i + \max\{\kappa_{11} + 3\kappa_{13} \overline{\rho},\ \kappa_{22} + 3\kappa_{22}^*,\ \kappa_4, \kappa_5\},$$

$$\overline{\rho} = \max_{\xi \in S_2} \rho(\xi), \qquad \tilde{m}_i = \max_{\xi \in S_2,\, t \in [0,T]} |\tilde{a}_i(\xi,t)|, \quad i = 1,2.$$

Using the Gronwall lemma we get

$$\|\theta(\xi,t)\| \leq e^{Ct} \int\limits_0^t \big(\kappa_1 |a_1(\xi,s)| + \kappa_2 |a_2(\xi,s)|\big)\, ds. \tag{7}$$

Using that $\hat{\theta} \leq 1$ from (3) for each $i = 1,2$, we have

$$\frac{1}{2} \int\limits_\Omega a_i^2\, dx + \int\limits_0^\tau \int\limits_\Omega k_i |\nabla a_i|^2\, dx\, dt = \int\limits_0^\tau \int\limits_{S_2} \big(\kappa_i \rho(\tilde{a}_i \theta + (\hat{\theta} - 1) a_i) + \kappa_{ii} \rho \theta_i\big) a_i\, dS\, dt \leq$$

$$\leq C_1 \int\limits_0^\tau \int\limits_{S_2} \big(|a_1(\xi,t)| + |a_2(\xi,t)|\big) \|\theta(\xi,t)\|\, dS\, dt,$$

$$C_1 = \overline{\rho} \max_{i=1,2} \big\{\kappa_i \tilde{m}_i + \kappa_{ii}\big\}.$$

Adding these equalities and using the inequalities

$$\int\limits_0^\tau \int\limits_{S_2} |a_1(\xi,t)| \|\theta(\xi,t)\| dS dt \leq e^{C\tau} \int\limits_{S_2} \int\limits_0^\tau |a_1(\xi,t)| dt \int\limits_0^\tau (\kappa_1 |a_1(\xi,t)| + \kappa_2 |a_2(\xi,t)|) dt dS \leq$$

$$\leq \tau e^{C\tau} \left(\kappa_1 \int\limits_0^\tau \int\limits_{S_2} a_1^2(\xi,t)\, dS\, dt + \frac{\kappa_2}{2} \int\limits_0^\tau \int\limits_{S_2} \big(a_1^2(\xi,t) + a_2^2(\xi,t)\big)\, dS\, dt \right)$$

and

$$\int\limits_0^\tau \int\limits_{S_2} |a_2(\xi,t)| \|\theta(\xi,t)\| dS dt \leq e^{C\tau} \int\limits_{S_2} \int\limits_0^\tau |a_2(\xi,t)| dt \int\limits_0^\tau (\kappa_1 |a_1(\xi,t)| + \kappa_2 |a_2(\xi,t)|) dt\, dS \leq$$

$$\leq \tau e^{C\tau} \left(\kappa_2 \int\limits_0^\tau \int\limits_{S_2} a_2^2(\xi,t)\, dS\, dt + \frac{\kappa_1}{2} \int\limits_0^\tau \int\limits_{S_2} \big(a_1^2(\xi,t) + a_2^2(\xi,t)\big)\, dS\, dt \right)$$

we obtain

$$\frac{1}{2} \int\limits_\Omega \sum_{i=1}^2 a_i^2\, dx + \int\limits_0^\tau \int\limits_\Omega \sum_{i=1}^2 k_i |\nabla a_i|^2\, dx\, dt \leq C_2 \int\limits_0^\tau \int\limits_{S_2} \sum_{i=1}^2 a_i^2\, dS\, dt.$$

For every $\varepsilon > 0$, we have the estimate

$$\int\limits_S a^2\, dx \leq \varepsilon \int\limits_\Omega |\nabla a|^2\, dx + C_\varepsilon \int\limits_\Omega a^2\, dx,$$

where the constant C_ε is independent of the function a, and $C_\varepsilon \to \infty$ as $\varepsilon \to 0$. Therefore,

$$\int\limits_{\Omega} \sum_{i=1}^{2} a_i^2 \, dx + \int\limits_0^{\tau} \int\limits_{\Omega} \sum_{i=1}^{2} k_i |\nabla a_i|^2 \, dx \, dt \le C_2 \int\limits_0^{\tau} \int\limits_{\Omega} \sum_{i=1}^{2} a_i^2 \, dx \, dt.$$

From here by the Gronwall lemma we get

$$\int\limits_0^{\tau} \int\limits_{\Omega} \sum_{i=1}^{2} a_i^2 \, dx \, dt \le 0.$$

Hence, $a_i = 0$ for $i = 1, 2$. Now estimate (7) shows that $\theta_i = 0$ for $i = 1, 2, 3, 4, 5$.

Theorem 5.1 is proved.

6. Existence of classical solution. In this section, we prove that problem (2), (3) has a classical solution. Let $\Omega_0 = \Omega$ if $a_{10} = 0$ and $a_{20} = 0$ in some neighborhood of the surface S, and $\Omega_0 \supset \overline{\Omega}$ if a_{10} or a_{20} is continuously differentiable on some neighbourhood of the surface S. In the last case, we extend the functions a_{10} and a_{20} to $\Omega_0 \setminus \overline{\Omega}$ preserving the same smoothness. Let

$$\Gamma_k(x,t) = \frac{1}{\big(4\pi k t\big)^{n/2}} e^{-\frac{|x|^2}{4kt}}, \qquad x \in \mathbb{R}^n, \quad t > 0,$$

be the fundamental solution to the equation $a_t - k\Delta a = 0$, $k > 0$. Then, for any continuous on $S_2 \times [0,T]$ functions $\theta_1, \theta_2, \ldots, \theta_5$ and continuous on S function ρ, problem (3) has a unique solution $a_i \in C^{2,1}(\Omega \times (0,T]) \cap C(\overline{\Omega} \times [0,T])$, $i = 1, 2$, which can be presented by the formula (see [5])

$$a_i(x,t) = \int\limits_0^t \int\limits_S \Gamma_{k_i}(x-\xi, t-\tau) \varphi_i(\xi,\tau) \, dS_\xi \, d\tau + \int\limits_{\Omega_0} \Gamma_{k_i}(x-y,t) a_{i0}(y) \, dy, \tag{8}$$

where φ_i, $i = 1, 2$, is a continuous and bounded solution on $S \times [0,T]$ to the Volterra integral equation

$$\frac{1}{2}\varphi_i(\eta,t) + \int\limits_0^t \int\limits_S \left(\frac{\partial \Gamma_{k_i}(\eta-\xi,t-\tau)}{\partial \mathbf{n}_\eta} + \frac{1}{k_i}\sigma_i(\eta,t,\theta)\Gamma_{k_i}(\eta-\xi,t-\tau) \right) \varphi_i(\xi,\tau) \, dS_\xi d\tau =$$

$$= \frac{1}{k_i}\psi_i(\eta,t,\theta) - \int\limits_{\Omega_0} \left(\frac{\partial \Gamma_{k_i}(\eta-x,t)}{\partial \mathbf{n}_\eta} + \frac{1}{k_i}\sigma_i(\eta,t,\theta)\Gamma_{k_i}(\eta-x,t) \right) a_{i0}(x) \, dx, \tag{9}$$

$$\sigma_i(\xi,t,\theta) = \begin{cases} 0 & \text{if} \quad (\xi,t) \in S_1 \times [0,T], \\ \kappa_i \rho(\xi)\big(1-\theta(\xi,t)\big) & \text{if} \quad (\xi,t) \in S_2 \times [0,T], \end{cases}$$

$$\psi_i(\xi,t,\theta) = \begin{cases} 0 & \text{if} \quad (\xi,t) \in S_1 \times [0,T], \\ \kappa_{ii} \rho(\xi)\theta_i(\xi,t) & \text{if} \quad (\xi,t) \in S_2 \times [0,T], \end{cases}$$

$$\big|\varphi_i(\xi,t)\big| \le M_i, \qquad \xi \in S, \quad t \in [0,T].$$

Here constant M_i is independent of functions $\theta_1, \theta_2, \ldots, \theta_5$ such that $\theta_i(\xi, t) \geq 0,\ i = 1, 2, \ldots, 5,$ and $\theta(\xi, t) := \sum_{i=1}^{5} \theta_i(\xi, t) \leq 1$ for all $(\xi, t) \in S_2 \times [0, T]$.

Let $a_{i1},\ i = 1, 2,$ defined by formulas (8), and $\varphi_{i1},\ i = 1, 2,$ be solutions to problem (3) and the integral equation (9) with functions $\theta_i = \theta_{i0},\ i = 1, 2, \ldots, 5$. Then by Lemma 3.2 functions a_{i1}, $i = 1, 2$, are nonnegative, $a_{i1}(x, t) \leq \beta_i$ for $x \in \overline{\Omega}$ and $t \in [0, T]$, and

$$\left|\varphi_{i1}(\xi, t)\right| \leq M_i \qquad \text{for} \quad \xi \in S, t \in [0, T].$$

Assume that $\theta_{i1},\ i = 1, 2, \ldots, 5,$ form a solution to Cauchy problem (2) with $a_i = a_{i1},\ i = 1, 2$. Then by Lemma 3.1 functions $\theta_{i1},\ i = 1, 2, \ldots, 5,$ are nonnegative and $\sum_{i=1}^{5} \theta_{i1}(\xi, t) < 1$, for all $\xi \in S_2,\ t \in [0, T]$.

Let $a_{i2},\ i = 1, 2,$ defined by formulas (8), and $\varphi_{i2},\ i = 1, 2,$ be solutions to problem (3) and the integral equation (9) with functions $\theta_i = \theta_{i1},\ i = 1, 2, \ldots, 5$. Then by Lemma 3.2 functions a_{i2}, $i = 1, 2$, are nonnegative, $a_{i2}(x, t) \leq \beta_i$ for $x \in \overline{\Omega}$ and $t \in [0, T]$, and

$$\left|\varphi_{i2}(\xi, t)\right| \leq M_i \qquad \text{for} \quad \xi \in S, \quad t \in [0, T], \quad i = 1, 2.$$

Assume that $\theta_{i2},\ i = 1, 2, \ldots, 5,$ form a solution to Cauchy problem (2) with $a_i = a_{i2},\ i = 1, 2$. Then by Lemma 3.1 functions $\theta_{i2},\ i = 1, 2, \ldots, 5,$ are nonnegative and $\sum_{i=1}^{5} \theta_{i2}(\xi, t) < 1$ for all $\xi \in S,\ t \in [0, T]$.

Proceeding with this argument, we get the sequences

$$\{a_{ij}\}_{j=1}^{\infty}, \quad i = 1, 2, \qquad \{\varphi_{ij}\}_{j=1}^{\infty}, \quad i = 1, 2, \qquad \{\theta_{ij}\}_{j=1}^{\infty}, \quad i = 1, 2, \ldots, 5,$$

which are uniformly bounded:

$$a_{ij}(x, t) \geq 0 \qquad \text{for} \quad x \in \overline{\Omega}, \quad t \in [0, T], \quad i = 1, 2, \quad j = 1, 2, \ldots,$$

$$a_{ij}(x, t) \leq \beta_i \qquad \text{for} \quad x \in \overline{\Omega}, \quad t \in [0, T], \quad i = 1, 2, \quad j = 1, 2, \ldots,$$

$$|\varphi_{ij}(\xi, t)| \leq M_i \qquad \text{for} \quad \xi \in S, \quad t \in [0, T], \quad i = 1, 2, \quad j = 1, 2, \ldots,$$

$$\theta_{ij}(\xi, t) \geq 0 \qquad \text{for} \quad \xi \in S_2, \quad t \in [0, T], \quad i = 1, 2, \ldots, 5, \quad j = 1, 2, \ldots,$$

$$\sum_{i=1}^{5} \theta_{ij}(\xi, t) < 1 \qquad \text{for} \quad \xi \in S_2, \quad t \in [0, T], \quad j = 1, 2, \ldots.$$

Now we prove that they are equicontinuous. Functions a_{ij} are defined by the formula

$$a_{ij}(x, t) = \int_0^t \int_S \Gamma_{k_i}(x - \xi, t - \tau) \varphi_{ij}(\xi, \tau)\, dS_\xi\, d\tau + \int_{\Omega_0} \Gamma_{k_i}(x - y, t) a_{i0}(y)\, dy.$$

The potential of a simple layer (see [5] or [6])

$$\int_0^t \int_S \Gamma_{k_i}(x - \xi, t - \tau) \varphi_{ij}(\xi, \tau)\, dS_\xi\, d\tau$$

belongs to the Hölder space $C^\lambda\big(\overline{\Omega}\times[0,T]\big)$ with $\lambda\in(0,1)$. Hence, the sequences $\{a_{ij}\}_{j=1}^{\infty}$, $i=1,2$, are equicontinuous.

Functions θ_{ij}, $i=1,2,\ldots,5$, are solutions to the system of integral equations

$$\theta_{1j}(\xi,t)=\theta_{10}(\xi)+\int\limits_0^t\left[\kappa_1 a_{1j}(\xi,s)\left(1-\sum_{i=1}^5\theta_{ij}(\xi,s)\right)-\kappa_{11}\theta_{1j}(\xi,s)-\right.$$

$$\left.-\kappa_{13}\rho(\xi)\theta_{1j}(\xi,s)\theta_{3j}(\xi,s)\right]ds,$$

$$\theta_{2j}(\xi,t)=\theta_{20}(\xi)+\int\limits_0^t\left[\kappa_2 a_{2j}(\xi,s)\left(1-\sum_{i=1}^5\theta_{ij}(\xi,s)\right)-(\kappa_{22}+\kappa_{22}^*)\theta_{2j}(\xi,s)\right]ds,$$

$$\theta_{3j}(\xi,t)=\theta_{30}(\xi)+\int\limits_0^t\left[\kappa_{22}^*\theta_{2j}(\xi,s)-\kappa_{13}\rho(\xi)\theta_{1j}(\xi,s)\theta_{3j}(\xi,s)\right]ds,$$

$$\theta_{4j}(\xi,t)=\theta_{40}(\xi)+\int\limits_0^t\left[\kappa_{22}^*\theta_{2j}(\xi,s)-\kappa_4\theta_{4j}(\xi,s)\right]ds,$$

$$\theta_{5j}(\xi,t)=\theta_{50}(\xi)+\int\limits_0^t\left[\kappa_{13}\rho(\xi)\theta_{1j}(\xi,s)\theta_{3j}(\xi,s)-\kappa_5\theta_{5j}(\xi,s)\right]ds.$$

Therefore,

$$|\theta_{1j}(\xi,t)-\theta_{1j}(\xi,\tau)|\le|t-\tau|\big(\kappa_1\beta_1+\kappa_{11}+\kappa_{13}\overline{\rho}\big),\qquad \overline{\rho}=\max_{\xi\in S_2}\rho(\xi),$$

$$|\theta_{2j}(\xi,t)-\theta_{2j}(\xi,\tau)|\le|t-\tau|(\kappa_2\beta_2+\kappa_{22}+\kappa_{22}^*),$$

$$|\theta_{3j}(\xi,t)-\theta_{3j}(\xi,\tau)|\le|t-\tau|(\kappa_{22}^*+\kappa_{13}\overline{\rho}),$$

$$|\theta_{4j}(\xi,t)-\theta_{4j}(\xi,\tau)|\le|t-\tau|(\kappa_{22}^*+\kappa_4),$$

$$|\theta_{5j}(\xi,t)-\theta_{5j}(\xi,\tau)|\le|t-\tau|(\kappa_{13}\overline{\rho}+\kappa_5).$$

Moreover,

$$\sum_{i=1}^5|\theta_{ij}(\xi,t)-\theta_{ij}(\eta,t)|\le\sum_{i=1}^5|\theta_{i0}(\xi)-\theta_{i0}(\eta)|+3\kappa_{13}T|\rho(\xi)-\rho(\eta)|+$$

$$+C\int\limits_0^t\sum_{i=1}^5|\theta_{ij}(\xi,s)-\theta_{ij}(\eta,s)|\,ds+\int\limits_0^t\sum_{i=1}^2\kappa_i|a_{ij}(\xi,s)-a_{ij}(\eta,s)|\,ds$$

and

$$\sum_{i=1}^5|\theta_{ij}(\xi,t)-\theta_{ij}(\eta,t)|\le e^{CT}\left(\sum_{i=1}^5|\theta_{i0}(\xi)-\theta_{i0}(\eta)|+3\kappa_{13}T|\rho(\xi)-\rho(\eta)|\right)+$$

$$+\frac{e^{CT}-1}{C}\max_{s\in[0,T]}\sum_{i=1}^{2}\kappa_i\big|a_{ij}(\xi,s)-a_{ij}(\eta,s)\big|$$

for all $\xi,\eta\in S_2,\ t,\tau\in[0,T]$. Here $C=\overline{m}+\max\left\{\kappa_{11}+3\overline{\rho}\kappa_{13},\kappa_{22}+3\kappa_{22}^*,\kappa_4,\kappa_5\right\}$. These estimates show that the sequence $\{\theta_{ij}\}_{j=1}^{\infty},\ i=1,2,\ldots,5$, is equicontinuous.

Functions $\varphi_{ij},\ i=1,2$, are solutions to integral equation (9) with $\theta_i=\theta_{ij-1}$. The potential of a double-layer (see [5] or [6]),

$$\int_0^t\int_S\frac{\partial\Gamma_{k_i}(\eta-\xi,t-\tau)}{\partial\mathbf{n}_\eta}\varphi_{ij}(\xi,\tau)\,dS_\xi\,d\tau$$

belongs to the Hölder space $C^\lambda(S\times[0,T])$ with $\lambda<2\alpha/3$. Therefore, the sequences $\{\varphi_{ij}\}_{j=1}^{\infty}$, $i=1,2$, are equicontinuous. According to the Arzelà – Ascoli theorem we can select uniformly converging subsequences from sequences $\{a_{ij}\}_{j=1}^{\infty},\ i=1,2,\ \{\varphi_{ij}\}_{j=1}^{\infty},\ i=1,2$, and $\{\theta_{ij}\}_{j=1}^{\infty}$, $i=1,2,\ldots,5$. Since problem (2), (3) cannot possess two classical solutions, we claim that the sequences $\{a_{ij}\}_{j=1}^{\infty},\ i=1,2,\ \{\varphi_{ij}\}_{j=1}^{\infty},\ i=1,2$, and $\{\theta_{ij}\}_{j=1}^{\infty},\ i=1,2,\ldots,5$, converge uniformly. Set

$$a_i(x,t)=\lim_{j\to\infty}a_{ij}(x,t),\qquad x\in\overline{\Omega},\quad t\in[0,T],\quad i=1,2,$$

$$\varphi_i(\xi,t)=\lim_{j\to\infty}\varphi_{ij}(\xi,t),\qquad \xi\in S,\quad t\in[0,T],\quad i=1,2,$$

$$\theta_i(\xi,t)=\lim_{j\to\infty}\theta_{ij}(\xi,t),\qquad \xi\in S_2,\quad t\in[0,T],\quad i=1,2,\ldots,5.$$

Formula (8) holds for the limit functions $a_i,\ i=1,2$. Therefore, the limit functions $a_i\in C^{2,1}(\Omega\times\times(0,T])\cap C(\overline{\Omega}\times[0,T])$ are solutions to problem (3). The limit functions $\theta_i,\ i=1,2,\ldots,5$, are solutions to the system of integral equations

$$\theta_1(\xi,t)=\theta_{10}(\xi)+\int_0^t\left[\kappa_1a_1(\xi,s)\left(1-\sum_{i=1}^5\theta_i(\xi,s)\right)-\kappa_{11}\theta_1(\xi,s)-\kappa_{13}\rho(\xi)\theta_1(\xi,s)\theta_3(\xi,s)\right]ds,$$

$$\theta_2(\xi,t)=\theta_{20}(\xi)+\int_0^t\left[\kappa_2a_2(\xi,s)\left(1-\sum_{i=1}^5\theta_i(\xi,s)\right)-(\kappa_{22}+\kappa_{22}^*)\theta_{2j}(\xi,s)\right]ds,$$

$$\theta_3(\xi,t)=\theta_{30}(\xi)+\int_0^t\left[\kappa_{22}^*\theta_2(\xi,s)-\kappa_{13}\rho(\xi)\theta_1(\xi,s)\theta_3(\xi,s)\right]ds,$$

$$\theta_4(\xi,t)=\theta_{40}(\xi)+\int_0^t\left[\kappa_{22}^*\theta_2(\xi,s)-\kappa_4\theta_4(\xi,s)\right]ds,$$

$$\theta_5(\xi,t)=\theta_{50}(\xi)+\int_0^t\left[\kappa_{13}\rho(\xi)\theta_1(\xi,s)\theta_3(\xi,s)-\kappa_5\theta_5(\xi,s)\right]ds.$$

Therefore, θ_i, $i = 1, 2, \ldots, 5$, are uniformly differentiable with respect to variable t and form a solution to Cauchy problem (2). Hence, problem (2), (3) has a classical solution. According to Theorem 5.1, this solution is unique.

Acknowledgment. The author thanks Prof. V. Skakauskas for the formulation of the problem (system (2) – (4)) and fruitful discussions.

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Received 20.04.15,
after revision — 06.06.17