

Existence and uniqueness theorem to a unimolecular heterogeneous catalytic reaction model

A. Ambrazevičius

Faculty of Mathematics and Informatics of Vilnius University
Naugarduko str. 24, LT-03225 Vilnius, Lithuania
algirdas.ambrazevicius@mif.vu.lt

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Abstract. A mathematical model for an unimolecular heterogeneous catalytic reaction is considered in the case when the reaction product slowly desorb and then diffuse away from the surface. This model is described by a coupled system of nonlinear parabolic and two ordinary differential equations. The existence and uniqueness theorems of classical solution are proved for this system.

Keywords: parabolic equations, ordinary differential equations, heterogeneous catalysis, reaction-diffusion system.

1 Introduction and formulation of the problem

The process of the bulk diffusion and heterogeneous chemical reactions is modelled by coupled systems of parabolic and ordinary differential equations (see for example [1], [2], and [3] where similar models are studied). In [4], a mathematical model for an unimolecular heterogeneous catalytic reaction of type $A \rightarrow B$ is proposed, where A is a reactant and B is a product of this reaction. According to Langmuir [5], molecules of the reactant A bind to active sites of the surface of a catalyst (adsorbent) K to form an intermediate (adsorbate) that subsequently gives the finite product B . In [1] we proved the existence and uniqueness theorem to a surface reaction model given in [4] taking into account the bulk diffusion of A , adsorption and desorption of A by de surface of K , decay of AK , and *instantaneous desorption* of product B from the surface.

In the present paper we consider the other model given in [4] which in addition to the adsorption and desorption of A includes the *slow desorption* of product B , and does not allow to diffuse for adsorbate AK and product B along the surface of the adsorbent. The model also includes the diffusion of A from reaction environment to the adsorbent and diffusion of B into reaction environment from the adsorbent.

Suppose that reactant A occupies a bounded domain $\Omega \subset \mathbb{R}^n$, $n \geq 3$, $a = a(x, t)$ is the concentration of A at the point $x \in \Omega$ at time t , $S := \partial\Omega$ is a surface of dimension $n - 1$, of class $C^{1+\alpha}$, $\alpha \in (0, 1)$, S_2 is a connected and closed part of S such that the

Hausdorff measure $\mathcal{H}^{n-1}(S_2) > 0$ or a finite number of such parts of the surface S (surface of the adsorbent), $S_1 = S \setminus S_2$ (see Fig. 1), $\rho \in C(S)$, $\rho(x) \geq 0$ for $x \in S$, $\rho(x)$ is a concentration of active sites of the adsorbent at point $x \in S_2$, $\rho(x) = 0$ for $x \in S_1$, $\theta_1 = \theta_1(x, t)$ is a fraction of ρ such that $\theta_1\rho$ is a density of active sites of the surface occupied by molecules of reactant A at point $x \in S_2$ at time t , $\theta_2 = \theta_2(x, t)$ is a fraction of ρ such that $\theta_2\rho$ is a density of active sites of the surface occupied by molecules of the product B at point $x \in S_2$ at time t , $1 - \theta_1 - \theta_2$ is a free fraction of ρ , (then $\rho(1 - \theta_1 - \theta_2)$ is the concentration of free active sites), $b = b(x, t)$ is a concentration of product B at point $x \in \Omega$ at time t , κ and κ_1 are the adsorption and desorption rate constants, κ_2 is the reaction rate of the intermediate AK , and κ_3 is the desorption rate constant for the product B .

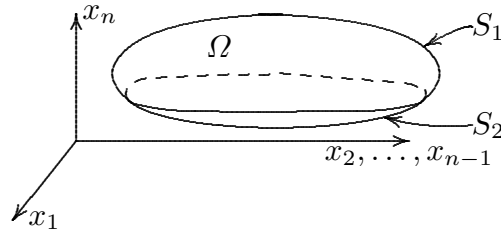


Fig. 1. Domain of definition of a and b .

According to Langmuir [5], adsorption, desorption and reaction rates of reactant A can be written as $\kappa\rho(1 - \theta_1 - \theta_2)a$, $\kappa_1\rho\theta_1$, and $\kappa_2\rho\theta_1$. Similarly, the desorption rate of product B can be written as $\kappa_3\rho\theta_2$. Therefore, the diffusion of reactant A can be described by the problem

$$\begin{cases} a_t - k\Delta a = 0 & \text{in } \Omega \times (0, T), \\ k \frac{\partial a}{\partial \mathbf{n}} = 0 & \text{on } S_1 \times (0, T), \\ k \frac{\partial a}{\partial \mathbf{n}} + \kappa\rho(1 - \theta_1 - \theta_2)a = \kappa_1\rho\theta_1 & \text{on } S_2 \times (0, T), \\ a|_{t=0} = a_0 & \text{in } \overline{\Omega}, \end{cases} \quad (1)$$

where $k = \text{const} > 0$ is a diffusion coefficient, $\partial a / \partial \mathbf{n}$ is the outward normal derivative to S , $\Delta a = \sum_{i=1}^n a_{x_i x_i}$, $a_0 = a_0(x)$ is the initial concentration of A at point $x \in \overline{\Omega}$.

For θ_1 and θ_2 , we have the Cauchy problem

$$\begin{cases} \theta_1' = \kappa(1 - \theta_1 - \theta_2)a - (\kappa_1 + \kappa_2)\theta_1, & \theta_1|_{t=0} = \theta_{10}, & x \in S_2, \\ \theta_2' = \kappa_2\theta_1 - \kappa_3\theta_2, & \theta_2|_{t=0} = \theta_{20}(x), & x \in S_2, \end{cases} \quad (2)$$

where $\theta_{10} = \theta_{10}(x) \geq 0$, $x \in S_2$ is the initial value of θ_1 , $\theta_{20} = \theta_{20}(x) \geq 0$ is the initial value of θ_2 , $\theta_{10}(x) + \theta_{20}(x) < 1$ for all $x \in S_2$.

The diffusion of the product B is described by the problem

$$\begin{cases} b_t - l\Delta b = 0 & \text{in } \Omega \times (0, T), \\ \frac{\partial b}{\partial \mathbf{n}} = 0 & \text{on } S_1 \times (0, T), \\ l \frac{\partial b}{\partial \mathbf{n}} = \kappa_3 \rho \theta_2 & \text{on } S_2 \times (0, T), \\ b|_{t=0} = b_0 & \text{in } \overline{\Omega}, \end{cases} \quad (3)$$

where $l = \text{const} > 0$ is a diffusion coefficient of the product B , $\partial b / \partial \mathbf{n}$ is the outward normal derivative to S , $\Delta b = \sum_{i=1}^n b_{x_i x_i}$, $b_0 = b_0(x)$ is the initial concentration of B at point $x \in \overline{\Omega}$.

Therefore, the unimolecular heterogeneous catalytic reaction described above we model by system (1), (2), and (3).

In the present paper, we prove the existence and uniqueness of the classical solution to problem (1), (2), and (3).

Definition 1. Functions a , θ_1 , θ_2 and b are classical solutions to problem (1), (2), and (3) if a and $b \in C^{2,1}(\Omega \times (0, T]) \cap C(\overline{\Omega} \times [0, T])$ and $\partial a / \partial \mathbf{n}$ and $\partial b / \partial \mathbf{n}$ are continuous on $S \times [0, T]$, $S = S_1 \cup S_2$, while $\theta_1, \theta_2 \in C([0, T] \times S_2)$, $\theta'_1, \theta'_2 \in C((0, T] \times S_2)$.

Remark 1. Results of this paper are not valid for $n = 1, 2$. The cases where $n = 1$ and $n = 2$ have to be studied separately.

The paper is organized as follows. In Section 1, we describe the model. In Section 2, we give a priori estimates. Section 3 is devoted to existence and uniqueness of the classical solution to problem (1), (2), and (3).

2 A priori estimates

Lemma 1. Let function $a = a(x, t)$ be continuous and nonnegative on $S_2 \times [0, T]$, θ_{10} and θ_{20} be continuous on S_2 and such that $\theta_{10}(x) + \theta_{20}(x) < 1$ for all $x \in S_2$. Let θ_1 and θ_2 be a solution of Cauchy problem (2). Then, for all $x \in S_2$ and $t \in [0, T]$, the following estimates are true:

$$\begin{aligned} \theta_1(x, t) &\geq \theta_{10}(x) e^{-\kappa \int_0^t a(x, s) ds - (\kappa_1 + \kappa_2)t} \geq 0, & \theta_2(t, x) &\geq 0, \\ \theta_1(x, t) &\leq 1 - (1 - \theta_{10}(x)) e^{-\int_0^t \kappa a(x, s) ds} < 1, \\ 1 - \theta_1(x, t) - \theta_2(x, t) &\geq (1 - \theta_{10}(x) - \theta_{20}(x)) e^{-\kappa \int_0^t a(x, s) ds - \kappa_2 t} > 0. \end{aligned}$$

Proof. Let $\gamma = \{(\theta_1, \theta_2): \theta_1 = \theta_1(t, x), \theta_2 = \theta_2(t, x), t \in [0, T], x \in S_2\}$ be a trajectory of system (2), which begins at the point $(\theta_{10}, \theta_{20})$ (see Fig. 2). We prove that γ does

not leave the triangle with vertices A, O, B . First, we note that inside of the triangle with vertices O, C, B , the derivative $\theta'_2 > 0$ and therefore θ_2 increases as t increases, but inside of the triangle with vertices O, A, C the derivative $\theta'_2 < 0$ and therefore θ_2 decreases as t increases.

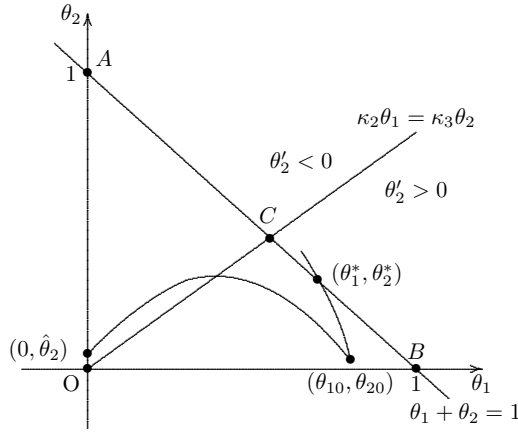


Fig. 2. Shema of trajectory.

Suppose, that γ crosses or touches the line CB at the point (θ_1^*, θ_2^*) . Then at this point

$$\frac{d\theta_2}{d\theta_1} = \frac{\kappa_2 \theta_1^* - \kappa_3 \theta_2^*}{-(\kappa_1 + \kappa_2) \theta_1^*} < 0.$$

Hence,

$$-1 \geq \frac{d\theta_2}{d\theta_1} = \frac{\kappa_2 \theta_1^* - \kappa_3 \theta_2^*}{-(\kappa_1 + \kappa_2) \theta_1^*} = -\frac{\kappa_2}{\kappa_1 + \kappa_2} + \frac{\kappa_3}{\kappa_1 + \kappa_2} \frac{\theta_2^*}{\theta_1^*} > -1.$$

The contradiction shows that γ does not either cross or touch the line CB .

Inside of the triangle with vertices O, C, B we have $\theta'_2 > 0$. Therefore θ_2 increases as t increases and therefore the trajectory γ does not cross line OB . Hence $\theta_2 \geq 0$. If trajectory γ crosses line OC , then at the point of intersection $\theta'_2 = 0$. Inside of the triangle with vertices O, A, C , derivative $\theta'_2 < 0$. Therefore inside of this triangle, θ_2 decreases as t increases. If trajectory γ crosses the line OA at the point $(0, \hat{\theta}_2)$, $\hat{\theta}_2 \in (0, 1)$, then at this point

$$0 < \frac{d\theta_2}{d\theta_1} = -\frac{\kappa_3 \hat{\theta}_2}{\kappa(1 - \hat{\theta}_2)a} \leq 0.$$

If $a = 0$, then trajectory γ touches line AO . Therefore trajectory γ does not cross line OA . Trajectory γ cannot cross point O as well, because at this point derivative $\theta'_2 = 0$.

At last, γ can reach point O only by touching line OA . Therefore, γ remains inside of the triangle with vertex A, O, B , i.e., $\theta_1(x, t) \geq 0$, $\theta_2(x, t) \geq 0$, $\theta_1(x, t) + \theta_2(x, t) < 1$ for $t > 0, x \in S_2$.

Solving the first equation of problem (2) with respect to θ_1 and $1 - \theta_1$ we get:

$$F(x, t)\theta_1(x, t) = \theta_{10}(x) + \int_0^t \kappa a(x, s)(1 - \theta_2(x, s))F(x, s) ds, \quad (4)$$

$$\begin{aligned} F(x, t)(1 - \theta_1(x, t)) &= (1 - \theta_{10}(x)) + \int_0^t (\kappa a(x, s)\theta_2(x, s) + \kappa_1 + \kappa_2)F(x, s) ds, \end{aligned} \quad (5)$$

where $F(x, t) = e^{(\kappa_1 + \kappa_2)t + \int_0^t \kappa a(x, s) ds}$. Hence,

$$0 \leq \theta_{10}(x)e^{-(\kappa_1 + \kappa_2)t} e^{-\int_0^t \kappa a(x, s) ds} \leq \theta_1(x, t) < 1 \quad \text{for all } x \in S_2, t \in [0, T],$$

since $0 \leq \theta_{10}(x) < 1$ for all $x \in S_2$. Moreover,

$$F(x, t) \geq (1 - \theta_{10}(x)) + (\kappa_1 + \kappa_2) \int_0^t F(x, s) ds.$$

From here by the Gronwall lemma we get the estimate

$$(\kappa_1 + \kappa_2) \int_0^t F(x, s) ds \geq (1 - \theta_{10}(x))(e^{(\kappa_1 + \kappa_2)t} - 1).$$

and then from (5) it follows that

$$F(x, t)(1 - \theta_1(x, t)) \geq (1 - \theta_{10}(x))e^{(\kappa_1 + \kappa_2)t}.$$

Hence,

$$\theta_1(x, t) \leq 1 - (1 - \theta_{10}(x))e^{-\int_0^t \kappa a(x, s) ds} \quad \text{for all } x \in S_2, t \in [0, T].$$

Solving $1 - \theta_1 - \theta_2$ from equations (2) we get

$$\begin{aligned} &F(x, t)(1 - \theta_1(x, t) - \theta_2(x, t)) \\ &= 1 - \theta_{10}(x) - \theta_{20}(x) \\ &\quad + \int_0^t \{ \kappa_1\theta_1(x, s) + \kappa_3\theta_2(x, s) + (\kappa_1 + \kappa_2)(1 - \theta_1(x, t) - \theta_2(x, t)) \} F(x, s) ds \\ &\geq 1 - \theta_{10}(x) - \theta_{20}(x) + \kappa_1 \int_0^t (1 - \theta_2(x, s))F(x, s) ds. \end{aligned}$$

From here it follows that

$$F(x, t)(1 - \theta_2(x, t)) \geq 1 - \theta_{10}(x) - \theta_{20}(x) + \kappa_1 \int_0^t (1 - \theta_2(x, s))F(x, s) \, ds.$$

Hence,

$$\kappa_1 \int_0^t (1 - \theta_2(x, s))F(x, s) \, ds \geq (1 - \theta_{10}(x) - \theta_{20}(x))(e^{\kappa_1 t} - 1).$$

Now, two last inequalities show that

$$F(x, t)(1 - \theta_1(x, t) - \theta_2(x, t)) \geq (1 - \theta_{10}(x) - \theta_{20}(x))e^{\kappa_1 t}.$$

The proof is complete. □

Corollary 1. *Under the conditions of Lemma 1 the following inequality is true*

$$\frac{\theta_1(x, t)}{1 - \theta_1(x, t) - \theta_2(x, t)} \leq \max \left\{ \frac{\theta_{10}(x)}{1 - \theta_{10}(x) - \theta_{20}(x)}, \frac{\kappa m}{\kappa_1} \right\}, \quad (6)$$

where $m = \max_{(x, t) \in S_2 \times [0, T]} a(x, t)$.

This inequality follows from the estimate

$$\begin{aligned} & \frac{\theta_1(x, t)}{1 - \theta_1(x, t) - \theta_2(x, t)} \\ & \leq \frac{\theta_{10}(x) + \kappa m \int_0^t (1 - \theta_2(x, s))F(x, s) \, ds}{1 - \theta_{10}(x) - \theta_{20}(x) + \kappa_1 \int_0^t (1 - \theta_2(x, s))F(x, s) \, ds} \\ & \leq \begin{cases} \frac{\kappa m}{\kappa_1}, & \text{as } \frac{\theta_{10}(x)}{1 - \theta_{10}(x) - \theta_{20}(x)} \leq \frac{\kappa m}{\kappa_1}, \\ \frac{\theta_{10}(x)}{1 - \theta_{10}(x) - \theta_{20}(x)}, & \text{as } \frac{\theta_{10}(x)}{1 - \theta_{10}(x) - \theta_{20}(x)} \geq \frac{\kappa m}{\kappa_1}. \end{cases} \end{aligned}$$

Lemma 2. *Let θ_1 and θ_2 be continuous on $S_2 \times [0, T]$ and $\theta_1(x, t) \geq 0$, $\theta_2(x, t) \geq 0$, $\theta_1(x, t) + \theta_2(x, t) < 1$ for all $x \in S_2$, $t \in [0, T]$. Let $0 \leq a_0 \in C(\overline{\Omega})$ and a be a classical solution of problem (1). Then*

$$0 \leq a(x, t) \leq \max \left\{ \max_{x \in \overline{\Omega}} a_0(x), \frac{\kappa_1}{\kappa} \max_{x \in S_2, t \in [0, T]} \frac{\theta_1(x, t)}{1 - \theta_1(x, t) - \theta_2(x, t)} \right\} \quad (7)$$

for all $x \in \overline{\Omega}$, $t \in [0, T]$.

Proof. Applying the positivity lemma (see [6], Chapter 2, Lemma 2.1, p. 54) to problem (1) we get

$$a(x, t) \geq 0 \quad \text{for all } (x, t) \in \overline{\Omega} \times [0, T].$$

Inserting $a = m - v$,

$$m = \max \left\{ \max_{x \in \overline{\Omega}} a_0(x), \frac{\kappa_1}{\kappa} \max_{x \in S_2, t \in [0, T]} \frac{\theta_1(x, t)}{1 - \theta_1(x, t) - \theta_2(x, t)} \right\},$$

into problem (1) and using the same lemma, we get the estimate

$$v = m - a(x, t) \geq 0 \quad \text{for all } (x, t) \in \overline{\Omega} \times [0, T].$$

The proof is complete. \square

Remark 2. Let θ_2 and ρ from problem (3) be nonnegative continuous functions, $b_0(x) \geq 0$ for all $x \in \overline{\Omega}$, and b be a classical solution of problem (3). By [6] (see Lemma 4.1, p. 19) $b(x, t) \geq 0$ for all $x \in \overline{\Omega}$, $t \in [0, T]$.

Let a, θ_1, θ_2 and b be a classical solution of problem (1), (2), and (3). Then (see [4])

$$\begin{aligned} & \int_{\Omega} (a(x, t) + b(x, t)) \, dx + \int_{S_2} \rho(x) (\theta_1(x, t) + \theta_2(x, t)) \, dS \\ &= \int_{\Omega} (a_0(x) + b_0(x)) \, dx + \int_{S_2} \rho(x) (\theta_{10}(x) + \theta_{20}(x)) \, dS. \end{aligned} \quad (8)$$

To prove this law it is sufficient to add equations (1) and (3), then integrate over cylinder $Q_t = \Omega \times (0, t)$, apply the formula of integration by parts, and use equation (2), boundary and initial conditions.

3 Existence and uniqueness of the solution

Theorem 1. *Problem (1), (2), (3) has at most one classical solution.*

Proof. We multiply equation (1) by a smooth function η and integrate the result over cylinder $Q_\tau = \Omega \times (0, \tau)$, $\tau \in (0, T]$ getting an identity which, by using the formula of integration by parts and taking into account the boundary condition, can be written as follows

$$\int_{Q_\tau} a_t \eta \, dx \, dt + k \int_{Q_\tau} a_x \eta_x \, dx \, dt = \int_0^\tau \int_{S_2} (\kappa \rho (\theta_1 + \theta_2 - 1) a + \kappa_1 \rho \theta_1) \eta \, dS \, dt. \quad (9)$$

Let \tilde{a} , $\tilde{\theta}_1$, $\tilde{\theta}_2$, \tilde{b} and \hat{a} , $\hat{\theta}_1$, $\hat{\theta}_2$, \hat{b} be two classical solutions of problem (1), (2), (3). Set $a = \tilde{a} - \hat{a}$, $\theta_1 = \tilde{\theta}_1 - \hat{\theta}_1$, $\theta_2 = \tilde{\theta}_2 - \hat{\theta}_2$, $b = \tilde{b} - \hat{b}$. Then, for pairs \tilde{a} , $\tilde{\theta}_1$, $\tilde{\theta}_2$ and \hat{a} , $\hat{\theta}_1$, $\hat{\theta}_2$, integral identity (9) with $\eta = a$ is true. Hence

$$\begin{aligned} & \int_{Q_\tau} a_t a \, dx \, dt + k \int_{Q_\tau} a_x^2 \, dx \, dt \\ &= \int_0^\tau \int_{S_2} [\kappa \rho (\tilde{\theta}_1 + \tilde{\theta}_2 - 1) \tilde{a} + \kappa_1 \rho \tilde{\theta}_1 - \kappa \rho (\hat{\theta}_1 + \hat{\theta}_2 - 1) \hat{a} - \kappa_1 \rho \hat{\theta}_1] a \, dS \, dt. \end{aligned}$$

This equality can be rewritten as follows

$$\begin{aligned} & \frac{1}{2} \int_{\Omega} a^2 \, dx |^{t=\tau} + k \int_{Q_\tau} a_x^2 \, dx \, dt \\ &= \int_0^\tau \int_{S_2} [\kappa \rho (\theta_1 + \theta_2) \tilde{a} + \kappa \rho (\hat{\theta}_1 + \hat{\theta}_2 - 1) a + \kappa_1 \rho \theta_1] a \, dS \, dt. \end{aligned} \quad (10)$$

By using Eq. (4) we get

$$\begin{aligned} \theta_1(x, t) &= \tilde{\theta}_1(x, t) - \hat{\theta}_1(x, t) \\ &= \theta_{10}(x) e^{-(\kappa_1 + \kappa_2)t} \left(e^{-\kappa \int_0^t \tilde{a}(x, s) \, ds} - e^{-\kappa \int_0^t \hat{a}(x, s) \, ds} \right) \\ &\quad + \kappa \int_0^t e^{-(\kappa_1 + \kappa_2)(t-\tau)} \left(\tilde{a}(x, s) (1 - \tilde{\theta}_2(x, s)) e^{-\kappa \int_\tau^t \tilde{a}(x, s) \, ds} \right. \\ &\quad \left. - \hat{a}(x, s) (1 - \hat{\theta}_2(x, s)) e^{-\kappa \int_\tau^t \hat{a}(x, s) \, ds} \right) d\tau \\ &= \theta_{10}(x) e^{-(\kappa_1 + \kappa_2)t} \left(e^{-\kappa \int_0^t \tilde{a}(x, s) \, ds} - e^{-\kappa \int_0^t \hat{a}(x, s) \, ds} \right) \\ &\quad + \kappa \int_0^t e^{-(\kappa_1 + \kappa_2)(t-\tau)} \left(a(x, s) (1 - \tilde{\theta}_2(x, s)) e^{-\kappa \int_\tau^t \tilde{a}(x, s) \, ds} \right. \\ &\quad \left. - \hat{a}(x, s) \theta_2(x, s) e^{-\kappa \int_\tau^t \tilde{a}(x, s) \, ds} \right. \\ &\quad \left. + \hat{a}(x, s) (1 - \hat{\theta}_2(x, s)) \left(e^{-\kappa \int_\tau^t \tilde{a}(x, s) \, ds} - e^{-\kappa \int_\tau^t \hat{a}(x, s) \, ds} \right) \right) d\tau \end{aligned}$$

and then

$$|\theta_1(x, t)| \leq \kappa \theta_0(x) e^{-(\kappa_1 + \kappa_2)t} \int_0^t |a(x, s)| \, ds$$

$$\begin{aligned}
 & + \kappa \int_0^t e^{-(\kappa_1 + \kappa_2)(t-\tau) - \kappa \int_\tau^t \tilde{a}(x,s) ds} |a(x, \tau)| d\tau \\
 & + m\kappa \int_0^t |\theta_2(x, \tau)| e^{-(\kappa_1 + \kappa_2)(t-\tau)} d\tau + m\kappa^2 \int_0^t e^{-(\kappa_1 + \kappa_2)(t-\tau)} \int_\tau^t |a(x, s)| ds d\tau.
 \end{aligned}$$

Using the formula of integration by parts we get

$$\int_0^t e^{-(\kappa_1 + \kappa_2)(t-\tau)} \int_\tau^t |a(x, s)| ds d\tau \leq \frac{1}{\kappa_1 + \kappa_2} \int_0^t |a(x, s)| ds.$$

From the second equation of system (2) we get

$$\begin{aligned}
 \theta_2(x, t) & = \tilde{\theta}_2(x, t) - \hat{\theta}_2(x, t) = \kappa_2 \int_0^t e^{-\kappa_3(t-\tau)} (\tilde{\theta}_1(x, \tau) - \hat{\theta}_1(x, \tau)) d\tau \\
 & = \kappa_2 \int_0^t e^{-\kappa_3(t-\tau)} \theta_1(x, \tau) d\tau.
 \end{aligned} \tag{11}$$

By integration by parts we get

$$\begin{aligned}
 \int_0^t |\theta_2(x, \tau)| e^{-(\kappa_1 + \kappa_2)(t-\tau)} d\tau & \leq \kappa_2 \int_0^t e^{-(\kappa_1 + \kappa_2)(t-\tau)} \int_0^\tau e^{-\kappa_3(\tau-s)} |\theta_1(x, t)| ds d\tau \\
 & \leq \frac{\kappa_2}{\kappa_1 + \kappa_2} \int_0^t |\theta_1(x, s)| ds.
 \end{aligned}$$

Therefore,

$$|\theta_1(x, t)| \leq \left(2\kappa + \frac{m\kappa^2}{\kappa_1 + \kappa_2} \right) \int_0^t |a(x, s)| ds + \frac{m\kappa\kappa_2}{\kappa_1 + \kappa_2} \int_0^t |\theta_1(x, s)| ds.$$

From here, by the Gronwall lemma, we get the estimate

$$\begin{aligned}
 |\theta_1(x, t)| & \leq e^{\frac{m\kappa\kappa_2}{\kappa_1 + \kappa_2} t} \left(2\kappa + \frac{m\kappa^2}{\kappa_1 + \kappa_2} \right) \int_0^t |a(x, s)| ds \\
 & \leq C e^{\kappa m t} \int_0^t |a(x, s)| ds;
 \end{aligned} \tag{12}$$

where $C = 2\kappa + \frac{m\kappa^2}{\kappa_1 + \kappa_2}$. Using these estimates we evaluate integrals of the right-hand side of equation (10). We have

$$\begin{aligned} \int_0^\tau \kappa\theta_1(x, t)\tilde{a}(x, t)a(x, t) dt &\leq \kappa m \int_0^\tau |\theta_1(x, t)||a(x, t)| dt \\ &\leq C \int_0^\tau e^{\kappa m t} \left(\int_0^t |a(x, s)| ds \right) |a(x, t)| dt \leq \frac{C}{2} e^{\kappa m \tau} \left(\int_0^\tau |a(x, s)| ds \right)^2 \\ &\leq \frac{C}{2} e^{\kappa m \tau} \tau \int_0^\tau a^2(x, s) ds. \end{aligned}$$

From Eq. (11) and inequality (12) it follows that

$$\begin{aligned} \int_0^\tau \kappa\theta_2(x, t)\tilde{a}(x, t)a(x, t) dt &\leq \kappa m \kappa_2 \int_0^\tau \left(\int_0^t e^{-\kappa_3(t-s)} |\theta_1(x, s)| ds \right) |a(x, t)| dt \\ &\leq \kappa m \kappa_2 \int_0^\tau \left(\int_0^t |\theta_1(x, s)| ds \right) \int_0^\tau |a(x, t)| dt \leq C \kappa_2 e^{\kappa m \tau} \left(\int_0^\tau |a(x, t)| dt \right)^2 \\ &\leq C \kappa_2 e^{\kappa m \tau} \tau \int_0^\tau a^2(x, t) dt. \end{aligned}$$

Obviously

$$\int_0^\tau \kappa(\hat{\theta}_1(x, t) + \hat{\theta}_2(x, t) - 1)a^2(x, t) dt \leq 0.$$

Using inequality (12) we get

$$\int_0^\tau \kappa_1 \theta_1(x, s)a(x, s) ds \leq \frac{C\kappa_1}{2\kappa m} e^{\kappa m \tau} \tau \int_0^\tau a^2(x, s) ds.$$

Then

$$\begin{aligned} \int_0^\tau \int_{S_2} [\kappa\rho(\theta_1 + \theta_2)\tilde{a} + \kappa\rho(\hat{\theta}_1 + \hat{\theta}_2 - 1)a + \kappa_1\rho\theta_1] a dS dt \\ \leq C_1 e^{\kappa m \tau} \tau \int_0^\tau \int_{S_2} \rho a^2 dS dt, \end{aligned}$$

$C_1 = C(1/2 + \kappa_2 + \kappa_1/2\kappa m)$ and from (10) we derive the inequality

$$\begin{aligned} \frac{1}{2} \int_{\Omega} a^2(x, \tau) dx + k \int_{Q_\tau} a_x^2 dx dt &\leq C_1 e^{\kappa m \tau} \tau \int_0^\tau \int_{\check{S}_2} \rho a^2 dS dt \\ &= C_1 e^{\kappa m \tau} \tau \int_0^\tau \int_S \rho a^2 dS dt \leq C_1 T e^{\kappa m T} \int_0^\tau \int_S \rho a^2 dS dt. \end{aligned}$$

It is well known (see [7]) that

$$\int_S a^2 dS \leq \varepsilon \int_{\Omega} a_x^2 dx + c_\varepsilon \int_{\Omega} a^2 dx \quad \text{for all } \varepsilon > 0,$$

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

$$\frac{1}{2} \int_{\Omega} a^2 dx + k \int_{Q_\tau} a_x^2 dx dt \leq \varepsilon \int_{Q_\tau} a_x^2 dx dt + c_\varepsilon \int_0^\tau \int_{\Omega} a^2 dx dt \quad \text{for all } \varepsilon > 0;$$

Letting $\varepsilon = k/2$ we get

$$\begin{aligned} \int_{\Omega} a^2 dx dt + k \int_{Q_\tau} a_x^2 dx dt &\leq C_2 \int_0^\tau \int_{\Omega} a^2 dx dt \\ \implies \int_{\Omega} a^2 dx dt &\leq C_2 \int_0^\tau \int_{\Omega} a^2 dx dt, \end{aligned}$$

where constant C is independent of the function a . Set

$$\Phi(\tau) = \int_0^\tau \int_{\Omega} a^2 dx dt.$$

Then

$$\Phi'(\tau) = \int_{\Omega} a^2 dx$$

and

$$\Phi'(\tau) \leq C_2 \Phi(\tau) \iff (e^{-C_2 \tau} \Phi(\tau))' \leq 0 \iff \Phi(\tau) \leq 0.$$

Hence, $a(x, t) \equiv 0$. Now estimate (12) shows, that $\theta_1(x, t) \equiv 0$. From formula (10) we get, that $\theta_2(x, t) \equiv 0$. Then $b \equiv 0$, since it is a solution of problem (3) with the homogeneous conditions. The proof is complete. \square

Theorem 2. Let $S \in C^{1+\alpha}$, $\alpha \in (0, 1)$, and $\rho \in C(S)$. Assume that a_0, b_0 are nonnegative continuous functions on $\bar{\Omega}$ and continuously differentiable on any neighbourhood of S . Let θ_{10}, θ_{20} be continuous on S_2 functions such, that $\theta_{10}(x) \geq 0$, $\theta_{20}(x) \geq 0$ and $\theta_{10}(x) + \theta_{20}(x) < 1$ for all $x \in S_2$. Then problem (1), (2), (3) has a unique classical solution.

Proof. Let $\Omega_0 = \Omega$, if $a_0 = 0$ in any neighbourhood of surface S , and $\Omega_0 \supset \bar{\Omega}$, if a_0 is continuously differentiable in any neighbourhood of surface S . In the last case we extend function a_0 on $\Omega_0 \setminus \bar{\Omega}$ preserving the same smoothness. Suppose, that

$$\Gamma(x, t, y, \tau) = \frac{1}{(4\pi k(t - \tau))^{n/2}} e^{-\frac{|x-y|^2}{4k(t-\tau)}}, \quad t > \tau,$$

is a fundamental solution of equation (1). Then, for any pair of continuous on $S_2 \times [0, T]$ functions θ_1, θ_2 and continuous on S function ρ , problem (1) has a unique solution $a \in C^{2,1}(\Omega \times (0, T]) \cap C(\bar{Q}_T)$ which can be represented by the formula (see [8])

$$a(x, t) = \int_0^t \int_S \Gamma(x, t, \xi, \tau) \varphi(\xi, \tau) dS_\xi d\tau + \int_{\Omega_0} \Gamma(x, t, \xi, 0) a_0(\xi) d\xi, \quad (13)$$

where φ is a continuous and bounded solution on $S \times [0, T]$ of the equation

$$\begin{aligned} \frac{1}{2} \varphi(\eta, t) + \int_0^t \int_S \left(\frac{\partial \Gamma(\eta, t, \xi, \tau)}{\partial \mathbf{n}_\eta} + \frac{1}{k} \sigma(\eta, t) \Gamma(\eta, t, \xi, \tau) \right) \varphi(\xi, \tau) dS_\xi d\tau \\ = \frac{1}{k} \psi(\eta, t) - \int_{\Omega_0} \left(\frac{\partial \Gamma(\eta, t, \xi, 0)}{\partial \mathbf{n}_\eta} + \frac{1}{k} \sigma(\eta, t) \Gamma(\eta, t, \xi, 0) \right) a_0(\xi) d\xi \end{aligned} \quad (14)$$

with

$$\sigma = \sigma(x, t) = \begin{cases} 0, & \text{if } x \in S_1, t > 0, \\ \kappa \rho(x) (1 - \theta_1(x, t) - \theta_2(x, t)), & \text{if } x \in S_2, t > 0, \end{cases}$$

$$\psi = \psi(x, t) = \begin{cases} 0, & \text{if } x \in S_1, t > 0, \\ \kappa_1 \rho(x) \theta_1(x, t), & \text{if } x \in S_2, t > 0, \end{cases}$$

and can be represented by the formula

$$\begin{aligned} \varphi(\eta, t) = g(\eta, t) + \sum_{i=1}^{\infty} \int_0^t \int_S Q_i(\eta, t, \xi, \tau) g(\xi, \tau) dS_\xi d\tau, \\ g(\eta, t) = 2 \left(\frac{\psi(\eta, t)}{k} - \int_{\Omega_0} \left(\frac{\partial \Gamma(\eta, t, \xi, 0)}{\partial \mathbf{n}_\eta} + \frac{\sigma(\eta, t)}{k} \Gamma(\eta, t, \xi, 0) \right) a_0(\xi) d\xi \right), \end{aligned} \quad (15)$$

$$Q_1(\eta, t, \xi, \tau) = -2 \left(\frac{\partial \Gamma(\eta, t, \xi, \tau)}{\partial \mathbf{n}_\eta} + \frac{\sigma(\eta, t)}{k} \Gamma(\eta, t, \xi, \tau) \right),$$

$$Q_{i+1}(\eta, t, \xi, \tau) = \int_{\tau}^t \int_S Q_1(\eta, t, \zeta, s) Q_i(\zeta, s, \xi, \tau) dS_\zeta ds, \quad i = 1, 2, \dots,$$

$$Q_i(\eta, t, \xi, \tau) \leq \frac{C^i}{|\xi - \eta|^{n-1-i\delta}} \frac{1}{(t - \tau)^{1-i\delta}} \frac{\Gamma^i(\gamma)}{\Gamma(i\gamma)};$$

where $\delta = \alpha - 2\gamma > 0$, $0 < \gamma < 1/2$, $\Gamma(t)$ is the gamma function, constant C is independent of function θ_1 , θ_2 and such that $\theta_1(x, t) \geq 0$, $\theta_2(x, t) \geq 0$, $\theta_1(x, t) + \theta_2(x, t) < 1$ for all $(x, t) \in S_2 \times [0, T]$. For small i , function Q_i has a weak singularity and Q_i becomes continuous for $i \geq i_0$. Since

$$\int_{\Omega_0} \left(\frac{\partial \Gamma(\eta, t, \xi, 0)}{\partial \mathbf{n}_\eta} + \frac{\sigma(\eta, t)}{k} \Gamma(\eta, t, \xi, 0) \right) a_0(\xi) d\xi$$

is continuous on $S \times [0, T]$, function g is continuous and bounded as well. Hence,

$$|g(x, t)| \leq K \quad \text{for all } x \in S, \quad t \in [0, T],$$

where constant K is independent of θ_1 , θ_2 which satisfies the conditions $\theta_1(x, t) \geq 0$, $\theta_2(x, t) \geq 0$, $\theta_1(x, t) + \theta_2(x, t) < 1$ for all $(x, t) \in S_2 \times [0, T]$. Therefore,

$$\left| \int_0^t \int_S Q_i(\eta, t, \xi, \tau) g(\xi, \tau) dS_\xi d\tau \right| \leq C^i K \frac{t^{i\gamma}}{\Gamma(i\gamma)} \quad \text{for all } i = 1, 2, \dots$$

These estimates show that series (15) converge uniformly and function φ is continuous and bounded, that is

$$|\varphi(\eta, t)| \leq M \quad \text{for all } \eta \in S, \quad t \in [0, T],$$

where constant M is independent of function θ_1 , θ_2 such that $\theta_1(x, t) \geq 0$, $\theta_2(x, t) \geq 0$, $\theta_1(x, t) + \theta_2(x, t) < 1$ for all $(x, t) \in S_2 \times [0, T]$.

Let a_1 and φ_1 defined by (13) and (15) be solutions of problem (1) and integral equation (14) with function $\theta_1 = \theta_{10}$, $\theta_2 = \theta_{20}$. Then by Lemma 2

$$0 \leq a_1(x, t) \leq \max \left\{ \max_{x \in \bar{\Omega}} a_0(x), \frac{\kappa_1}{\kappa} \max_{x \in S_2} \frac{\theta_{10}(x)}{1 - \theta_{10}(x) - \theta_{20}(x)} \right\} := m$$

for all $x \in \bar{\Omega}$, $t \in [0, T]$,

$$|\varphi_1(\eta, t)| \leq M \quad \text{for all } \eta \in S, \quad t \in [0, T].$$

Assume that θ_{11}, θ_{21} is a solution of Cauchy problem (2) with $a = a_1$. Then

$$\frac{\theta_{11}(x, t)}{1 - \theta_{11}(x, t) - \theta_{21}(x, t)} = \max \left\{ \max_{x \in S_2} \frac{\theta_{10}(x)}{1 - \theta_{10}(x) - \theta_{20}(x)}, \frac{\kappa m}{\kappa_1} \right\}$$

and

$$\frac{\kappa_1}{\kappa} \frac{\theta_{11}(x, t)}{1 - \theta_{11}(x, t) - \theta_{21}(x, t)} \leq \max \left\{ \frac{\kappa_1}{\kappa} \max_{x \in S_2} \frac{\theta_{10}(x)}{1 - \theta_{10}(x) - \theta_{20}(x)}, m \right\} \leq m.$$

Let a_1 and φ_1 defined by (13) and (15) be solutions of problem (1) and integral equation (14) with function $\theta_1 = \theta_{11}, \theta_2 = \theta_{21}$. Then according to Lemma 2

$$0 \leq a_2(x, t) \leq \max \left\{ \max_{x \in \bar{\Omega}} a_0(x), \frac{\kappa_1}{\kappa} \max_{x \in S_2, t \in [0, T]} \frac{\theta_{11}(x, t)}{1 - \theta_{11}(x, t) - \theta_{21}(x, t)} \right\} \leq m$$

and

$$|\varphi_2(\eta, t)| \leq M \quad \text{for all } \eta \in S, \quad t \in [0, T].$$

Suppose, that θ_{12}, θ_{22} is a solution of Cauchy problem (2) with $a = a_2$. Then

$$\frac{\theta_{12}(x, t)}{1 - \theta_{12}(x, t) - \theta_{22}(x, t)} \leq \max \left\{ \max_{x \in S_2} \frac{\theta_{10}(x)}{1 - \theta_{10}(x) - \theta_{20}(x)}, \frac{\kappa m}{\kappa_1} \right\}$$

and

$$\frac{\kappa_1}{\kappa} \frac{\theta_{12}(x, t)}{1 - \theta_{12}(x, t) - \theta_{22}(x, t)} \leq \max \left\{ \frac{\kappa_1}{\kappa} \max_{x \in S_2} \frac{\theta_{10}(x)}{1 - \theta_{10}(x) - \theta_{20}(x)}, m \right\} \leq m.$$

Proceeding this argument we get four following sequences:

$$a_i(x, t) = \int_0^t \int_S \Gamma(x, t, \xi, \tau) \varphi_i(\xi, \tau) dS_\xi d\tau + \int_{\Omega_0} \Gamma(x, t, \xi, 0) a_0(\xi) d\xi,$$

$$\varphi_i(\eta, t) = g_i(\eta, t) + \sum_{j=1}^{\infty} \int_0^t \int_S Q_j(\eta, t, \xi, \tau) g_i(\xi, \tau) dS_\xi d\tau,$$

$$\begin{aligned} \theta_{1i}(x, t) &= \theta_{10}(x) + \int_0^t \kappa (1 - \theta_{1i}(x, s) - \theta_{2i}(x, s)) a_i(x, s) - (\kappa_1 + \kappa_2) \theta_{1i}(x, s) ds, \quad (16) \end{aligned}$$

$$\theta_{2i}(x, t) = \theta_{20}(x) + \int_0^t \kappa_2 \theta_{1i}(x, s) - \kappa_3 \theta_{2i}(x, s) ds, \quad i = 1, 2, \dots \quad (17)$$

Here $g_i = g$ with $\theta_1 = \theta_{1i-1}, \theta_2 = \theta_{2i-1}$. These sequences are uniformly bounded

$$\begin{aligned} 0 \leq a_i(x, t) \leq m & \quad \text{for all } x \in \bar{\Omega}, \quad t \in [0, T], \quad i = 1, 2, \dots, \\ |\varphi_i(\eta, t)| \leq M & \quad \text{for all } \eta \in S, \quad t \in [0, T], \quad i = 1, 2, \dots, \end{aligned}$$

$$0 \leq \theta_{1i}(\eta, t) < 1, \quad 0 \leq \theta_{2i}(\eta, t) < 1 \quad \text{for all } \eta \in S_2, \quad t \in [0, T], \quad i = 1, 2, \dots$$

The potential of a simple layer (see [8] or [9]),

$$\int_0^t \int_S \Gamma(x, t, \xi, \tau) \varphi_i(\xi, \tau) dS_\xi d\tau$$

belong to the Hölder space $C^\lambda(\bar{\Omega} \times [0, T])$ with $\lambda \in (0, 1)$. Therefore, sequence $\{a_i\}_{i=1}^\infty$ is equicontinuous. Functions θ_{1i}, θ_{2i} are solutions of a system (16), (17). Therefore

$$\begin{aligned} |\theta_{1i}(x, t) - \theta_{1i}(x, \tau)| &\leq (\kappa m + \kappa_1 + \kappa_2)(t - \tau), \\ |\theta_{2i}(x, t) - \theta_{2i}(x, \tau)| &\leq (\kappa_2 + \kappa_3)(t - \tau), \\ |\theta_{1i}(x, t) - \theta_{1i}(y, t)| &\leq |\theta_{10}(x) - \theta_{10}(y)| + (\kappa m + \kappa_1 + \kappa_2) \int_0^t |\theta_{1i}(x, s) - \theta_{1i}(y, s)| ds \\ &\quad + \kappa m \int_0^t |\theta_{2i}(x, s) - \theta_{2i}(y, s)| ds + \kappa \int_0^t |a_i(x, s) - a_i(y, s)| ds, \\ |\theta_{2i}(x, t) - \theta_{2i}(y, t)| &\leq |\theta_{20}(x) - \theta_{20}(y)| + \kappa_2 \int_0^t |\theta_{1i}(x, s) - \theta_{1i}(y, s)| e^{-\kappa_3(t-s)} ds \end{aligned}$$

From here we get

$$\begin{aligned} &\int_0^t |\theta_{2i}(x, s) - \theta_{2i}(y, s)| ds \\ &\leq t |\theta_{20}(x) - \theta_{20}(y)| + \int_0^t \kappa_2 \left(\int_0^\tau |\theta_{1i}(x, s) - \theta_{1i}(y, s)| e^{-\kappa_3(\tau-s)} ds \right) d\tau \\ &\leq t |\theta_{20}(x) - \theta_{20}(y)| + \frac{\kappa_2}{\kappa_3} \int_0^t |\theta_{1i}(x, s) - \theta_{1i}(y, s)| ds \end{aligned}$$

and

$$\begin{aligned} &|\theta_{1i}(x, t) - \theta_{1i}(y, t)| \\ &\leq |\theta_{10}(x) - \theta_{10}(y)| + \kappa m |\theta_{20}(x) - \theta_{20}(y)| \\ &\quad + C \int_0^t |\theta_{1i}(x, s) - \theta_{1i}(y, s)| ds + \kappa \int_0^t |a_i(x, s) - a_i(y, s)| ds \end{aligned}$$

where $C = \kappa m + \kappa_1 + \kappa_2 + \kappa m \kappa_2 / \kappa_3$. Now, by the Gronwall lemma, we get the estimate

$$\begin{aligned} & |\theta_{1i}(x, t) - \theta_{1i}(y, t)| \\ & \leq e^{Ct} (|\theta_{10}(x) - \theta_{10}(y)| + \kappa m |\theta_{20}(x) - \theta_{20}(y)|) \\ & \quad + \frac{\kappa}{C} \max_{x \in S_2, t \in [0, t]} |a_i(x, s) - a_i(y, s)|. \end{aligned}$$

Hence,

$$\begin{aligned} & |\theta_{2i}(x, t) - \theta_{2i}(y, t)| \\ & \leq |\theta_{20}(x) - \theta_{20}(y)| + \kappa_2 \frac{e^{Ct}}{C} |\theta_{10}(x) - \theta_{10}(y)| \\ & \quad + \frac{\kappa \kappa_2}{C \kappa_3} \max_{x \in S_2, t \in [0, t]} |a_i(x, s) - a_i(y, s)|. \end{aligned}$$

These estimates show that sequences $\{\theta_{1i}\}_{i=1}^\infty, \{\theta_{2i}\}_{i=1}^\infty$ are equicontinuous. Function φ_i is a solution of integral equation (13) with $\theta_1 = \theta_{1i-1}, \theta_2 = \theta_{2i-1}$. The potential of a double-layer (see [8] or [9]),

$$\int_0^t \int_S \frac{\partial \Gamma(\eta, t, \xi, \tau)}{\partial \mathbf{n}_\eta} \varphi_i(\xi, \tau) dS_\xi dt$$

belongs to the Hölder space $C^\lambda(S \times [0, T])$ with $\lambda < 2\alpha/3$ (see [8]). Therefore, sequence $\{\varphi_i\}_{i=1}^\infty$ is equicontinuous. According to the Arzelà–Ascoli theorem we get four subsequences which converge uniformly. Since problem (1) and (2) cannot possess two classical solutions we claim that sequences $\{a_i\}_{i=1}^\infty, \{\varphi_i\}_{i=1}^\infty, \{\theta_{1i}\}_{i=1}^\infty, \{\theta_{2i}\}_{i=1}^\infty$ converge uniformly.

Set

$$\begin{aligned} a(x, t) &= \lim_{i \rightarrow \infty} a_i(x, t), \quad x \in \overline{\Omega}, \quad t \in [0, T], \\ \varphi(x, t) &= \lim_{i \rightarrow \infty} \varphi_i(x, t), \quad x \in S, \quad t \in [0, T], \\ \theta_1(x, t) &= \lim_{i \rightarrow \infty} \theta_{1i}(x, t), \quad x \in S_2, \quad t \in [0, T], \\ \theta_2(x, t) &= \lim_{i \rightarrow \infty} \theta_{2i}(x, t), \quad x \in S_2, \quad t \in [0, T]. \end{aligned}$$

For limit function a we have formula (13). Therefore, $a \in C^{2,1}(\Omega \times (0, T]) \cap C(\overline{Q_T})$ and it is a solution of problem (1). Pair of functions θ_{1i}, θ_{2i} is a solution of system (16) and (17). Since sequences $\{a_i\}_{i=1}^\infty, \{\theta_i\}_{i=1}^\infty$ are uniformly bounded, we can go to a limit. Pair of limit functions θ_1 and θ_2 is a solution of the system

$$\begin{aligned} \theta_1(x, t) &= \theta_{10}(x) + \int_0^t \kappa (1 - \theta_1(x, s) - \theta_2(x, s)) a(x, s) - (\kappa_1 + \kappa_2) \theta_1(x, s) ds, \\ \theta_2(x, t) &= \theta_{20}(x) + \int_0^t \kappa_2 \theta_1(x, s) - \kappa_3 \theta_2(x, s) ds. \end{aligned}$$

Therefore, θ_1, θ_2 is continuously differentiable with respect to variable t and this pair is a solution of Cauchy problem (2). For this θ_2 problem (3) has a unique classical solution. The proof is complete. \square

Remark 3. Formula (8) is true for limit functions a, θ_1, θ_2 and b .

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