



Reaction on a solid surface supplied by an anomalous mass transfer source



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HIGHLIGHTS

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ABSTRACT

The reaction process occurring on a solid surface where active sites are present is investigated. The phenomenon is described by a linear kinetic equation capable of accounting for memory effects in the adsorption–desorption and a first order reaction process. In order to broaden the formulation of the problem, the surface is in contact with a system defined in a half space where the dynamics is governed by a fractional diffusion equation, meaning, in principle, that the approach can be applied to complex systems such as biological fluids. Our results prove that the anomalous behavior has great importance on the reaction and, consequently, on the densities rates of particles at the surface and on the distribution of particles in the bulk. The results are particularly relevant for heterogeneous catalysis.

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1. Introduction

The limiting surfaces in a sample give rise to distinct chemical reactivity from the bulk. Particles from the bulk may be adsorbed to the surface by either two mechanisms, physisorption, where the particles are attracted to the surfaces through van der Waals forces, or chemisorption, where covalent bonds at active sites adsorb them. Frequently, more than one chemical species are present in the system and chemical reactions on solid substrates may occur, being subject of studies for

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over a century [1]. In fact, this phenomena is intimately related to several technologies such as for catalysts for producing fuel [2], in biochemical sensors [3], electro-optical [4] and solar energy devices [5] and so on. With catalysts support, there are some kinds of surface reactions mechanisms [6]: the Langmuir–Hinshelwood, where two different molecules, A and B , are adsorbed, bond together and then the combination $A - B$ is desorbed; the Rideal–Eley mechanism, where A , for example, is adsorbed by the surface. Then, B encounters A , react, and $A - B$ later is desorbed. The Precursor method assumes that one molecule is adsorbed (A). The second molecule latter collides with the surface and forms a precursor, which latter collides with A , react, forming $A - B$ and desorbs [6]. Another mechanism is the so-called Mars–van Krevelen, which is also very important and widely used in heterogeneously catalyzed oxidation reactions [7–13]. In the Mars–van Krevelen mechanism, the surface itself is a very important part of the reaction. One of the reactants forms a chemical bond with the surface. This catalytic surface layer then reacts with the other reactant from the gas phase and then desorbs. It was proposed as a first order process with respect to the reactant A and the fraction of sites covered by oxygen. In this oxidation reaction, only certain lattice ions O (oxygen) at the surface are involved. In addition, the rate of surface reoxidation, i.e., O_2 adsorption, is proportional to $P_{O_2}^n$ (pressure) and to the concentration of active sites not covered by oxygen. More details about this mechanism can be found in Ref. [7].

In the above mentioned reactions, the reacting species are in different phases, which is the case of heterogeneous catalysis [14]. Different from the homogeneous catalysis, the heterogeneous case has the reacting species embedded in the solid surface while the reactant is, for example, dissolved in a liquid. Therefore, the reaction rate depends crucially on the diffusion of the species involved [15]. Diffusion can be found in several situations such as drug dissolution [16] or delivery [17], transport in biological films [18], cytoplasmic crowding in living cells [19], irregular catalyst structures on diffusion and reaction, diffusion of proteins in sheared lipid membranes [20]. The diffusion can be usual, i.e., characterized by Markovian processes, or anomalous where non-Markovian processes govern the system. In this context, an important point is to understand the mechanisms behind these phenomena and how the presence of memory effects [21,22], long-range interactions [23] and the characteristics of the media (e.g., fractal structure [24–26]) may contribute to the presence of a usual or anomalous diffusion. The surface [27], i.e., the region where the boundary conditions are defined, also plays an important role in diffusion and in others processes such as adsorption and/or desorption [28,29] and catalysis [30,31].

In this article, our goal is to investigate a system with bulk anomalous diffusion in contact with a surface where adsorption–desorption and chemical reactions happen. We consider that the dynamics of particles in the bulk is governed by a fractional diffusion equation, which has been successfully applied in several physical situations [32,33]. We further consider that the processes occurring on the surface are described by linear kinetic equations. These kinetic equations are coupled in order to cover reversible reaction processes. This analysis is performed in Section 2 and in Section 3 the discussions and conclusions are presented. The analysis carried out here may be relevant for several systems of interest (such as heterogeneous catalysis) where surface reaction occur when the substrate is in contact with a media supplying mass to these reactions by means of an anomalous mechanism.

2. Mathematical model and discussions

Let us start our analysis by considering that the particle dynamics in the bulk is governed by the following fractional diffusion equation

$$\frac{\partial}{\partial t} \rho(x, t) = \mathcal{K} \mathcal{D}_t^{1-\alpha} \left[\frac{\partial^2}{\partial x^2} \rho(x, t) \right] \quad (1)$$

where $\rho(x, t)$ is the density of particles, \mathcal{K} is the diffusion coefficient, and the fractional time derivative is the Riemann–Liouville one, defined as [34],

$${}_0 \mathcal{D}_t^{\bar{\alpha}} [\rho(x, t)] = \frac{1}{\Gamma(n - \bar{\alpha})} \frac{d^n}{dt^n} \int_0^t dt' \frac{\rho(x, t')}{(t - t')^{\bar{\alpha}-n+1}} \quad (2)$$

with $n - 1 < \bar{\alpha} < n$, where $\bar{\alpha}$ is a real number and n integer. For the processes which may occur on the surface such as adsorption, desorption, and chemical reactions, we assume that they satisfy the following kinetic equations

$$\tau \frac{d}{dt} \Gamma_a(t) = \kappa \tau \rho(0, t) - \int_0^t dt' k_a(t - t') \Gamma_a(t') + \int_0^t dt' k_{b \rightarrow a}(t - t') \Gamma_b(t'), \quad (3)$$

$$\tau \frac{d}{dt} \Gamma_b(t) = \int_0^t dt' k_{a \rightarrow b}(t - t') \Gamma_a(t') - \int_0^t dt' k_b(t - t') \Gamma_b(t'). \quad (4)$$

In Eqs. (3) and (4), $\Gamma_{a(b)}(t)$ is the surface density, κ , $k_{a(b)}(t)$, and $k_{a \rightarrow b(b \rightarrow a)}(t)$ are the parameters connected to the processes on the surface. In particular, we can write a characteristic time $\tau_\kappa = (\mathcal{K}/\kappa^2)^{2/(2-\alpha)}$ which governs the diffusion and adsorption near the surface. Eq. (3) relates the rate of adsorption of species a with the amount in the bulk, minus the amount of a desorbed plus whatever quantity is reversibly reacted from b to a . Eq. (4) relates the rate of adsorption of species b with the

amount of a that reacts to form b minus the amount of b desorbed. Typical situations governed by first order kinetic equations can be found in several chemical contexts [35] and, in particular, in heavy metal sorption [36]. In addition, depending on the choice of $k_a(t)$ and $k_{b \rightarrow a}(t)$, Eq. (3) may be connected, in the steady-state, to the Henry isotherm [37].

In order to solve these equations, we initially consider them subjected to the condition $\rho(x, 0) = \Phi(x)$, with $\int_0^\infty dx \Phi(x) = 1$, and $\Gamma_a(0) = \Gamma_b(0) = 0$. We also consider that $\rho(\infty, t) = 0$ and

$$\Gamma_a(t) + \Gamma_b(t) + \int_0^\infty \rho(x, t) dx = \text{constant}, \tag{5}$$

which accounts for the conservation of the number of particles present in the system. The boundary conditions may be written as

$$\mathcal{K}_0 \mathcal{D}_t^{1-\alpha} \left[\frac{\partial}{\partial x} \rho(x, t) \right] \Big|_{x=0} = \frac{d}{dt} \Gamma_t(t), \tag{6}$$

where $\Gamma_t(t) = \Gamma_a(t) + \Gamma_b(t)$. Eq. (5) may also be extended to cover other scenarios in which the conservation of the number of particles is not satisfied, such as the situation worked out in Ref. [38]. In this case, suitable changes in Eq. (6) are required to incorporate this scenario to the approach.

By using the Laplace transform and the Green function approach, the solution of Eq. (1) by taking Eq. (5) into account can be written as

$$\rho(x', s) = - \int_0^\infty \mathcal{G}(x, x'; s) \Phi(x) dx' + \rho(0, s) e^{-\sqrt{s^\alpha/\mathcal{K}} x'}, \tag{7}$$

with the Green function given by

$$\mathcal{G}(x, x'; s) = - \frac{1}{2 s \sqrt{\mathcal{K}/s^\alpha}} \left(e^{-\sqrt{s^\alpha/\mathcal{K}} |x-x'|} - e^{-\sqrt{s^\alpha/\mathcal{K}} |x+x'|} \right). \tag{8}$$

The last term of Eq. (7) represents the effect of the surface on the bulk, i.e., how the processes which occur on the surface can modify the evolution of the system's initial condition. For the present case, the surface terms are connected to processes represented by Eqs. (3) and (4). In particular, to simulate a reversible kinetic process, e.g., a chemical reaction $\mathcal{A} \rightleftharpoons \mathcal{B}$, we have that $k_{b \rightarrow a}(t) = k_b(t)$ and $k_{a \rightarrow b}(t) = k_a(t)$. Thus, after performing some calculations, it is possible to show that

$$\rho(0, s) = \frac{\kappa}{\kappa + s \sqrt{\mathcal{K}/s^\alpha}} \int_0^\infty \Phi(x) e^{-\sqrt{s^\alpha/\mathcal{K}} x} dx, \tag{9}$$

and that the concentrations of the species a and b on the surface are given by

$$\Gamma_a(s) = \frac{\kappa}{\kappa + s \sqrt{\mathcal{K}/s^\alpha}} \frac{(s\tau + k_b(s)) \Theta(s)}{s\tau + k_a(s) + k_b(s)}, \tag{10}$$

$$\Gamma_b(s) = \frac{\kappa}{\kappa + s \sqrt{\mathcal{K}/s^\alpha}} \frac{k_a(s) \Theta(s)}{s\tau + k_a(s) + k_b(s)}, \tag{11}$$

with

$$\Theta(s) = \frac{1}{s} \int_0^\infty \Phi(x) e^{-\sqrt{s^\alpha/\mathcal{K}} x} dx. \tag{12}$$

These equations were obtained from Eqs. (7) and (5) by using Eqs. (3) and (4) in the Laplace space. The boundary and initial conditions for $\rho(x, t)$, $\Gamma_a(t)$ and $\Gamma_b(t)$ were also used.

Notice the presence of a stationary state when $t \rightarrow \infty$ ($s \rightarrow 0$), if $k_a(s) \rightarrow k_a = \text{constant}$ and $k_b(s) \rightarrow k_b = \text{constant}$, which results in

$$\Gamma_a(t) \sim \frac{k_b}{k_a + k_b} \quad \text{and} \tag{13}$$

$$\Gamma_b(t) \sim \frac{k_a}{k_a + k_b}. \tag{14}$$

Eqs. (5), (13) and (14) imply that, after some time, the substance, initially present in the bulk, is totally adsorbed by the surface where the reversible reaction process is occurring and is not desorbed to the bulk due to the type of the kinetic process on the surface. Another interesting point about this case, by analyzing Eq. (9), is that this choice of kinetic process on the surface has no influence on the spreading of the species into the bulk. The influence of the surface on the bulk is manifested by κ , the adsorption rate.

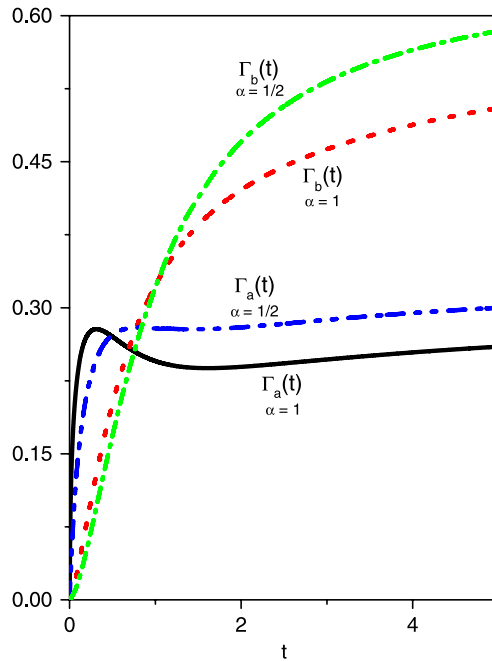


Fig. 1. This figure illustrates the behavior of Eqs. (15) and (16) for $\alpha = 1$ and $\alpha = 1/2$. We use, for simplicity, $k_a = 2, k_b = 1, \tau = 1,$ and $\tau_k = 1$ in arbitrary unities.

In addition, by applying the inverse Laplace transform to Eqs. (10) and (11), for $k_a(s) = k_a = \text{constant}$ and $k_b(s) = k_b = \text{constant}$, we obtain

$$\Gamma_a(t) + \Gamma_b(t) = \frac{\kappa}{\sqrt{\mathcal{K}}} \int_0^t \frac{dt'}{\sqrt{(t-t')^\alpha}} E_{1-\alpha/2, 1-\alpha/2} \left[\frac{\kappa}{\sqrt{\mathcal{K}}} (t-t')^{1-\alpha/2} \right] h(t'), \tag{15}$$

$$\Gamma_b(t) = \int_0^t dt' \Delta(t-t') h(t'), \tag{16}$$

with

$$\Delta(t) = \frac{k_a}{\tau} e^{-k_a+b t/\tau} \int_0^t \frac{\kappa}{\sqrt{\mathcal{K} t'^\alpha}} E_{1-\alpha/2, 1-\alpha/2} \left(\frac{\kappa}{\sqrt{\mathcal{K}}} t'^{1-\alpha/2} \right) e^{k_a+b t'/\tau} dt' \tag{17}$$

and

$$h(t') = \int_0^\infty dx \Phi(x) H_{1,1}^{1,0} \left[\frac{x'}{\sqrt{\mathcal{K} t'^\alpha}} \middle|_{(0,1)}^{(1, \frac{\alpha}{2})} \right], \tag{18}$$

where $k_{a+b} = k_a + k_b$, $E_{\beta, \alpha}(x)$ is a generalized Mittag-Leffler function [34] and $H_{p,q}^{m,n} \left[x \middle|_{(b_q B_q)}^{(a_p A_p)} \right]$ is the Fox H function [39]. The presence of these previous functions is connected to the anomalous spreading of the system due to the fractional time derivative in Eq. (1).

Fig. 1 shows the behavior of $\Gamma_a(t)$ and $\Gamma_b(t)$ for $\alpha = 1$ and $\alpha \neq 1$ in order to illustrate the influence of the fractional coefficient and, thereafter, the anomalous behavior, on the solutions of the kinetic equations. Fig. 3 shows the time evolution of the bulk distribution for the same cases reported in Fig. 1 for three different times. Clearly, the anomalous nature has great influence in both, the surface and bulk densities, as it is evidenced from Figs. 1 and 2.

Interesting cases are obtained when the memory effect is introduced in Eqs. (3) and (4) due to a time dependence on $k_a(t)$ and/or $k_b(t)$, which implies that $k_a(s)$ and/or $k_b(s)$, are not constants as in the previous case. As a particular case, for $k_a(s) = k_a/(1+s\tau_a)$ ($k_a(t) = k_a/\tau_a e^{t/\tau_a}$) [40] and for $k_b(s) = k_b = \text{constant}$ ($k_b(t) = k_b \delta(t)$), the behavior of $\Gamma_a(t)$ and $\Gamma_b(t)$, obtained from the kinetic equations, presents a few oscillations, which can be interpreted as repeated adsorption–desorption phenomena often found in the physisorption process [40], as illustrated in Fig. 3.

Now, let us consider the most general case, where $k_{b \rightarrow a}(t) \neq k_b(t)$ and $k_{b \rightarrow a}(t) \neq k_a(t)$, with $k_a k_b - k_{b \rightarrow a} k_{a \rightarrow b} > 0$. We obtain that

$$\rho(0, s) = \frac{\kappa \{ [s\tau + k_a(s)] [s\tau + k_b(s)] - k_{b \rightarrow a}(s) k_{a \rightarrow b}(s) \} \Theta(s)}{\kappa [s\tau + k_{b \rightarrow a}(s) + k_b(s)] + \sqrt{\frac{\mathcal{K}}{s^\alpha}} \{ [s\tau + k_a(s)] [s\tau + k_b(s)] - k_{b \rightarrow a}(s) k_{a \rightarrow b}(s) \}} \tag{19}$$

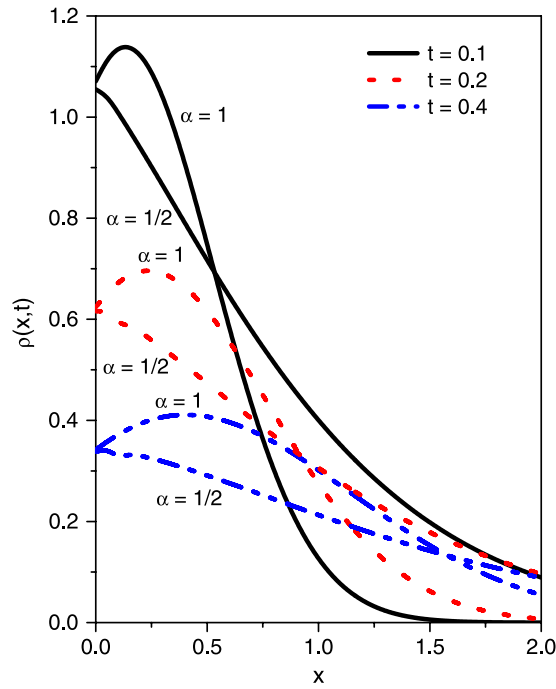


Fig. 2. Behavior of Eq. (7) for $\alpha = 1$ and $\alpha = 1/2$ with $\tau_\kappa = 1$ in arbitrary unities.

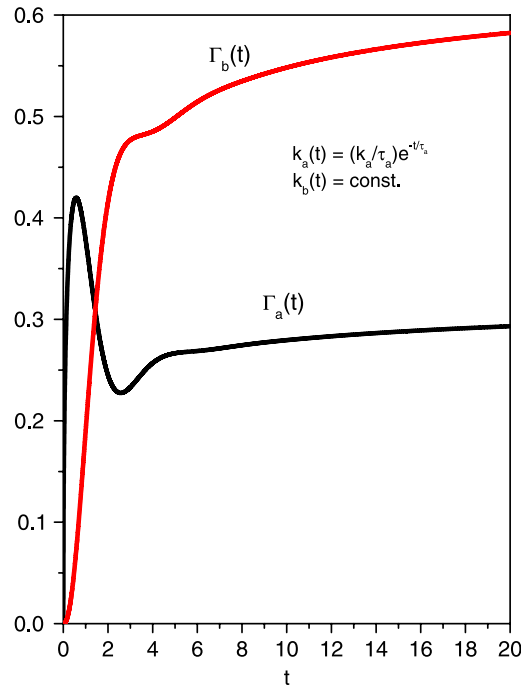


Fig. 3. This figure illustrates the behavior of Eqs. (15) and (16) for $\alpha = 1$ when the memory effect is incorporated in the kinetic equations. For simplicity, we also consider $k_a = 2, k_b = 1, \tau = 1, \tau_a = 1,$ and $\tau_\kappa = 1$ in arbitrary unities.

with

$$\Gamma_a(s) = \frac{\kappa [s\tau + k_b(s)] \Theta(s)}{\kappa [s\tau + k_{b \rightarrow a}(s) + k_b(s)] + \sqrt{\mathcal{K}/s^\alpha} \{ [s\tau + k_a(s)] [s\tau + k_b(s)] - k_{b \rightarrow a}(s) k_{a \rightarrow b}(s) \}}, \quad (20)$$

$$\Gamma_b(s) = \frac{\kappa k_a(s) \Theta(s)}{\kappa [s\tau + k_{b \rightarrow a}(s) + k_b(s)] + \sqrt{\mathcal{K}/s^\alpha} \{ [s\tau + k_a(s)] [s\tau + k_b(s)] - k_{b \rightarrow a}(s) k_{a \rightarrow b}(s) \}}. \quad (21)$$

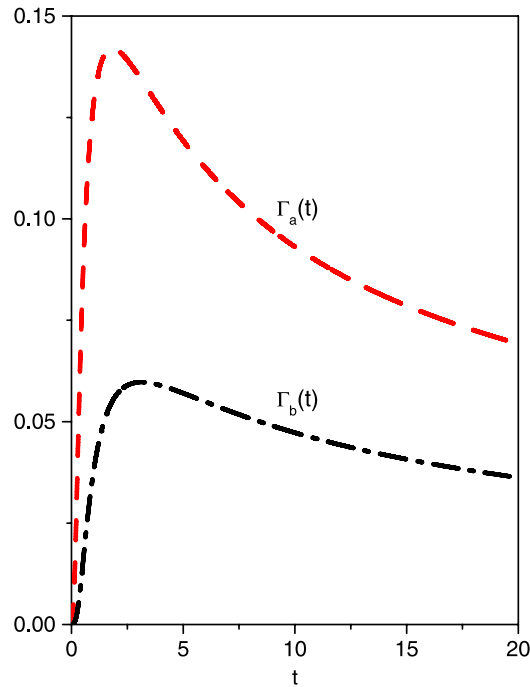


Fig. 4. This figure illustrates the behavior of Eqs. (10) and (11) for $\alpha = 1$. For simplicity, we also consider $k_a = 2$, $k_b = k_{b \rightarrow a} = k_{a \rightarrow b} = 1$, $\tau = 1$, and $\tau_\kappa = 1$ in arbitrary unities.

Notice that, in this case, the processes occurring on the surface have a direct influence on the diffusive process of the system in the bulk, i.e., the quantities k_a , k_b , $k_{b \rightarrow a}$, and $k_{a \rightarrow b}$ in Eq. (19) represent the contribution given by the surface in Eq. (7). Another interesting feature of this case is found in the asymptotic limit of Eqs. (20) and (21), for k_a , k_b , $k_{b \rightarrow a}$, and $k_{a \rightarrow b}$ constants. In this case,

$$\Gamma_a(t) \sim \frac{\kappa k_b}{k_a k_b - k_{b \rightarrow a} k_{a \rightarrow b}} \frac{1}{\sqrt{\mathcal{K} t^\alpha}} \int_0^\infty dx \Phi(x) H_{1,1}^{1,0} \left[\frac{x}{\sqrt{\mathcal{K}_a t^\alpha}} \middle| \begin{matrix} (1 - \frac{\alpha}{2}, \frac{\alpha}{2}) \\ (0,1) \end{matrix} \right], \quad (22)$$

$$\Gamma_b(s) \sim \frac{\kappa k_a}{k_a k_b - k_{b \rightarrow a} k_{a \rightarrow b}} \frac{1}{\sqrt{\mathcal{K} t^\alpha}} \int_0^\infty dx \Phi(x) H_{1,1}^{1,0} \left[\frac{x}{\sqrt{\mathcal{K}_a t^\alpha}} \middle| \begin{matrix} (1 - \frac{\alpha}{2}, \frac{\alpha}{2}) \\ (0,1) \end{matrix} \right]. \quad (23)$$

These equations show that, after some time, the concentration on the surface begins to saturate and the desorption process starts. The substance present in the bulk is initially adsorbed by the surface where the kinetic processes governed by Eqs. (3) and (4) occur and, after some time, is desorbed back to the bulk. This behavior is illustrated in Figs. 4 and 5 which, for small times, show that the system has an accumulation of particles on the surface and, after some time, it spreads out back to the bulk. This case is specially important in connection to heterogeneous catalysis, in the process mentioned before such as the Langmuir–Hinshelwood mechanism, but with memory effects and anomalous diffusion included, which generalizes the problem.

3. Conclusions

We have investigated the solutions of a fractional diffusion equation in connection with kinetic processes present on a surface in contact with the bulk system under consideration. The evolution of the bulk equation and the adsorption–desorption, as well as the chemical reactions that occur on the surface, are coupled by means of kinetic equations incorporating memory effects. We firstly considered the case for which $k_{b \rightarrow a}(t) = k_b(t)$ and $k_{b \rightarrow a}(t) = k_a(t)$, which implies a first order reversible kinetic process, i.e., $\mathcal{A} \rightleftharpoons \mathcal{B}$. The results show that the particles initially present in the bulk, after some time, are adsorbed by the surface, being trapped in the kinetic process. Another case studied was the one for which $k_a \neq k_{a \rightarrow b}$ and $k_b \neq k_{b \rightarrow a}$. It shows a different behavior for long times. The surface, after some time, starts a desorption process which is asymptotically governed by $1/\sqrt{t^\alpha}$. Our approach yields results for the densities of particles on the surface and in the bulk, when memory effects happen in the process of adsorption–desorption and chemical reaction. In particular, we analyze the importance of the diffusion in the bulk when the media presents an anomalous behavior, proving it has great influence on the reaction and consequently, on the densities rates.

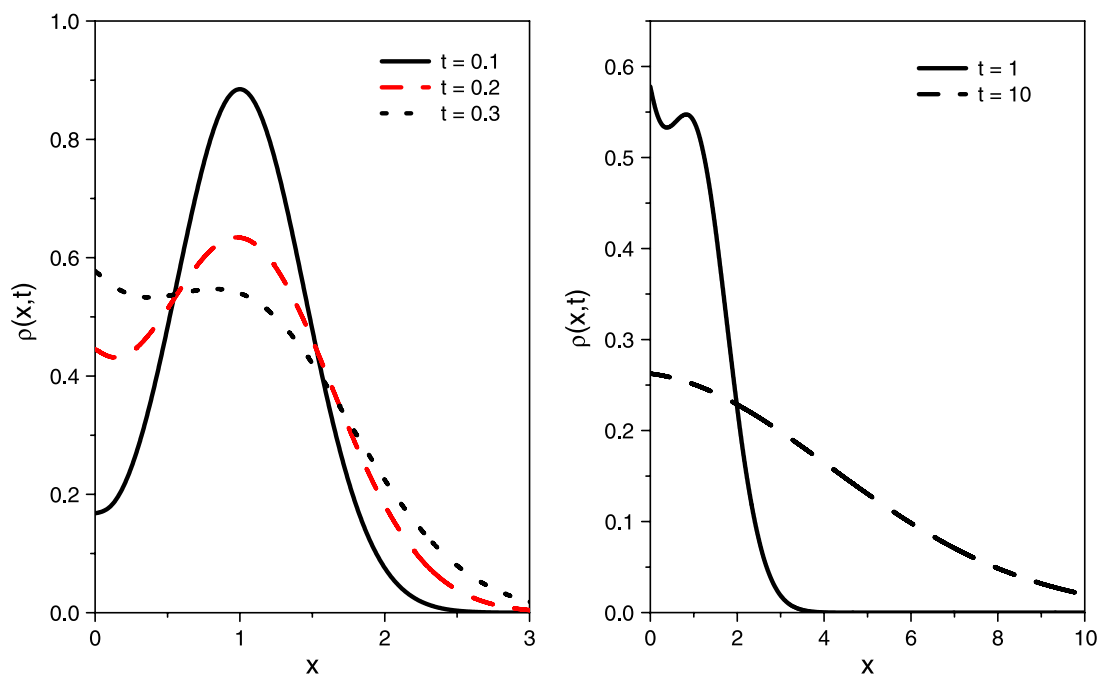


Fig. 5. This figure illustrates the behavior of Eq. (7) for different times for $\alpha = 1$ when $k_{b \rightarrow a}(t) \neq k_b(t)$ and $k_{b \rightarrow a}(t) \neq k_a(t)$. For simplicity, we also consider $k_a = 2$, $k_b = k_{b \rightarrow a} = k_{a \rightarrow b} = 1$, $\tau_c = 1$, and $\tau = 1$ in arbitrary unities. Note that for $t = 1$ the particles are very close to the surface where the kinetic processes are occurring. For $t = 10$ the particles leave the surface by a desorption process and spread out.

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