

Research Article

Diffusion Process and Reaction on a Surface

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We investigate the influence of the surface effects on a diffusive process by considering that the particles may be sorbed or desorbed or undergo a reaction process on the surface with the production of a different substance. Our analysis considers a semi-infinite medium, where the particles may diffuse in contact with a surface with active sites. For the surface effects, we consider integrodifferential boundary conditions coupled with a kinetic equation which takes non-Debye relation process into account, allowing the analysis of a broad class of processes. We also consider the presence of the fractional derivatives in the bulk equations. In this scenario, we obtain solutions for the particles in the bulk and on the surface.

1. Introduction

The description of the dynamic processes present on the surface such as adsorption-desorption or reactions and their influence on the diffusion is very important due to the broadness of applications in several fields such as optimization of industrial processes, electronic devices, the action of pharmaceuticals in the organism, and materials science [1]. Particularly in chemical engineering applications of reactions that occur on solid surfaces are of the great interest and advances have recently been achieved by using the heterogeneous catalysis in the pollutants removal [2], with emphasis on oxidative pathways involving the formation of radicals [3–5], and in the biodiesel production [6–8]. In this sense, it is worth not only studying chemical reactions on substrates, but also analyzing different phenomena occurring in a given surface, for example, the adsorption of proteins [9–12], which can be controlled by nanoroughness on the surface of the material [13, 14] or hybridization of DNA [15–17], among others. In these systems, different species may diffuse and react [18–21], which implies considering suitable changes in the diffusion equation or in the boundary conditions to account for the processes of interest [18, 19, 22, 23]. These contexts usually are analyzed by using the standard approaches

related to Markovian processes. Therefore, extensions of these approaches become very important in order to consider non-Markovian processes where anomalous diffusion or non-Debye relaxations on the kinetic processes are present.

Here, we analyze the surface effects on a diffusion process by considering one-dimensional semi-infinite media in contact with a surface, which may adsorb (and desorb) particles or absorb the particles to perform a reaction process with the formation of another kind of particle. Figure 1 illustrates the scenario analyzed in this manuscript, where two species (particles or substances) 1 and 2 diffuse in the bulk which is in contact with a surface. Species 1 may be adsorbed or sorbed by the surface. For the first process, species 1 is removed from the bulk by the surface and after some time is desorbed with a characteristic time. In the other case, when species 1 is sorbed, a reaction process may occur with the formation of species 2. In this sense, the reaction only occurs on the surface, for instance, in the presence of an specific catalyst. Thus, species 1 reacts promoting the formation of species 2, in other words, following a first-order irreversible reaction $1 \rightarrow 2$. This reaction can be described by a kinetic equation [21, 24, 25]. In order to describe these processes, the model considered here is a set of coupled equations that may represent reaction, adsorption, and desorption processes

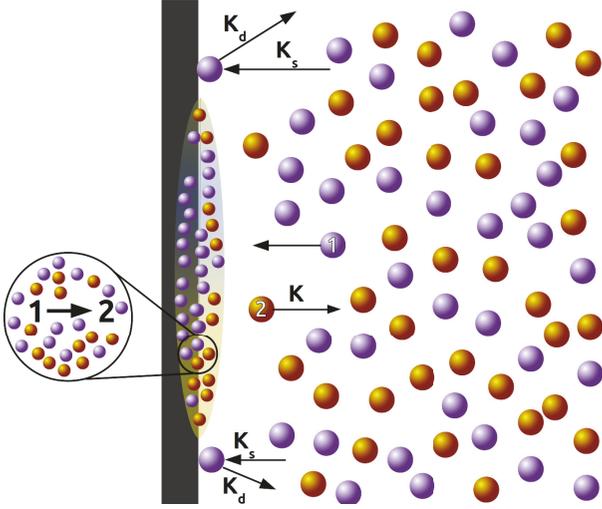


FIGURE 1: Illustration of the interactions occurring between particles and, for example, a catalyst surface. Species 1 in contact with the surface can either be adsorbed or sorbed and by a reaction process produce species 2, i.e., $1 \rightarrow 2$.

of a substrate on a surface and also the diffusion process in the bulk. These developments are performed in Section 2 by considering generalized diffusion equations [26–40] for the species (1 and 2) in the bulk coupled with kinetic equations as boundary conditions. The discussion and conclusions are presented in Section 3.

2. Diffusion and Surface Effects

Let us start our analysis about the surface effects on the diffusion processes by considering the system governed in the bulk by the following generalized diffusion equations

$$\frac{\partial}{\partial t} \rho_1(x, t) = \mathcal{K}_1 \mathcal{F}_{\alpha_1} \left[\frac{\partial^2}{\partial x^2} \rho_1(x, t) \right], \quad (1)$$

and

$$\frac{\partial}{\partial t} \rho_2(x, t) = \mathcal{K}_2 \mathcal{F}_{\alpha_2} \left[\frac{\partial^2}{\partial x^2} \rho_2(x, t) \right], \quad (2)$$

with $0 \leq x < \infty$, where \mathcal{K}_1 and \mathcal{K}_2 are the generalized diffusion coefficients related to particles 1 and 2, respectively. The quantities $\rho_1(x, t)$ and $\rho_2(x, t)$ represent the density of each particle present in the bulk and $\mathcal{F}_{\alpha_{1(2)}}\{\dots\}$, in (1) and (2), is the integrodifferential operator:

$$\begin{aligned} \mathcal{F}_{\alpha_{1(2)}}\{\rho_{1(2)}(x, t)\} \\ = \frac{\partial}{\partial t} \int_0^t dt' k_{\alpha_{1(2)}}(t-t') \rho_{1(2)}(x, t'), \end{aligned} \quad (3)$$

where $k_{\alpha_{1(2)}}(t)$ defines how the past history of each system influences the time evolution of $\rho_{1(2)}(x, t)$ and depending on

the choice of $k_{\alpha_{1(2)}}(t)$ different fractional differential operators may be obtained. For example, the case

$$k_{\alpha_{1(2)}}(t) = \frac{t^{\alpha_{1(2)}-1}}{\Gamma(\alpha_{1(2)})} \quad (4)$$

corresponds to the well-known Riemann-Liouville fractional operator [41] for $0 < \alpha_{1(2)} < 1$, which has been applied in several situations related to anomalous diffusion [42–45]. The exponential kernel

$$k_{\alpha_{1(2)}}(t) = \frac{\mathcal{R}(\alpha_{1(2)})}{1 - \alpha_{1(2)}} e^{-\bar{\alpha}_{1(2)} t}, \quad (5)$$

with $\bar{\alpha}_{1(2)} = \alpha_{1(2)}/(1 - \alpha_{1(2)})$ and $\mathcal{R}(\alpha_{1(2)})$ being a normalization factor, corresponds to the fractional operator of Caputo-Fabrizio [46]. Further possibilities for describing the kernel $k_{\alpha_{1(2)}}(t)$ are discussed by Gómez-Aguilar et al. [47], in particular the kernel $k_{\alpha_{1(2)}}(t) = \mathcal{R}(\alpha_{1(2)}) E_{\alpha_{1(2)}}(-\bar{\alpha}_{1(2)} t^{\alpha_{1(2)}})/(1 - \alpha_{1(2)})$, where $E_{\alpha}(\dots)$ is the Mittag-Leffler function [41], which leads us to Atangana-Baleanu fractional operator [48–50]. It has been considered in the Bloch system [51], by resulting in a different behavior from the one obtained in the usual context where differential operators of integer order are considered. These operators, by taking a variable order into account, have been used to extend the Gray-Scott reaction-diffusion model which describe irreversible reaction between two species. In this scenario, the Atangana-Baleanu-Caputo fractional differential operator has shown a faster stabilization behavior and Liouville-Caputo fractional differential operator presented a stronger memory effect [52]. From these choices for $k_{\alpha_{1(2)}}(t)$, we observe that the Riemann-Liouville operator has a singularity at the origin ($t = 0$), while the recently proposed Caputo-Fabrizio and Atangana-Baleanu operators are nonsingular [46–48, 53–55], and the last one may manifest different regimes of diffusion.

On the surface, we consider that the processes are governed by the following equations:

$$\begin{aligned} \mathcal{K}_1 \frac{\partial}{\partial x} \mathcal{F}_{\alpha_1} \{\rho_1(x, t)\} \Big|_{x=0} \\ = \frac{d}{dt} \mathcal{C}(t) + \int_0^t k(t-t') \rho_1(0, t') dt' \end{aligned} \quad (6)$$

and

$$\begin{aligned} \mathcal{K}_2 \frac{\partial}{\partial x} \mathcal{F}_{\alpha_2} \{\rho_2(x, t)\} \Big|_{x=0} \\ = - \int_0^t k(t-t') \rho_1(0, t') dt'. \end{aligned} \quad (7)$$

Here $k(t)$ is related to the rate of particle sorption by the surface which by a reaction process produces substance 2. In addition to (6) and (7), we also have the boundary conditions $\partial_x \rho_1(\infty, t) = 0$ and $\partial_x \rho_2(\infty, t) = 0$ to solve (1) and (2). In (6), $\mathcal{C}(t)$ represents the density of particles which are adsorbed by the surface. For the adsorption and desorption processes

on the surface, we assume that they may be modeled by the following kinetic equation [56]:

$$\frac{d}{dt} \mathcal{C}(t) = k_s \rho_1(0, t) - \int_0^t k_d(t-t') \mathcal{C}(t') dt'. \quad (8)$$

In (8), $\rho_1(0, t)$ is the bulk density just in front of the surface, which may be adsorbed by the surface. The parameter k_s is connected to the adsorption phenomenon, being related to a characteristic adsorption time $\tau \propto 1/k_s$, and $k_d(t)$ is a kernel that governs the desorption phenomenon. Thus, the surface density of adsorbed particles depends on the bulk density of particles just in front of the membrane, and on the surface density of particles already sorbed [56]. Equation (8) also extends the usual kinetic equations of first order to situations characterized by unusual relaxations, i.e., non-Debye relaxations for which a nonexponential behavior of the densities can be obtained, depending on the choice of the kernels [56, 57].

From (1), (2), (6), and (7), it is possible to show that

$$\begin{aligned} \frac{d}{dt} \left(\int_0^\infty \rho_1(x, t) dx + \mathcal{C}(t) \right) \\ = - \int_0^t k(t-t') \rho_1(0, t') dt', \end{aligned} \quad (9)$$

and

$$\frac{d}{dt} \left(\int_0^\infty \rho_2(x, t) dx \right) = \int_0^t k(t-t') \rho_1(0, t') dt'. \quad (10)$$

In (9) the term $\int_0^t k(t-t') \rho_1(0, t') dt'$ implies the removal of the particles from the bulk by the surface to promote the production of species 2 by a reaction process. Thus, (11) shows that the mass (number of particles) variation on species 1 is connected to the variations on species 2. In particular, the negative sign shows that the variation of particles of one species produces an opposite variation on the other. The particles produced by a reaction process on the surface are being released from the surface to the bulk. Equations (9) and (10) imply

$$\begin{aligned} \frac{d}{dt} \left(\int_0^\infty \rho_1(x, t) dx + \mathcal{C}(t) \right) \\ = - \frac{d}{dt} \left(\int_0^\infty \rho_2(x, t) dx \right), \end{aligned} \quad (11)$$

which consequently yields

$$\mathcal{C}(t) + \int_0^\infty \rho_1(x, t) dx + \int_0^\infty \rho_2(x, t) dx = \text{constant}, \quad (12)$$

i.e., a direct consequence of the conservation of the total number of particles present in the system.

These systems of coupled equations can be solved by using the Laplace transform and the Green function approach. In fact, by applying the Laplace transform $\{\mathcal{L}\{\rho_{1(2)}(x, t)\} = \int_0^\infty dt e^{-st} \rho_{1(2)}(x, t) = \bar{\rho}_{1(2)}(x, s)$ and $\mathcal{L}^{-1}\{\bar{\rho}_{1(2)}(x, s)\} =$

$\int_0^\infty dt e^{-st} \bar{\rho}_{1(2)}(x, s) = \rho_{1(2)}(x, t)$) it is possible to simplify the previous equations and obtain that

$$\mathcal{D}_1(s) \frac{\partial^2}{\partial x^2} \bar{\rho}_1(x, s) - s \bar{\rho}_1(x, s) = -\rho_1(x, 0), \quad (13)$$

and

$$\mathcal{D}_2(s) \frac{\partial^2}{\partial x^2} \bar{\rho}_2(x, s) - s \bar{\rho}_2(x, s) = -\rho_2(x, 0), \quad (14)$$

which are subjected, in the Laplace domain, to the boundary conditions:

$$\begin{aligned} \mathcal{D}_1(s) \frac{\partial}{\partial x} \bar{\rho}_1(x, s) \Big|_{x=0} \\ = \left[\frac{sk_s}{s + \bar{k}_d(s)} + \bar{k}(s) \right] \bar{\rho}_1(0, s) - \frac{\bar{k}_d(s)}{s + \bar{k}_d(s)} \mathcal{C}(0), \end{aligned} \quad (15)$$

$$\mathcal{D}_2(s) \frac{\partial}{\partial x} \bar{\rho}_2(x, s) \Big|_{x=0} = -\bar{k}(s) \bar{\rho}_1(0, s) \quad (16)$$

with $\rho_1(x, 0) = \varphi_1(x)$ and $\rho_2(x, 0) = \varphi_2(x)$, where $\bar{\mathcal{D}}_{1(2)}(s) = \mathcal{K}_{1(2)} s \bar{k}_{\alpha_{1(2)}}(s)$. By using the Green function approach, the solutions for $\bar{\rho}_1(x, s)$ and $\bar{\rho}_2(x, s)$ can be found by considering the previous equations and they are given by

$$\begin{aligned} \bar{\rho}_1(x, s) = - \int_0^\infty \bar{\mathcal{G}}_1(x, x'; s) \varphi_1(x') dx' \\ + \frac{sk_s}{s + \bar{k}_d(s)} \bar{\rho}_1(0, s) \bar{\mathcal{G}}_1(x, 0; s) \\ - \frac{\bar{k}_d(s)}{s + \bar{k}_d(s)} \mathcal{C}(0) \bar{\mathcal{G}}_1(x, 0; s) \end{aligned} \quad (17)$$

and

$$\begin{aligned} \bar{\rho}_2(x, s) = - \int_0^\infty \bar{\mathcal{G}}_2(x, x'; s) \varphi_2(x') dx' \\ - \bar{k}(s) \bar{\mathcal{G}}_2(x, 0; s) \bar{\rho}_1(0, s). \end{aligned} \quad (18)$$

where $\bar{\mathcal{G}}_1(x, x'; s)$ and $\bar{\mathcal{G}}_2(x, x'; s)$ correspond to the Green function for each species and $\mathcal{C}(0)$ represents the quantity of particles which may initially be present on the surface. These Green functions are obtained by solving the following equation:

$$\bar{\mathcal{D}}_i(s) \frac{\partial^2}{\partial x^2} \bar{\mathcal{G}}_i(x, x'; s) - s \bar{\mathcal{G}}_i(x, x'; s) = \delta(x - x') \quad (19)$$

by taking into account the suitable boundary conditions for each species related to (17) and (18). Thus, the solutions given by (17) and (18) are obtained from the combination of (15), (16), and (19) by taking into account their boundary conditions, following the procedure employed in [58]. It is also interesting to note that the boundary conditions chosen for the Green function were performed in order to evidence the adsorption-desorption process manifested by the surface

on the distribution $\rho(x, t)$. For species 1, by performing some calculations, it is possible to show that the Green function is given by

$$\begin{aligned} \bar{\mathcal{G}}_1(x, x'; s) &= -\frac{1}{2\sqrt{s\bar{\mathcal{D}}_1(s)}} \left(e^{-\sqrt{s/\bar{\mathcal{D}}_1(s)}|x+x'|} + e^{-\sqrt{s/\bar{\mathcal{D}}_1(s)}|x-x'|} \right) \\ &+ \frac{\bar{k}(s)}{\sqrt{s\bar{\mathcal{D}}_1(s)} + \bar{k}(s)} \frac{1}{\sqrt{s\bar{\mathcal{D}}_1(s)}} e^{-\sqrt{s/\bar{\mathcal{D}}_1(s)}|x+x'|}, \end{aligned} \quad (20)$$

when the boundary conditions, which are consistent with (17),

$$\bar{\mathcal{D}}_1(s) \frac{\partial}{\partial x} \bar{\mathcal{G}}_1(x, x'; s) \Big|_{x=0} - \bar{k}(s) \bar{\mathcal{G}}_1(0, x'; s) = 0 \quad (21)$$

and $\partial_x \bar{\mathcal{G}}_1(x, x'; s)|_{x=\infty} = 0$ are considered to solve (21). Note that the boundary condition used to solve the equation for the Green function incorporates the reaction on the surface. The term related to the adsorption-desorption process was not incorporated in (21) to be evident on the solution as an additional term. In (20), the first term corresponds to the spreading of the initial condition and the second term shows the influence of the surface on the spreading of the system, which in this case corresponds to a reaction sorption process where species 1 is removed from the system. It is worth mentioning that depending on the choice of $\bar{k}_{\alpha_1}(s)$ (or $k_{\alpha_1}(t)$) the previous Green function may present a stationary solution. In particular, for case $\bar{k}_{\alpha_1}(s) \rightarrow k_{\alpha_1} = \text{constant}$ with $\bar{k}(s) \rightarrow k = \text{constant}$, for $s \rightarrow 0$ (i.e., for long times), we have, for the Green function, the following stationary behavior:

$$\mathcal{G}_1(x, x') = -\frac{1}{2\sqrt{\bar{D}_1}} \left(e^{-|x-x'|/\sqrt{\bar{D}_1}} - e^{-|x+x'|/\sqrt{\bar{D}_1}} \right), \quad (22)$$

which in this case is independent of $\bar{k}(s)$, where $\bar{D}_1 = k_{\alpha_1} \mathcal{K}_1 = \text{constant}$. This feature is interesting and shows that the surface effects with this characteristic play an important role at intermediate times. For the case $\bar{k}(s) = 0$ (or $k(t) = 0$), we also have a stationary solution and it is given by

$$\mathcal{G}_1(x, x') = -\frac{1}{2\sqrt{\bar{D}_1}} \left(e^{-|x+x'|/\sqrt{\bar{D}_1}} + e^{-|x-x'|/\sqrt{\bar{D}_1}} \right), \quad (23)$$

which is different from (22) due to the boundary conditions used to obtain the Green function. The scenario is typical, for example, of the Caputo-Fabrizio operator, which is characterized by an exponential kernel as mentioned before.

For species 2, we have that

$$\begin{aligned} \bar{\mathcal{G}}_2(x, x'; s) &= -\frac{1}{2\sqrt{s\bar{\mathcal{D}}_2(s)}} \left(e^{-\sqrt{s/\bar{\mathcal{D}}_2(s)}|x+x'|} + e^{-\sqrt{s/\bar{\mathcal{D}}_2(s)}|x-x'|} \right), \end{aligned} \quad (24)$$

when the boundary conditions, which are consistent with Eq. (18),

$$\begin{aligned} \bar{\mathcal{D}}_2(s) \frac{\partial}{\partial x} \bar{\mathcal{G}}_2(x, x'; s) \Big|_{x=0} \\ = \bar{\mathcal{D}}_2(s) \frac{\partial}{\partial x} \bar{\mathcal{G}}_2(x, x'; s) \Big|_{x=\infty} = 0 \end{aligned} \quad (25)$$

are employed to solve (21). Equation (24) may also present a stationary solution depending on the choice of $\bar{k}_{\alpha_2}(s)$, similar to (20).

In order to perform the inverse of Laplace transform and obtain the time dependent behavior of (20) and (24), we first consider the Riemann-Liouville fractional time derivative and after that the Caputo-Fabrizio fractional time derivative. We also consider that $\bar{k}(s) = k = \text{constant}$; i.e., the sorption of species 1 by the surface occurs in a constant rate. For the Riemann-Liouville fractional time derivative, we have that

$$\bar{\mathcal{D}}_{1(2)}(s) = \mathcal{K}_{1(2)} s^{1-\alpha_{1(2)}/2}, \quad (26)$$

and, consequently,

$$\begin{aligned} \mathcal{G}_{1,RL}(x, x'; t) &= -\frac{1}{\sqrt{4\mathcal{K}_1} t^{\alpha_1}} \left(H_{1,1}^{1,0} \left[\frac{|x-x'|}{\sqrt{\mathcal{K}_1} t^{\alpha_1}} \Big|_{(0,1)}^{(1-\alpha_1/2, \alpha_1/2)} \right] \right. \\ &+ H_{1,1}^{1,0} \left[\frac{|x+x'|}{\sqrt{\mathcal{K}_1} t^{\alpha_1}} \Big|_{(0,1)}^{(1-\alpha_1/2, \alpha_1/2)} \right] \left. \right) + \frac{k}{\sqrt{4\mathcal{K}_1}} \\ &\cdot \int_0^t \frac{dt'}{\sqrt{t'^{\alpha_1} (t-t')^{\alpha_1}}} E_{\alpha_1, \alpha_1}' \left(-\frac{kt'^{\alpha_1}}{\sqrt{\mathcal{K}_1}} \right) \\ &\cdot H_{1,1}^{1,0} \left[\frac{|x+x'|}{\sqrt{\mathcal{K}_1} (t-t')^{\alpha_1}} \Big|_{(0,1)}^{(1-\alpha_1/2, \alpha_1/2)} \right] \end{aligned} \quad (27)$$

with $\alpha_1' = 1 - \alpha_1/2$, where $H_{p,q}^{m,n}[x]_{(b,B)}^{(a,A)}$ is the Fox H function [59] and $E_{\alpha,\beta}(x)$ is the generalized Mittag-Leffler function [59] (see Figure 2). For the Caputo-Fabrizio fractional time operator, we have that

$$\bar{\mathcal{D}}_{1(2)}(s) = \frac{sD_{1(2)}}{s + \bar{\alpha}_{1(2)}} \quad (28)$$

with $D_{1(2)} = [\mathcal{R}(\alpha_{1(2)})/(1 - \alpha_{1(2)})] \mathcal{K}_{1(2)}$. By substituting the previous equation in (20) and performing some calculations

with $\bar{k}(s) = k = \text{constant}$, we obtain that

$$\begin{aligned} \mathcal{G}_{1,CF}(x, x'; t) &= - \int_0^t \frac{dt'}{\sqrt{4\pi D_1 t'}} \Xi(t, t') \\ &\cdot e^{-\bar{\alpha}_1 t'} \left(e^{-(x-x')^2/4D_1 t'} + e^{-(x+x')^2/4D_1 t'} \right) \\ &- \frac{k}{\sqrt{k^2 + 4D_1 \bar{\alpha}_1}} \int_0^t dt' \Xi(t, t') e^{-\bar{\alpha}_1 t'} \int_0^{t'} dt'' \\ &\cdot \frac{\Lambda(t'')}{\sqrt{4\pi D_1 (t' - t'')^3}} e^{-x^2/4D_1 (t-t')} \end{aligned} \quad (29)$$

for the Caputo-Fabrizio fractional time operator with $\Xi(t, t') = \bar{\alpha}_1 + \delta(t - t')$:

$$\Lambda(t) = \xi_- \operatorname{erfc}(\xi_- \sqrt{t}) e^{\xi_-^2 t} - \xi_+ \operatorname{erfc}(\xi_+ \sqrt{t}) e^{\xi_+^2 t} \quad (30)$$

where $\operatorname{erfc}(x)$ is the complementary error function and $\xi_{\pm} = k/\sqrt{4D_1} \mp \sqrt{k^2/(4D_1) + \bar{\alpha}_1}$. Figure 3 shows the behavior of (29) for different values of α_1 . In Figure 4, we present the behavior of (29) for different times in order to show that for long times a stationary state is reached. For (24), we have

$$\begin{aligned} \mathcal{G}_{2,RL}(x, x'; t) &= - \frac{1}{\sqrt{4\mathcal{K}_2 t^{\bar{\alpha}_2}}} \left(H_{1,1}^{1,0} \left[\frac{|x - x'|}{\sqrt{\mathcal{K}_2 t^{\bar{\alpha}_2}}} \right]_{(0,1)}^{(1-\bar{\alpha}_2/2, \bar{\alpha}_2/2)} \right) \\ &+ H_{1,1}^{1,0} \left[\frac{|x + x'|}{\sqrt{\mathcal{K}_2 t^{\bar{\alpha}_2}}} \right]_{(0,1)}^{(1-\bar{\alpha}_2/2, \bar{\alpha}_2/2)} \end{aligned} \quad (31)$$

for the Riemann-Liouville operator (see Figure 5) and

$$\begin{aligned} \mathcal{G}_{2,CF}(x, x'; t) &= - \int_0^t \frac{dt'}{\sqrt{4\pi D_2 t'}} \Xi(t, t') \\ &\cdot e^{-\alpha t'} \left(e^{-(x-x')^2/4D_2 t'} + e^{-(x+x')^2/4D_2 t'} \right) \end{aligned} \quad (32)$$

for the Caputo-Fabrizio fractional time operator (see Figure 6).

In (17) and (18), the first term promotes the spreading of the initial condition and the other terms represent the influence of the surface on the diffusive process in the bulk. Thus, we need to quantify these terms, in order to obtain the solution for each species. From (17), after some calculations, it is also possible to show that

$$\begin{aligned} \bar{\rho}_1(0, s) &= - \frac{1}{1 - sk_s \bar{\omega}(s) \bar{\mathcal{G}}_1(0, 0; s)} \left(\int_0^\infty \bar{\mathcal{G}}_1(0, x'; s) \right. \\ &\cdot \varphi(x') dx' + \bar{k}_d(s) \bar{\omega}(s) \bar{\mathcal{G}}_1(0, 0; s) \mathcal{C}(0) \end{aligned} \quad (33)$$

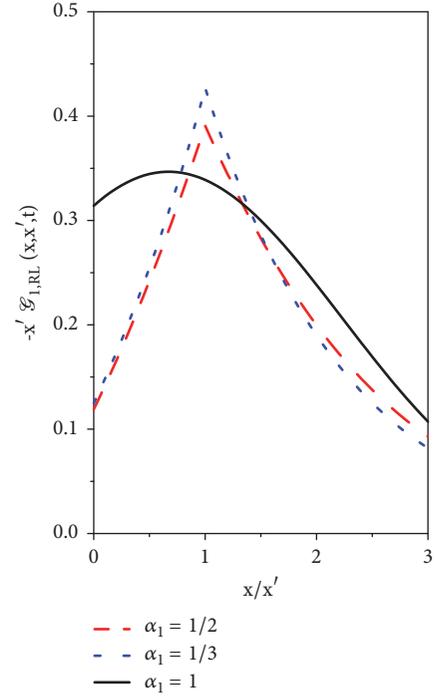


FIGURE 2: This figure illustrates the behavior of (27) by considering different values of α_1 . We consider, for simplicity, $\tau_{1,RL} = (x'^2/\mathcal{K}_1)^{1/\alpha_1}$, $k\tau_{1,RL}/x' = 1$, and $t = \tau_{1,RL}$, in arbitrary unities.

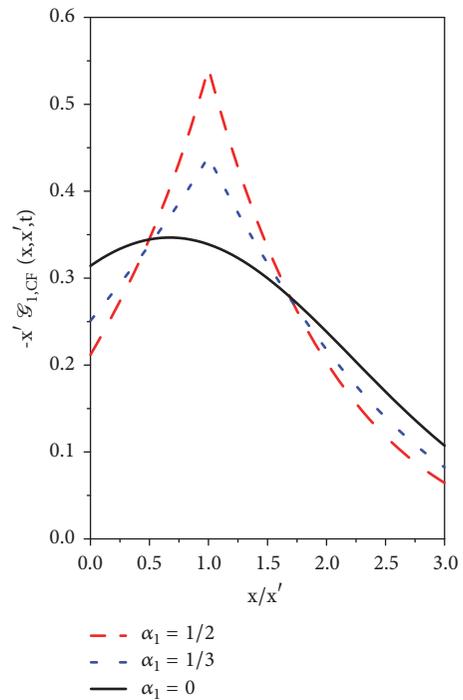


FIGURE 3: This figure illustrates the behavior of (29) by considering different values of α_1 . We consider, for simplicity, $\tau_{1,CF} = \sqrt{x'^2/D_1}$, $k\tau_{1,CF}/x' = 1$, $\tau_{1,CF} = 1$, and $\alpha_1 = 1/2$, in arbitrary unities.

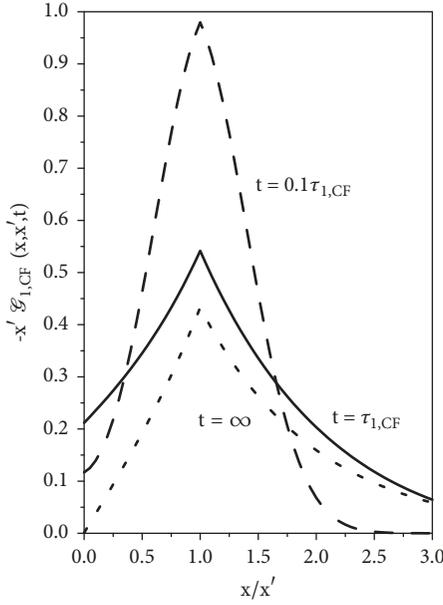


FIGURE 4: This figure illustrates the behavior of (29) by considering different times. We consider, for simplicity, $\tau_{1,CF} = \sqrt{x'^2/D_1}$, $k\tau_{1,CF}/x' = 1$, and $\alpha_1\tau_{1,CF} = 1/2$, in arbitrary unities.

where $\bar{\omega}(s) = 1/(s + \bar{k}_d(s))$. By using the previous result, it is possible to show that the quantity of adsorbed particles by the surface is given by

$$\begin{aligned} \bar{\mathcal{E}}(s) = & -\frac{k_s \bar{\omega}(s)}{1 - sk_s \bar{\omega}(s) \bar{\mathcal{E}}_1(0, 0, 0; s)} \left(\int_0^\infty \bar{\mathcal{E}}_1(0, x'; s) \right. \\ & \cdot \varphi(x') dx' + \bar{\omega}(s) \left(1 - k_s \bar{k}_d(s) \right. \\ & \left. \left. \cdot \frac{\bar{\omega}(s) \bar{\mathcal{E}}_1(0, 0, 0; s)}{1 - sk_s \bar{\omega}(s) \bar{\mathcal{E}}_1(0, 0, 0; s)} \right) \mathcal{E}(0) \right). \end{aligned} \quad (34)$$

By using (33) and (34) and by applying the inverse Laplace transform in (13) and (14), it is possible to obtain $\rho_1(x, t)$ and $\rho_2(x, t)$. We consider, as performed for the Green functions, the fractional operators characterized by (4) and (5) in order to obtain the inverse Laplace transform of (33) and (34). For the first case, i.e., the Riemann-Liouville fractional time derivative, we obtain for (33) and (34) that

$$\begin{aligned} \rho_{1,RL}(0, t) = & \int_0^t \frac{dt'}{t'} \Upsilon_{RL}(t - t') \int_0^\infty dx' \varphi_1(x') \\ & \cdot H_{1,1}^{1,0} \left[\frac{|x - x'|}{\sqrt{\mathcal{D}_2 t'^{\alpha}}} \right]_{(0,1)}^{(0,\alpha/2)} + \left(\Upsilon_{RL}(t) \right. \\ & \left. - \int_0^t dt' \Upsilon_{RL}(t - t') \mathcal{J}_{RL}(t') \right) \mathcal{E}_{RL}(0) \end{aligned} \quad (35)$$

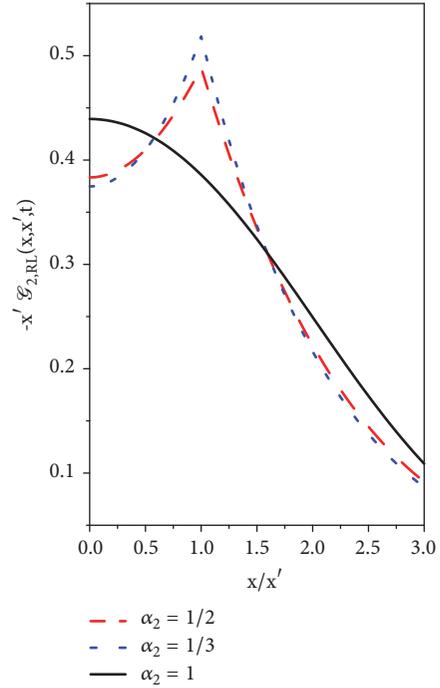


FIGURE 5: This figure illustrates the behavior of (31) by considering different values of α_2 . We consider, for simplicity, $\tau_{2,RL} = (x'^2/\mathcal{K}_2)^{1/\alpha_2}$, $k\tau_{2,RL}/x' = 1$, and $t = \tau_{2,RL}$, in arbitrary unities.

$$\begin{aligned} \mathcal{E}_{RL}(t) = & \int_0^t dt'' \omega(t - t'') \left\{ \int_0^{t''} \frac{dt'}{t'} \Upsilon_{RL}(t'' - t') \right. \\ & \cdot \int_0^\infty dx' \varphi_1(x') H_{1,1}^{1,0} \left[\frac{|x - x'|}{\sqrt{\mathcal{D}_2 t'^{\alpha}}} \right]_{(0,1)}^{(0,\alpha/2)} \\ & + \left((1 - k_s) \Upsilon_{RL}(t'') \right. \\ & \left. \left. - k_s \int_0^{t''} dt' \Upsilon_{RL}(t'' - t') \mathcal{J}_{RL}(t') \right) \mathcal{E}_{RL}(0) \right\}, \end{aligned} \quad (36)$$

For the case characterized by (4), i.e., the Caputo-Fabrizio fractional time operator, we obtain that

$$\begin{aligned} \rho_{1,CF}(0, t) = & \int_0^t \frac{dt'}{(4\pi D_1 t')^{3/2}} \Upsilon_{CF}(t - t') \\ & \cdot \int_0^\infty dx' \varphi_1(x') e^{-x'^2/4D_1 t'} + \left(\Upsilon_{CF}(t) \right. \\ & \left. - \int_0^t dt' \Upsilon_{CF}(t - t') \mathcal{J}_1(t') \right) \mathcal{E}_{CF}(0), \end{aligned} \quad (37)$$

$$\begin{aligned}
 \mathcal{E}_{CF}(t) = & \int_0^t dt'' \omega(t-t'') \left\{ \int_0^{t''} \frac{dt'}{(4\pi D_1 t')^{3/2}} \Upsilon_{CF}(t'' - t') \int_0^\infty dx' \varphi_1(x') e^{-x'^2/4D_1 t'} \right. \\
 & + \left. \left((1-k_s) \Upsilon_{CF}(t'') - k_s \int_0^{t''} dt' \Upsilon_{CF}(t''-t') \mathcal{F}_{CF}(t') \right) \mathcal{E}_{CF}(0) \right\}. \quad (38)
 \end{aligned}$$

The quantities $\Upsilon_{RL(CF)}(t)$ and $\mathcal{F}_{RL(CF)}(t)$ presented in previous equations are defined as follows:

$$\begin{aligned}
 \Upsilon_{RL(CF)}(t) = & -\mathcal{G}_{RL(CF)}(0,0;t) - \sum_{n=1}^{\infty} k_s^n \\
 & \cdot \int_0^t dt_n \mathcal{F}_{RL(CF)}(t-t_n) \mathcal{G}_{RL(CF)}(0,0;t-t_n) \\
 & \times \int_0^{t_n} dt_{n-1} \mathcal{F}_{RL(CF)}(t_n-t_{n-1}) \\
 & \cdot \mathcal{G}_{RL(CF)}(0,0;t_n-t_{n-1}) \cdots \\
 & \cdot \int_0^{t_2} dt_1 \mathcal{F}_{RL(CF)}(t_2-t_1) \\
 & \cdot \int_0^{t_1} dt' \mathcal{G}_{RL(CF)}(0,0;t_1-t') \mathcal{G}_{RL(CF)}(0,0;t') \quad (39)
 \end{aligned}$$

with

$$\begin{aligned}
 \mathcal{F}_{RL(CF)}(t) = & \delta(t) - \sum_{n=1}^{\infty} (-1)^n \int_0^t dt_n k_d(t-t_n) \\
 & \cdot \int_0^{t_n} dt_{n-1} k_d(t_n-t_{n-1}) \cdots \\
 & \cdot \int_0^{t_3} dt_2 k_d(t_2-t_1) \int_0^{t_2} dt_1 k_d(t_1). \quad (40)
 \end{aligned}$$

By using the previous equations, the inverse of Laplace transform of (17) and (18)

$$\begin{aligned}
 \rho_{1,RL(CF)}(x,t) = & - \int_0^\infty \mathcal{G}_{1,RL(CF)}(x,x';t) \varphi_1(x') dx' \\
 & + k_s \int_0^t dt'' \mathcal{F}_{RL(CF)}(t-t'') \int_0^{t''} \rho_1(0,t''-t') \\
 & \cdot \mathcal{G}_{1,RL(CF)}(x,0;t') - \left(\mathcal{G}_{1,RL(CF)}(x,0;t) \right. \\
 & \left. - \int_0^t dt' \mathcal{F}_{RL(CF)}(t-t') \mathcal{G}_{1,RL(CF)}(x,0;t') \right) \mathcal{E}(0) \quad (41)
 \end{aligned}$$

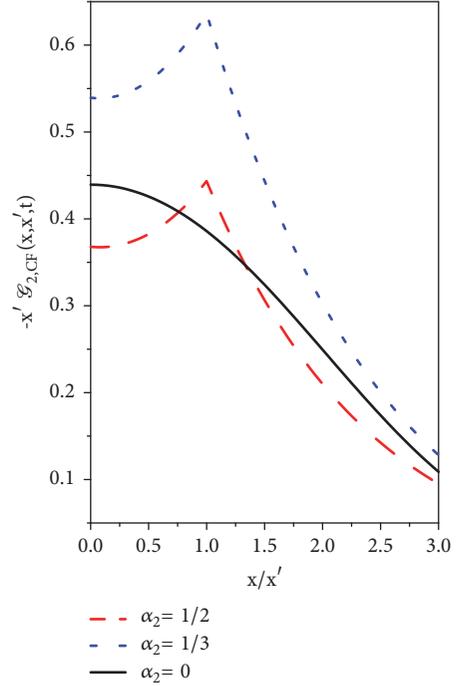


FIGURE 6: This figure illustrates the behavior of (32) by considering different values of α_2 . We consider, for simplicity, $\tau_{2,CF} = \sqrt{x'^2/D_2}$, $k\tau_{2,CF}/x' = 1$, $\tau_{2,CF} = 1$, and $\alpha_2 = 1/2$, in arbitrary unities.

and

$$\begin{aligned}
 \rho_{2,RL(CF)}(x,t) = & - \int_0^\infty \mathcal{G}_{2,RL(CF)}(x,x';t) \varphi_2(x') dx' \\
 & - \int_0^t dt'' k(t-t'') \\
 & \cdot \int_0^{t''} dt' \mathcal{G}_{2,RL(CF)}(x,0;t''-t') \rho_{1,RL(CF)}(0,t'). \quad (42)
 \end{aligned}$$

Equations (41) and (42) are the solutions for the scenario analyzed here, which is characterized by a diffusion with processes occurring on a surface. By using these results, it is possible to obtain the survival probability, which is defined as $S_{1(2)}(t) = \int_0^\infty dx \rho_{1(2)}(x,t)$ and it is related to the quantity of particles each species present in the bulk. In Figure 7, we illustrate the time behavior of the survival probability for species 1 and 2 by considering different fractional time operators. We observe that for the fractional operator related to the exponential kernel a stationary state is reached. Another interesting quantity to be analyzed is the mean square displacement, i.e., $(\Delta_{1(2)}x)^2 = \langle (x - \langle x \rangle_{1(2)})^2 \rangle_{1(2)}$, related to the spreading of the distributions of each species. For example, in absence of adsorption-desorption process ($k_s = 0$ and $\mathcal{E}(0) = 0$) with $k(t) = \text{constant}$, species 1 manifests two different regimes, when the Riemann-Liouville fractional time derivative is considered. In particular, for small times $(\Delta_1 x)^2 \sim t^\gamma$ and for long times $(\Delta_1 x)^2 \sim t^{\gamma/2}$. In this context, for the Caputo-Fabrizio fractional time operator, we have $(\Delta_1 x)^2 \sim t$ for

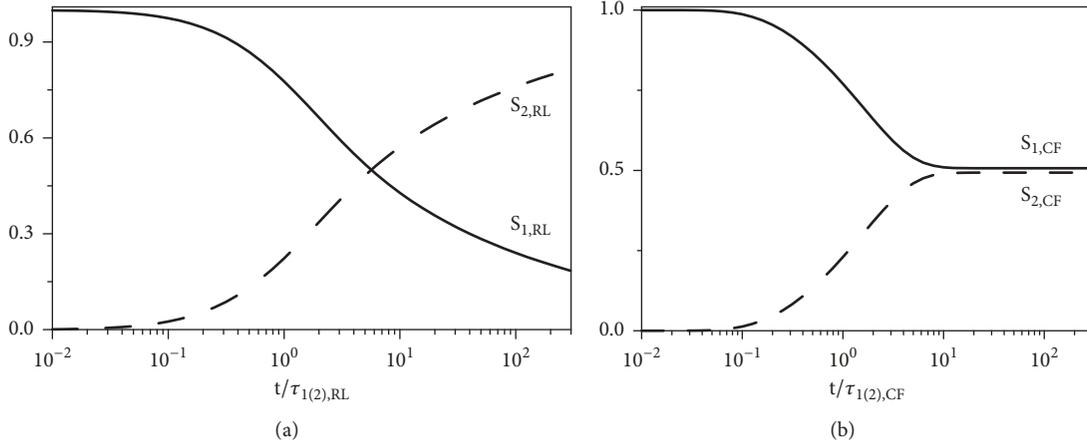


FIGURE 7: (a) illustrates the survival probability for species 1 and 2 by considering the Riemann-Liouville fractional time operator. We consider, for simplicity, $\tau_{1(2),RL} = (x'^2/\mathcal{K}_{1(2)})^{1/\alpha_{1(2)}}$, $k\tau_{1(2),RL}/x' = 1$, $\varphi_1(x) = \delta(x - x')$, $\varphi_2(x) = 0$, and $\alpha_{1(2)} = 1/2$, in arbitrary unities. (b) illustrates the survival probability for species 1 and 2 by considering the Caputo-Fabrizio fractional time operator. We consider, for simplicity, $\tau_{1(2),CF} = \sqrt{x'^2/D_{1(2)}}$, $k\tau_{1(2),CF}/x' = 1$, $\tau_{1(2),CF} = 1$, $\varphi_1(x) = \delta(x - x')$, $\varphi_2(x) = 0$, and $\alpha_{1(2)} = 1/2$, in arbitrary unities.

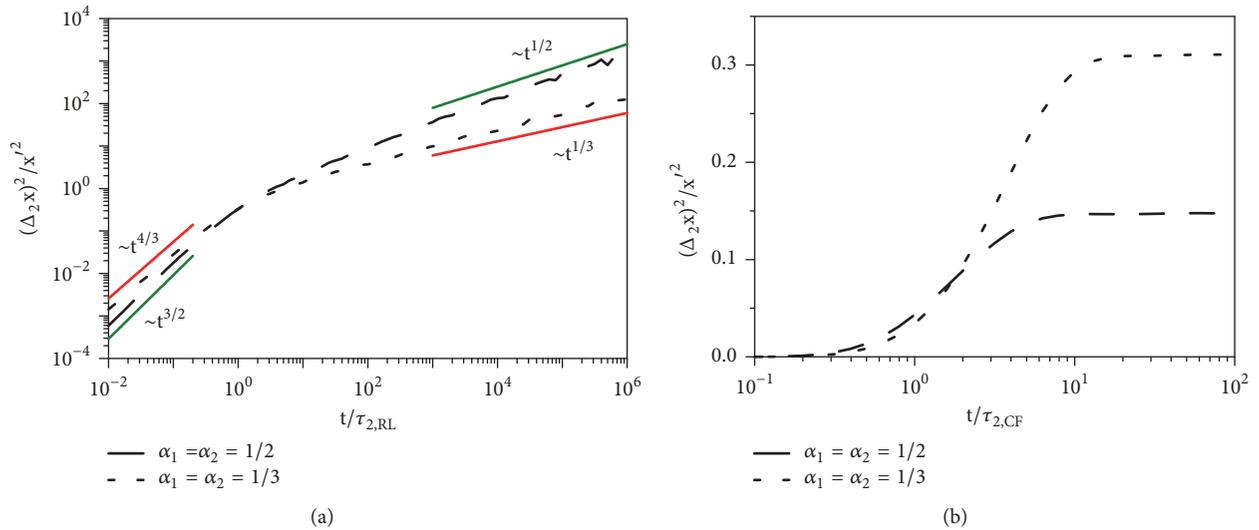


FIGURE 8: (a) illustrates the mean square displacement for species 2 for different values of $\alpha_{1(2)}$ by taking into account the Riemann-Liouville fractional time operator. We consider, for simplicity, $\tau_{1(2),RL} = (x'^2/\mathcal{K}_{1(2)})^{1/\alpha_{1(2)}}$, and $k\tau_{1,CF}/x' = 1$, $\varphi_1(x) = \delta(x - x')$, $\varphi_2(x) = 0$, and $\tau_{1(2),RL} = 1$, in arbitrary unities. (b) illustrates the mean square displacement for species 2 for different values of $\alpha_{1(2)}$ for species 2 by taking into account the Caputo-Fabrizio fractional time operator. We consider, for simplicity, $\tau_{1(2),CF} = \sqrt{x'^2/D_{1(2)}}$, and $k\tau_{1,CF}/x' = 1$, $\varphi_1(x) = \delta(x - x')$, $\varphi_2(x) = 0$, and $\tau_{1(2),CF} = 1$, in arbitrary unities.

small times and $(\Delta_1 x)^2 \sim \text{constant}$ for long times. For species 2, the behavior of the mean square displacement is illustrated in Figure 8 and, similar to species 1, the Riemann-Liouville fractional time derivative presents two different regimes. For the Caputo-Fabrizio fractional time operator, we observe a superdiffusion (see Figure 8(a)) or a transient (see Figure 8(b)) before reaching a stationary state. The initial behavior observed for species 2 is a consequence of the initial condition considered in this application.

3. Discussions and Conclusions

We have investigated a diffusion process in semi-infinite media in contact with a surface, where the particles may be adsorbed (and desorbed) or sorbed by the surface in order to promote though a reaction process the formation of another species of particle. For particles in the bulk, we have considered generalized diffusion equations, which may be related to a different types of fractional differential operator. In particular, we have considered the Riemann-Liouville and

Caputo-Fabrizio fractional time derivative. For each one of these operators, we have found the solutions and obtained the quantity of particles adsorbed by the surface as a function of time. The reaction process considered here occurs only on the surface and it is irreversible, i.e., $1 \rightarrow 2$. The particles obtained from the reaction process are released from the surface to the bulk. In the bulk, we have considered that reaction between the particles is absent; i.e., only the diffusion governs the dynamics of the particles in the bulk. The solutions for these cases have been obtained in terms of the Fox H-functions and generalized Mittag-Leffler functions which are usually related to anomalous diffusion. They have exhibited different diffusive behaviors depending on the conditions employed to analyze the system. In particular, for the situation discussed in Figure 8(a), we have verified that species 2 manifests a superdiffusive behavior for small times when the Riemann-Liouville fractional time operator is considered. For long times, the behavior is essentially governed by the diffusion equations and, consequently, subdiffusive behaviors were obtained. In Figure 8(b), we have analyzed the behavior of the mean square displacement for the Caputo-Fabrizio fractional time operator. For small times, we have a transient until the system reaches a stationary solution, i.e., a time independent solution, for long times. The stationary solution manifested by the system, when the Caputo-Fabrizio is employed, may be related to the resetting process connected to this operator as pointed out in [60]. Finally, we hope that the results presented here may be useful to discussion of situations related to anomalous diffusion.

Data Availability

No data were used to support this study.

Conflicts of Interest

The authors declare no conflicts of interest.

Acknowledgments

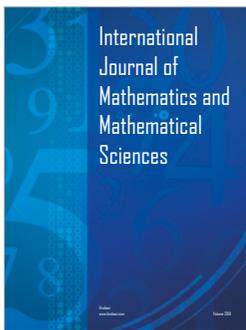
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