Non-Markovian diffusion equation and diffusion in a porous catalyst

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ABSTRACT

We revisit the problem of diffusion in a porous catalyst by incorporating in the diffusion equation fractional time derivatives and a spatial dependent diffusion coefficient in order to extend the usual description to situations which have an unusual behavior. In our analysis, we also consider a nonlocal reaction term of linear order. We obtain exact solutions for the profile of substance in the porous catalyst in terms of the Green function approach. The results show an anomalous behavior of the concentration profile spreading which may be connected to anomalous diffusion.

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1. Introduction

Diffusion is one of the most ubiquitous phenomena in nature. It is present in several contexts related, for example, to engineering [1], physics [2], and biology [3]. Typical diffusive phenomena can be found in a usual diffusion equation with an appropriate description which has as main characteristic the square mean displacement with a linear dependence on time, i.e., $\langle (r - \langle r \rangle)^2 \rangle \propto t$ [2,4]. This feature is related to the central limit theorem and also to the Markovian [4] nature of this stochastic process. However, a large number of experimental observations show that more complex processes, in which the mean-square displacement is not proportional to t, are also established in nature. These scenarios can be found, for instance, in atom deposition into a porous substrate [5], diffusion of high molecular weight polyisopropylacrylamide in nanopores [6], highly confined hard disk fluid mixtures [7], fluctuating particle fluxes [8], conservative motion in a $d = 2$ periodic potential [9], transport of fluid in porous media, diffusion on fractals [10–12], impedance spectroscopy [13] and many other interesting systems. Another typical example is the surface of a catalyst (see, for example, Fig. 1) which may require changes in the standard model of the diffusion in a porous catalyst [14] to describe the physical and chemical processes suitably [15]. In this sense, we investigated the

predictions which are obtained when fractional time derivatives [16] and an spatial time dependence on the diffusion coefficient [17,18] are incorporated in the diffusion. In particular, according to Giona and Roman in Ref. [19], the diffusion in porous structures is better described by using mathematical tools that provide memory effects, which is one of the main features of fractional derivatives [20,21].

Our analysis is focused on the diffusion equation:

$$\frac{\partial^\gamma}{\partial t^\gamma} \mathcal{C}(r, t) = \frac{1}{r^2} \frac{\partial}{\partial r} \left(r^2 \mathcal{D}(r) \frac{\partial}{\partial r} \mathcal{C}(r, t) \right) + \mathcal{R}(\mathcal{C}), \tag{1}$$

which accomplishes the radial symmetry where the diffusion coefficient is given by $\mathcal{D}(r) = \overline{\mathcal{D}}/r^\eta$. We also consider $0 < \gamma \le 1$, with $-1 < \eta$ and $0 \le r \le R$. This spatial dependence for the diffusion coefficient has been used to investigate unusual situations related to anomalous diffusion [24,25] such as diffusion on fractals [10,16,17], turbulence [26], fast electrons in a hot plasma in the presence of a electric field, financial models [27,28], and non-Markovian random walk [29]. Eq. (1) was also worked out by Povstenko [30] in the absence of nonlocal reaction terms with a constant diffusion coefficient. The fractional derivative considered here is the Caputo one [16] which is defined as

$$\frac{\partial^\gamma}{\partial t^\gamma} \mathcal{C}(r, t) = \frac{1}{\Gamma(n - \gamma)} \int_0^t d\bar{t} \frac{\mathcal{C}^{(n)}(r, \bar{t})}{(t - \bar{t})^{\gamma + 1 - n}}$$

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Nomenclature

$\mathcal{C}$	concentration
$\mathcal{G}$	Green function
$\mathcal{G}_f$	Green function in absence of reaction terms
$\mathcal{C}_s$	surface concentration
α	reaction rate
$\bar{\alpha}$	reaction rate constant
α_1	reaction rate constant
α_2	reaction rate constant
$\mathcal{D}$	diffusion coefficient
$\overline{\mathcal{D}}$	diffusion coefficient
r	radial coordinate
t	time
R	radius of the pellet
γ	dimensionless variable
η	dimensionless variable
μ	dimensionless variable

with $n-1 < \gamma < n$ and $\mathcal{C}^{(n)}(r,t)$ corresponding to the nth – time derivative of the concentration. The last term of Eq. (1) is a reaction term which is given by

$$\mathcal{R}(\mathcal{C}) = -\int_0^t \alpha(t-t')\mathcal{C}(r,t')dt',$$

where $\alpha(t)$ is a time function related to the nonlocal effects. In the absence of nonlocal effects we have $\alpha(t) = \bar{\alpha}\delta(t)$ and consequently the usual form of the reaction term of linear order is recovered. Eq. (1) is investigated by considering the conditions $\mathcal{C}(R,t) = \mathcal{C}_s(t)$ and $\mathcal{C}(r,0) = \overline{\mathcal{C}}(r)$, which correspond to the boundary and initial conditions. Note that the boundary condition considered here has a time dependence in order to cover situations that have a finite quantity of substance on the surface of the porous catalyst. The initial condition has a spatial dependence in order to simulate an initial concentration of substance in the porous catalyst. We obtain analytical solutions in terms of the Green function approach [31] and analyze the effect on the solution due to the nonlocal term when particular forms of $\alpha(t)$ are assumed. These developments are performed in Section 2. In Section 3, we present our discussions and conclusions.

2. Results and discussions

Let us start our analysis by considering Eq. (1) with $\alpha(t) = \bar{\alpha}\delta(t)$, i.e., a reaction term of linear order, and subjected to the conditions

discussed at the introduction. We also consider the initial concentration in the porous catalyst normalized, i.e., $\int_0^R dr\, r^2 \mathcal{C}(r,0) = 1$, when it is present. After these requirements, Eq. (1) can be written as

$$\frac{\partial^\gamma}{\partial t^\gamma}\mathcal{C}(r,t) = \frac{\overline{\mathcal{D}}}{r^2}\frac{\partial}{\partial r}\left(r^{2-\eta}\frac{\partial}{\partial r}\mathcal{C}(r,t)\right) - \bar{\alpha}\mathcal{C}(r,t) \quad (2)$$

with $0 < \gamma \le 1$ and $-1 < \eta$. Note that depending on the choice of the parameters γ and η, we may have a subdiffusive or a superdiffusive process. This feature may be verified by analyzing the free case, i.e., a simple diffusion without reaction terms, which has the mean square displacement given by $\langle (r - \langle r \rangle)^2 \rangle \propto t^{2\gamma/(2+\eta)}$ with $2\gamma/(2+\eta)$ less, equal or greater than one corresponding to sub-, normal or superdiffusion, respectively. The presence of nonlinear reaction terms with $\gamma = 1$ and $\eta = 0$ is discussed in [32,33]. Before using the Green function approach, we apply the Laplace transform in Eq. (2) yielding

$$\frac{\bar{\mathcal{D}}}{r^2}\frac{\partial}{\partial r}\left(r^{2-\eta}\frac{\partial}{\partial r}\mathcal{C}(r,s)\right) - (\bar{\alpha} + s^\gamma)\mathcal{C}(r,s) = -s^{\gamma-1}\mathcal{C}(r,0). \quad (3)$$

In terms of the Green function approach the solution of Eq. (3) can be written as follows:

$$\mathcal{C}(r,s) = -s^{\gamma-1}\int_0^R dr'r'^2\,\mathcal{G}(r,r',s)\mathcal{C}(r',0) + \bar{\mathcal{D}}R^{2-\eta}\mathcal{C}(R,s)\frac{\partial}{\partial r}\mathcal{G}(r,r',s)\bigg|_{r=R} \quad (4)$$

with the Green function $\mathcal{G}(r,r',s)$ determined by the equation

$$\frac{\overline{\mathcal{D}}}{r^2}\frac{\partial}{\partial r}\left(r^{2-\eta}\frac{\partial}{\partial r}\mathcal{G}(r,r',s)\right) - (\bar{\alpha} + s^\gamma)\mathcal{G}(r,r',s) = \frac{1}{r^2}\delta(r-r') \quad (5)$$

rendering the conditions $\mathcal{G}(R,t) = 0$ and $(\partial/\partial r)\mathcal{G}(r,t)|_{r=0} = 0$. Note that the first term of Eq. (4) corresponds to the spreading of the initial condition in the porous catalyst with the dynamic governed by Eq. (5). The second term is the influence of the surface on the diffusion in the porous catalyst. After some calculations, it is possible to show that the solution for Eq. (5) is given by

$$\mathcal{G}(r,s) = -\sum_{n=1}^\infty \frac{\mathcal{N}_n}{\bar{\alpha} + \mathcal{D}k_n^2 + s^\gamma}\psi(r,k_n)\psi(r',k_n) \quad (6)$$

with $\mathcal{N}_n = (2+\eta)/[R^{2+\eta}J_{\upsilon+1}(2k_n R^{1/2(2+\eta)}/(2+\eta))]$,

$$\psi(r,k_n) = r^{-1/2(1-\eta)}J_\upsilon\left(\frac{2k_n}{2+\eta}r^{1/2(2+\eta)}\right),$$
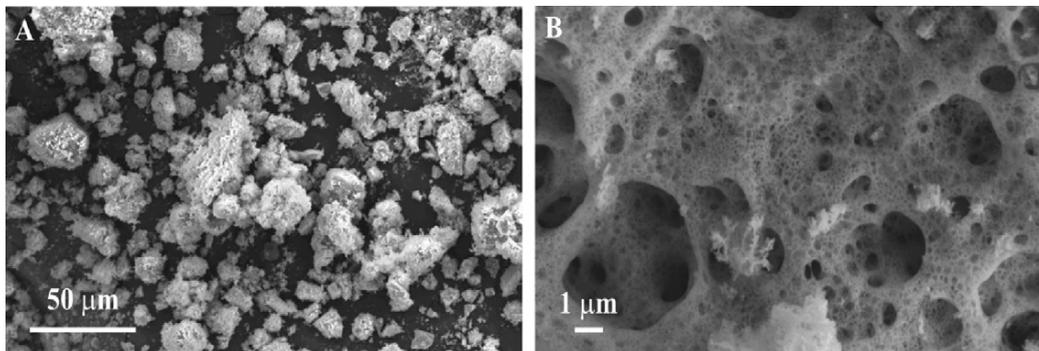
Fig. 1. These SEM figures at different magnification show the surface of different catalysts in, order to illustrate the irregular geometry and the catalytic pores which may be conveniently, described in terms of fractal considerations. Catalyst A is Ag/TiO$_2$ prepared by impregnation, procedure [22]; catalyst B is Pd/LaMnO$_3$·ZrO$_2$ prepared by solution combustion synthesis [23].
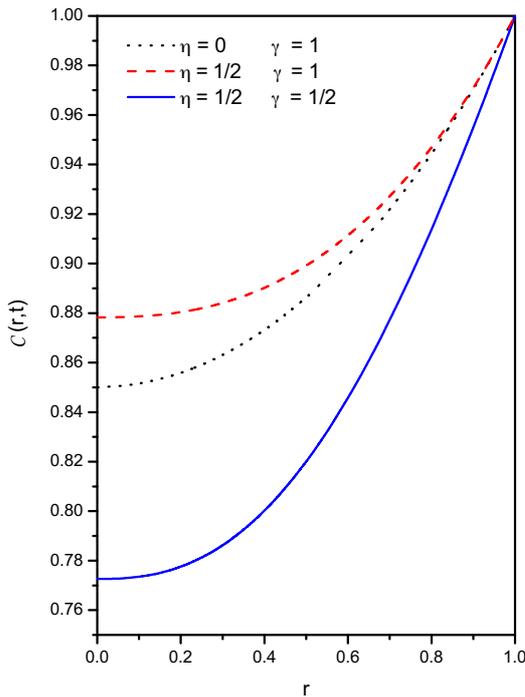
Fig. 2. Behavior of $\mathcal{C}(r, t)$ versus r in order to illustrate Eq. (8). We consider, for simplicity, $t = 2.0$, $\bar{D} = 1$, $\mathcal{C}_s = 1$, and $\bar{a} = 1$.

Fig. 3. Behavior of $\mathcal{C}(r, t)$ versus r in order to illustrate the influence of the spatial dependence on the solutions of Eq. (8). We consider, for simplicity, $t = 2.0$, $\gamma = 1/2$, $\bar{D} = 1$, $\mathcal{C}_s = 1$ and $\bar{a} = 1$.

where $J_\upsilon(x)$ is the Bessel function, $\upsilon = (1 - \eta)/(2 + \eta)$ and, k_n are determined by the equation $J_\upsilon(2k_n R^{1/2(2+\eta)}/(2 + \eta)) = 0$. The inverse Laplace transform of Eq. (6) yields
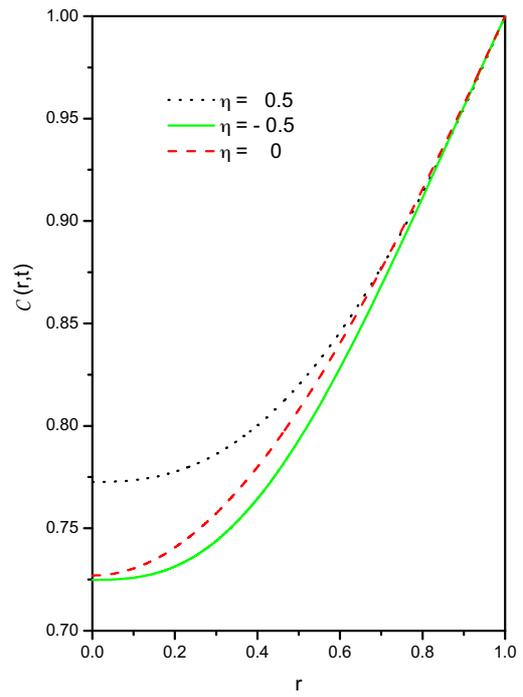
$$\mathcal{G}(r, t) = -t^{\gamma - 1} \sum_{n=1}^{\infty} \mathcal{N}_n \psi(r, k_n) \psi(r', k_n) E_{\gamma, \gamma}(-(\mathcal{D}k_n^2 + \bar{\alpha})t^\gamma), \quad (7)$$

where $E_{\alpha, \beta}(x)$ is the generalized Mittag–Leffler function [14] and it is defined as $E_{\gamma, \beta}(x) = \sum_{n=0}^{\infty} x^n / \Gamma(\beta + \gamma n)$. Note that for $\gamma = \beta = 1$, the exponential function is recovered. Also, the asymptotic limit is a power law [14], i.e., $E_{\gamma, \beta}(x) \sim -1/(\Gamma(\beta + \gamma)x)$ when $|x| \to \infty$. In this sense, it is also interesting to note that the relaxation of the solution governed by Eq. (7) is asymptotically characterized by a power law and not by an exponential one as the usual case. Another interesting property of the solution is that the fractional time derivative does not change the system's stationary state. However, the spatial dependence present on the diffusion coefficient changes the stationary state of the system which recovers the standard situation when $\eta = 0$.

By performing the inverse Laplace transform in Eq. (4), it follows that:

$$\mathcal{C}(r, t) = -\frac{1}{\Gamma(1 - \gamma)} \int_0^R dr' r'^2 \mathcal{C}(r', 0) \int_0^t d\bar{t} \frac{1}{(t - \bar{t})^\gamma} \mathcal{G}(r, r', \bar{t})$$

$$+ \bar{D} R^{2 - \eta} \int_0^t d\bar{t} \, \mathcal{C}(R, \bar{t}) \frac{\partial}{\partial r'} \mathcal{G}(r, r', t - \bar{t}) \bigg|_{r' = R}. \quad (8)$$

The second term of Eq. (8) represents the surface effect (as mentioned before in the Laplace space) and the stationary solution depends on the choice performed to the boundary condition. In particular, for the case $\lim_{t \to \infty} \mathcal{C}(R, t) = const$. A stationary solution is verified for the system, independently of the initial condition considered. In Fig. 2, we illustrate Eq. (8) by considering different

values of γ and η for the boundary conditions $(\partial/\partial r)\mathcal{C}(r, t)|_{r=0} = 0$ and $\mathcal{C}(R, t) = \mathcal{C}_s$ with $\mathcal{C}_s = const$. and the initial condition $\mathcal{C}(r, 0) = 0$. In order to perform the numerical calculations, we use the software Mathematica [34]. It shows that for the situation $\gamma \neq 1$ and $\eta \neq 0$ the substance diffusion in the porous catalyst is slower than the others cases shown in this figure.

In Fig. 3, we consider a fixed value of γ and different values of η to analyze the effect of the spatial dependence on the substance diffusion. It shows that the stationary solution obtained from Eq. 8 for $\mathcal{C}(R, t) = \mathcal{C}_s = const$. and $\mathcal{C}(r, 0) = 0$ is given by

$$\mathcal{C}(r, t) = \mathcal{C}_s \left(\frac{R}{r}\right)^{1/2(1 - \eta)} \frac{I_\upsilon((2/(2 + \eta))\sqrt{\bar{\alpha}/\bar{D}}r^{1/2(2+\eta)})}{I_\upsilon((2/(2 + \eta))\sqrt{\bar{\alpha}/\bar{D}}R^{1/2(2+\eta)})} \quad (9)$$

and for $\eta = 0$ it recovers the usual solution [1]. By using Eq. (8) it is possible to find the quantity $\mathcal{M}(t) = \int_0^R dr r^2 \mathcal{C}(r, t)$ which shows the quantity of substance in the catalytic layer as a function of time. In particular, it is given by

$$\mathcal{M}(r, t) = -\frac{1}{\Gamma(1 - \gamma)} \int_0^R dr r^2 \int_0^R dr' r'^2 \mathcal{C}(r', 0) \int_0^t d\bar{t} \frac{1}{(t - \bar{t})^\gamma} \mathcal{G}(r, r', \bar{t})$$

$$+ \bar{D} R^{2 - \eta} \int_0^R dr r^2 \int_0^t d\bar{t} \, \mathcal{C}(R, \bar{t}) \frac{\partial}{\partial r'} \mathcal{G}(r, r', t - \bar{t}) \bigg|_{r' = R}. \quad (10)$$

In Fig. 4, the behavior of $\mathcal{M}(t)$ versus t is illustrated for different values of γ and η by employing the same boundary conditions for the concentration. It shows that the system is governed by fractional derivatives (anomalous relaxation), which take a long time to become saturated, as compared to the system governed by the usual relaxations.

Now, we consider an arbitrary time dependence for the nonlocal reaction term and analyze possible effect on the concentration when some time dependent forms are performed for $\alpha(t)$. In particular, we consider two situations: $\alpha(t) = \bar{\alpha}t^{\mu - 1}/\Gamma(\mu)$ and
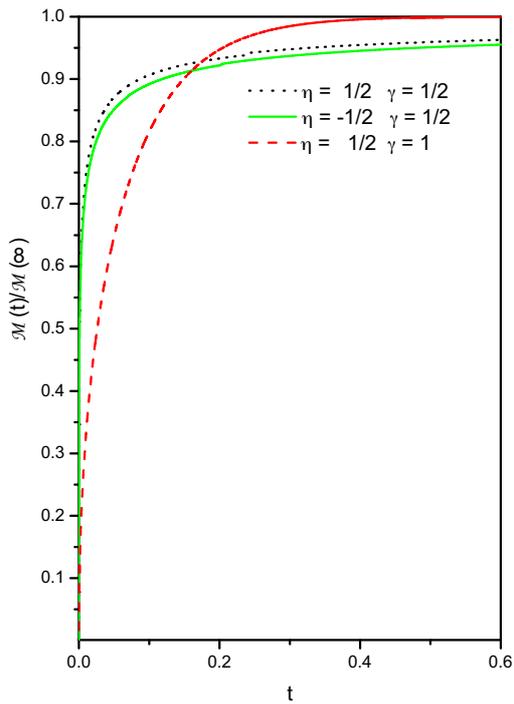
Fig. 4. Behavior of $\mathcal{M}(t)$ versus for different values of γ and η in order to illustrate Eq. (10). We consider, for simplicity $\bar{D} = 1$, $R = 1$, $\mathcal{C}_s = 1$, and $\bar{a} = 1$.

$\alpha(t) = \alpha_1 \delta(t) + \alpha_2 t^{\mu-1} / \Gamma(\mu)$. The first one is characterized by a power law time dependence and the second is a mixing between the usual reaction term and a nonlocal reaction term. The solution for this case is also given by Eq. (8). However the Green function for this case is given by

$$\mathcal{G}(r, s) = \mathcal{G}_f(r, s) - \sum_{n=1}^{\infty} \mathcal{N}_n \psi(r, k_n) \psi(r', k_n) \sum_{m=0}^{\infty} \frac{(-1)^m}{\Gamma(1+m)}$$
$$\times \int_0^t dt_m \alpha(t_m - t_{m-1}) \int_0^{t_m} dt_{m-1} \alpha(t_{m-1} - t_{m-2}) \cdots$$
$$\times \int_0^{t_2} dt_1 \alpha(t_2 - t_1) t_1^{(m+1)\gamma-1} E_{\gamma, \gamma+m\mu}^{(m)}(-\mathcal{D}k_n^2 t_1^\gamma), \quad (11)$$

where $E_{\alpha, \beta}^{(n)}(x) \equiv (d^n/dx^n) E_{\alpha, \beta}(x)$ and $\mathcal{G}_f(r, s)$ is given by

$$\mathcal{G}_f(r, s) = -t^{\gamma-1} \sum_{n=1}^{\infty} \mathcal{N}_n \psi(r, k_n) \psi(r', k_n) E_{\gamma, \gamma}(-\mathcal{D}k_n^2 t^\gamma). \quad (12)$$

Eq. (12) is the Green function of the diffusion equation in absence of reaction terms. For the case, $\alpha(t) = \bar{\alpha} t^{\mu-1} / \Gamma(\mu)$ Eq. (11) can be written as

$$\mathcal{G}(r, t) = \mathcal{G}_f(r, s) - \sum_{n=1}^{\infty} \mathcal{N}_n \psi(r, k_n) \psi(r', k_n) \Xi(\bar{\alpha}, t, k_n) \quad (13)$$

$$\Xi(\bar{\alpha}, t, k_n) = t^{\gamma-1} \sum_{m=1}^{\infty} \frac{(-\bar{\alpha})^m}{\Gamma(1+m)} t^{(\gamma+\mu)m} E_{\gamma, \gamma+m\mu}^{(m)}(-\mathcal{D}k_n^2 t^\gamma),$$

where the second term represents the effect of the nonlocal reaction term present in the diffusion equation. Fig. 5 shows the behavior of Eq. (13) for some values of μ and γ when $\alpha(t) = \bar{\alpha} t^{\mu-1} / \Gamma(\mu)$ taking the previous boundary conditions into account. The red dashed line is the usual case and the black dotted line and
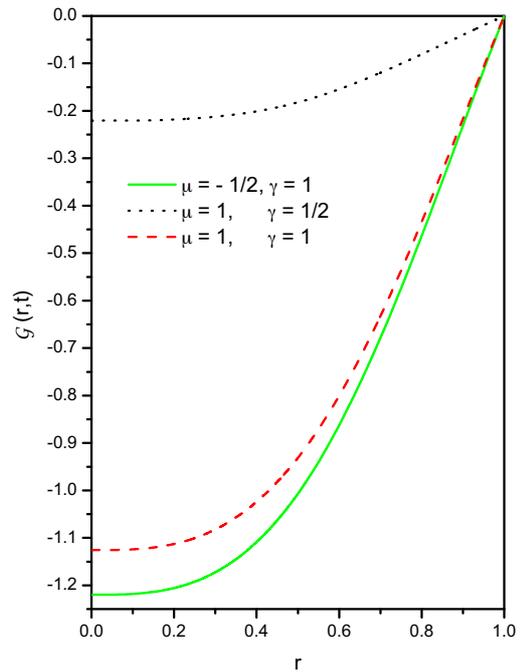

Fig. 5. Behavior of $\mathcal{G}(r, t)$ versus r for different values of γ, μ, α and η in order to illustrate Eq. (12) with $\alpha(t) = \bar{\alpha} t^{\mu-1} / \Gamma(\mu)$. We consider, for simplicity, $\bar{D} = 1$, $\eta = 1$, $t = 0.15$, $r' = 1/2$, $\alpha = 1$, $\mathcal{C}_s = 1$, and $R = 1$.

green solid line are the unusual situations which present an anomalous spreading when compared to the red dashed line. The black dotted line relaxes faster than the green solid line, which has a time dependence on the reaction term. Fig. 6 shows the behavior of Eq. (11) by considering the same boundary and initial conditions
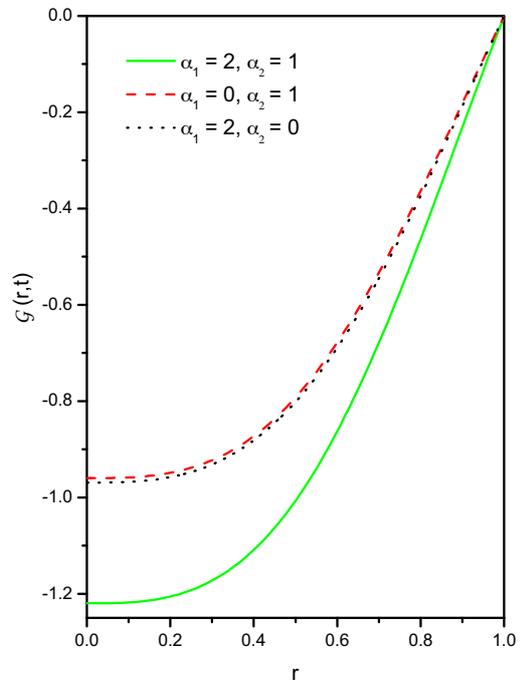

Fig. 6. Behavior of $\mathcal{G}(r, t)$ versus r for different values of α_1 and α_2 in order to illustrate Eq. (11) with $\alpha(t) = \alpha_1 \delta(t) + \alpha_2 t^{\mu-1} / \Gamma(\mu)$. We consider, for simplicity, $\bar{D} = 1$, $\eta = 1$, $t = 0.15$, $\mu = -1/2$, and $R = 1$.

when $\alpha(t) = \alpha_1 \delta(t) + \alpha_2 t^{\mu-1}/\Gamma(\mu)$, i.e., the reaction term is a mixing between the usual and the nonlocal terms. For this case the relaxation is slower when α_1 and α_2 are different from zero. (For interpretation of the references to color in this sentence, the reader is referred to the web version of the article.)

3. Conclusions

We have investigated the diffusion in a porous catalyst by considering a non-Markovian diffusion equation with a spatial dependent diffusion coefficient and fractional time derivatives. The solutions have shown an anomalous behavior when compared to the usual solution. In fact, the spatial dependence on the diffusion coefficient introduces dishomogeneity and the fractional derivative has memory effects. These features are ingredients to describe situations where the anomalous diffusion is present. In Figs. 1 and 3 the effects of fractional derivative are evident, the relaxation is slower than the usual case for $0 < \gamma \leq 1$. The spatial dependence on the diffusion coefficient may produce a relaxation which may be faster or slower than the usual one depending on the choice of the parameter η. Figs. 5 and 6 show the influence of the nonlocal term on the spreading of the solution, i.e., how the diffusion of the substance is influenced by this term. Finally, we hope that the results present here may be useful to investigate situations where the anomalous diffusion is present.

Acknowledgements

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