

A model for reversible reaction in a subdiffusive regime

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In this study, a model of reversible reaction in subdiffusive regime is set up by incorporating a reversible reaction term to a subdiffusion equation. Some models discussed previously are special cases of the model here and can be obtained by selecting proper parameters in the equations. Two different forms of the solution are given among which one is more suitable for computation. Though the physical interpretation is not clear, the discussions show that it is reasonable for describing the reaction-diffusion process. © 2009 American Institute of Physics.

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I. INTRODUCTION

The continuous time random walk (CTRW) method maybe the most powerful tool for deriving equations for anomalous diffusion.¹⁻³ Under different assumptions and based on different fractional operators,⁴ many equations were presented. However, the characteristics they perform are alike each other somehow. They all can describe the nature of the anomalous diffusion characterized by the temporal scaling of the mean-square displacement $\langle r^2(t) \rangle \sim t^\alpha$. The process is called subdiffusive if $0 < \alpha < 1$ while it is called superdiffusive if $\alpha > 1$.

When one substance is absorbed by another through which it can diffuse and with which it can react chemically during the diffusive process, the problems become more complicated. As for the normal reaction-diffusion system, the simplest models are of the form

$$\frac{\partial n}{\partial t} = D \frac{\partial^2}{\partial x^2} + f[n], \quad n = n(x, t), \quad (1)$$

where D is the diffusion coefficient and $f[n]$ is a linear or nonlinear function representing reaction kinetics. Saxena *et al.*⁵ discussed some generalizations of Eq. (1) to the fractional order. By the CTRW method, kinds of anomalous reaction-diffusion equations were derived,⁶⁻¹³ while the diffusion-controlled reactions in solution are usually treated by using a diffusion equation with appropriate boundary conditions.¹⁴ Hernández *et al.*¹⁵ studied the conditions for the appearance of a diffusion-driven instability and show that the restrictive conditions for a Turing instability are relaxed.

The reactions discussed before were mostly irreversible ones, while in this study, we will consider a reversible reaction occurring during the subdiffusive process. The problem discussed in the study is that one substance diffuses from the origin $x=0$ and can react with another whose concentration is always uniform.

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II. EQUATIONS OF THE DIFFUSION PROCESS WHEN A REVERSIBLE REACTION OCCURS

As for the problem introduced at the end of Sec. I, Henry *et al.*¹⁰ discussed equations such as

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

where $\mathcal{D}^{1-\alpha}$ is a fractional order temporal differential operator whose Laplace transform is

$$\mathcal{L}\{\mathcal{D}^{1-\alpha} f(t)\} = p^{1-\alpha} \mathcal{L}\{f(t)\}.$$

n , D , and K are the number density (or the concentration of the solute free to diffuse), the diffusion coefficient, and the generalized reaction rate, correspondingly. In the case $\beta = \alpha$, Eq. (2) is obtained by the CTRW method for instantaneous creation and annihilation processes by setting $K=1$ and $K=-1$, correspondingly, while in the case $\beta=1$, the equation is the fractional diffusion-reaction equation with standard reaction rate kinetics. In some cases, the rates of the diffusion and the reaction are comparable and the reaction is reversible. For the convenience of generalizing the above equation to reversible reaction case, we rewrite Eq. (2) as¹⁶

$$\frac{\partial n}{\partial t} = D\mathcal{D}^{1-\alpha} \left[\frac{\partial^2 n}{\partial t^2} \right] + K \frac{\partial S}{\partial t} \quad (3)$$

and

$$\frac{\partial S}{\partial t} = \mathcal{D}^{1-\beta} n, \quad (4)$$

where S is the concentration of the product. Equation (3) can be explained from the macroscopic point of view. To incorporate with reversible reaction, we generalize expression (4) for the simultaneous reaction to be of the type

$$\frac{\partial S}{\partial t} = \mathcal{D}^{1-\beta} \{\lambda n - \mu S\}, \quad (5)$$

where λ and μ are the rate constants of the forward and backward reactions, respectively. Thus the product is formed at a rate proportional to the concentration of the solute free to diffuse. When $\lambda=1$ and $\mu=0$, Eq. (3) recovers Eq. (2). In order to solve the equation, the following initial and boundary conditions are appended:

$$n(x,0) = \delta(x), \quad n(\pm\infty, t) = 0, \quad (6)$$

$$S(x,0) = 0. \quad (7)$$

III. SOLUTIONS TO THE EQUATIONS

To solve the equation subjects to the boundary and initial conditions, we use the Fourier and Laplace transform method. The Fourier and Laplace transforms of Eqs. (3) and (5) yield

$$p\hat{n} - 1 = -Dp^{1-\alpha} q^2 \hat{n} + Kp\hat{S} \quad (8)$$

and

$$p\hat{S} = p^{1-\beta} (\lambda \hat{n} - \mu \hat{S}). \quad (9)$$

From the above two equations, \hat{n} can be written as

$$\hat{n} = \frac{p^{\alpha-1}}{p^\alpha + Dq^2 - \frac{\lambda K p^\alpha}{p^\beta + \mu}} \quad (10)$$

$$= \frac{p^{\alpha-1}}{p^\alpha + Dq^2} + \sum_{j=1}^{\infty} \frac{(\lambda K)^j}{(p^\beta + \mu)^j j!} \frac{j! p^{\alpha-[1-\alpha j]}}{(p^\alpha + Dq^2)^{j+1}}. \quad (11)$$

Considering that

$$\mathcal{L}^{-1} \left\{ \frac{j! p^{\alpha-\gamma}}{(p^\alpha + a)^{j+1}} \right\} = t^{\alpha+\gamma-1} E_{\alpha,\gamma}^{(j)}(-at^\alpha), \quad j \geq 0, \quad (12)$$

and

$$\mathcal{L}^{-1} \left\{ \frac{1}{(p^\beta + \mu)^{j+1}} \right\} = \frac{t^{\beta(j+1)-1}}{\Gamma(j+1)} E_{\beta,\beta}^{(j)}(-\mu t^\beta), \quad j \geq 0, \quad (13)$$

where $E_{\alpha,\gamma}^{(j)}(y)$ is the j th order derivative of the Mittag-Leffler function, the reverse Laplace transform of \hat{n} can be obtained,

$$\hat{n} = E_{\alpha,1}(-Dq^2 t^\alpha) + \sum_{j=1}^{\infty} \frac{(\lambda K)^j t^{\beta j-1} E_{\beta,\beta}^{(j-1)}(-\mu t^\beta)}{\Gamma(j)j!} * E_{\alpha,1-\alpha j}^{(j)}(-Dq^2 t^\alpha), \quad (14)$$

where $f(t) * g(t)$ denotes the convolution of $f(t)$ and $g(t)$ defined by

$$f(t) * g(t) = \int_0^t f(t-\tau)g(\tau)d\tau.$$

Noting that for any j the first part of the convolution is independent of q and considering the linear properties of the inverse Fourier transform, we have

$$n = \mathcal{F}^{-1}\{E_{\alpha,1}(-Dq^2 t^\alpha)\} + \sum_{j=0}^{\infty} \frac{(\lambda K)^j t^{\beta j-1} E_{\beta,\beta}^{(j-1)}(-\mu t^\alpha)}{\Gamma(j)j!} * \mathcal{F}^{-1}\{E_{\alpha,1-\alpha j}^{(j)}(-Dq^2 t^\alpha)\}. \quad (15)$$

To calculate the inverse Fourier transformation of $E_{\alpha,1-\alpha j}^{(j)}(-Dq^2 t^\alpha)$, the procedure in the paper of Henry *et al.*¹⁰ is used. Refraining from too much repeats of the method, we just list some relations and formulas needed in the driving process:

$$E_{\alpha,1-\alpha j}^{(j)}(-Dq^2 t^\alpha) = H_{1,2}^{1,1}[Dq^2 t^\alpha |_{(0,1)(0,\alpha)}^{(-j,1)}], \quad (16)$$

$$\mathcal{M}\{\mathcal{F}[f(x)](q)\}(z) = 2\Gamma(z)\cos\left(\frac{\pi z}{2}\right)\mathcal{M}\{f(x)\}(1-z), \quad (17)$$

$$\mathcal{M}\{\phi(ax^p)\}(z) = \frac{1}{p} a^{-z/p} \mathcal{M}\{\phi(x)\}\left(\frac{z}{p}\right), \quad (18)$$

$$\sin(\pi s) = \frac{\pi}{\Gamma(1-s)\Gamma(s)}, \quad (19)$$

and

$$\Gamma(2s) = \frac{2^{(2s-1)}}{\sqrt{\pi}} \Gamma(s) \Gamma\left(s + \frac{1}{2}\right), \tag{20}$$

where $H_{p,q}^{m,n}[z]$ is the Fox H function¹⁷ defined by

$$H_{p,q}^{m,n}(z) = H_{p,q}^{m,n}\left[z \mid \begin{matrix} (a_1, A_1)_{1,p} \\ (b_1, B_1)_{1,q} \end{matrix}\right] = H_{p,q}^{m,n}\left[z \mid \begin{matrix} (a_1, A_1), \dots, (a_p, A_p) \\ (b_1, B_1), \dots, (b_q, B_q) \end{matrix}\right] = \frac{1}{2\pi i} \int_L \mathcal{H}_{p,q}^{m,n}(s) (z)^{-s} ds,$$

with

$$\mathcal{H}_{p,q}^{m,n}(s) = \frac{\prod_{j=1}^m \Gamma(b_j + B_j s) \prod_{l=1}^n \Gamma(1 - a_l - A_l s)}{\prod_{l=1}^p \Gamma(a_l + A_l s) \prod_{j=m+1}^q \Gamma(1 - b_j - B_j s)}.$$

$\mathcal{M}\{f(x)\}(z)$ is the Mellin transform of $f(x)$ defined by

$$\mathcal{M}\{f(x)\}(z) = \int_0^\infty x^{z-1} f(x) dx.$$

Consequently,

$$\begin{aligned} \mathcal{F}^{-1}\{E_{\alpha,1-\alpha}^{(j)}(-Dq^2 t^\alpha)\} &= \frac{1}{2} \frac{1}{\sqrt{4\pi D t^\alpha}} H_{1,2}^{2,0} \left[\frac{|x|}{\sqrt{4D t^\alpha}} \left| \begin{matrix} (1-\alpha/2, \alpha/2) \\ (0,1/2)(1/2+j,1/2) \end{matrix} \right. \right] \\ &= \frac{1}{\sqrt{4\pi D t^\alpha}} H_{1,2}^{2,0} \left[\frac{x^2}{4D t^\alpha} \left| \begin{matrix} (1-\alpha/2, \alpha) \\ (0,1)(1/2+j,1) \end{matrix} \right. \right]. \end{aligned} \tag{21}$$

As a result,

$$\begin{aligned} n &= \frac{1}{\sqrt{4\pi D t^\alpha}} H_{1,2}^{2,0} \left[\frac{x^2}{4D t^\alpha} \left| \begin{matrix} (1-\alpha/2, \alpha) \\ (0,1)(1/2,1) \end{matrix} \right. \right] \\ &+ \sum_{j=1}^\infty \frac{(\lambda K)^j t^{\beta j-1} E_{\beta,\beta}^{(j-1)}(-\mu t^\beta)}{\Gamma(j)j!} * \frac{1}{\sqrt{4\pi D t^\alpha}} H_{1,2}^{2,0} \left[\frac{x^2}{4D t^\alpha} \left| \begin{matrix} (1-\alpha/2, \alpha) \\ (0,1)(1/2+j,1) \end{matrix} \right. \right]. \end{aligned} \tag{22}$$

Similarly, since $\hat{S} = \lambda \hat{n} / (p^\beta + \mu)$ and $\widetilde{\partial S / \partial t} = \lambda \hat{n} p / (p^\beta + \mu)$ the solution for S and $\partial S / \partial t$ can be written as

$$S = \lambda \sum_{j=0}^\infty \frac{(\lambda K)^j t^{\beta(j+1)-1} E_{\beta,\beta}^{(j)}(-\mu t^\beta)}{\Gamma(j+1)j!} * \frac{1}{\sqrt{4\pi D t^\alpha}} H_{1,2}^{2,0} \left[\frac{x^2}{4D t^\alpha} \left| \begin{matrix} (1-\alpha/2, \alpha) \\ (0,1)(1/2+j,1) \end{matrix} \right. \right] \tag{23}$$

and

$$\frac{\partial S}{\partial t} = \lambda \sum_{j=0}^\infty \frac{(\lambda K)^j t^{\beta(j+1)-2} E_{\beta,\beta-1}^{(j)}(-\mu t^\beta)}{\Gamma(j+1)j!} * \frac{1}{\sqrt{4\pi D t^\alpha}} H_{1,2}^{2,0} \left[\frac{x^2}{4D t^\alpha} \left| \begin{matrix} (1-\alpha/2, \alpha) \\ (0,1)(1/2+j,1) \end{matrix} \right. \right], \tag{24}$$

correspondingly.

IV. DISCUSSION ON THE EQUATION AND SOLUTION

A. Computation of the solution

The solution (22) obtained in Sec. III is clear and compact. Moreover, in the case $\Delta = B_1 + B_2 - A_1 > 0$ and $B_1(b_2+k) \neq B_2(b_1+l)(k, l \in \mathbb{N})$, the H function of the following form can be computed using its series expansion:

$$H_{1,2}^{2,0}[z]_{(b_1, B_1)(b_2, B_2)}^{(a_1, A_1)} = \sum_{l=0}^{\infty} h_{1,l} z^{(b_1+l)/B_1} + \sum_{l=0}^{\infty} h_{2,l} z^{(b_2+l)/B_2}, \quad (25)$$

where

$$h_{1,l} = \frac{(-1)^l \Gamma\left(b_2 - [b_1 + l] \frac{B_2}{B_1}\right)}{l! B_1 \Gamma\left(a_1 - [b_1 + l] \frac{A_1}{B_1}\right)}$$

and

$$h_{2,l} = \frac{(-1)^l \Gamma\left(b_1 - [b_2 + l] \frac{B_1}{B_2}\right)}{l! B_2 \Gamma\left(a_1 - [b_2 + l] \frac{A_1}{B_2}\right)}.$$

It is easy to verify that the conditions mentioned above are satisfied by the H function appears in solutions (22)–(24) and the series is appropriate to be used for computation of the H function. However, because of the convolution, in order to compute the solution (22), all values of the function

$$\frac{1}{\sqrt{4Dt^\alpha}} H_{1,2}^{2,0} \left[\frac{x^2}{4Dt^\alpha} \middle| \begin{matrix} (1-\alpha/2, \alpha) \\ (0,1)(1/2+j,1) \end{matrix} \right]$$

must be obtained in the interval $[0, t]$. Noting that $1/\sqrt{4Dt^\alpha}$ tends to infinity when t tends to 0, computing the solution using the series form of H function and using the traditional method of computing the convolutions may go beyond the computational capabilities of even an advanced computer. To avoid the appearance of the infinite value, we will rewrite the solution using the integral form of the H function and calculate the convolution first. With a proper integral path L [separating the poles of $\Gamma(s)$ and $\Gamma(1/2+j+s)$], the H function that appeared in (22) can be written as

$$H_{1,2}^{2,0} \left[\frac{x^2}{4Dt^\alpha} \middle| \begin{matrix} (1-\alpha/2, \alpha) \\ (0,1)(1/2+j,1) \end{matrix} \right] = \frac{1}{2\pi i} \int_L \mathcal{H}_{1,2}^{2,0}(s) \left(\frac{x^2}{4Dt^\alpha} \right)^{-s} ds, \quad (26)$$

where

$$\mathcal{H}_{1,2}^{2,0}(s) = \frac{\Gamma(s) \Gamma\left(\frac{1}{2} + j + s\right)}{\Gamma\left(1 - \frac{\alpha}{2} + \alpha s\right)}.$$

So, for every $j \geq 1$, the convolution can be written as

$$\frac{(\lambda K)^j}{j! \Gamma(j) \sqrt{4\pi D}} \int_0^t \tau^{\beta j - 1} E_{\beta, \beta}^{(j-1)}(-\mu \tau^\beta) (t - \tau)^{-\alpha/2 + \alpha s} \frac{1}{2\pi i} \int_L \mathcal{H}_{1,2}^{2,0}(s) \left(\frac{x^2}{4D} \right)^{-s} ds d\tau. \quad (27)$$

Changing the order of the integral and writing $E_{\beta, \beta}^{(j-1)}(-\mu \tau^\beta)$ in its series form

$$E_{\beta, \beta}^{(j-1)}(-\mu \tau^\beta) = \sum_{k=0}^{\infty} \frac{\Gamma(k+j) (-\mu \tau^\beta)^k}{k! \Gamma(\beta(j+k))},$$

(27) becomes

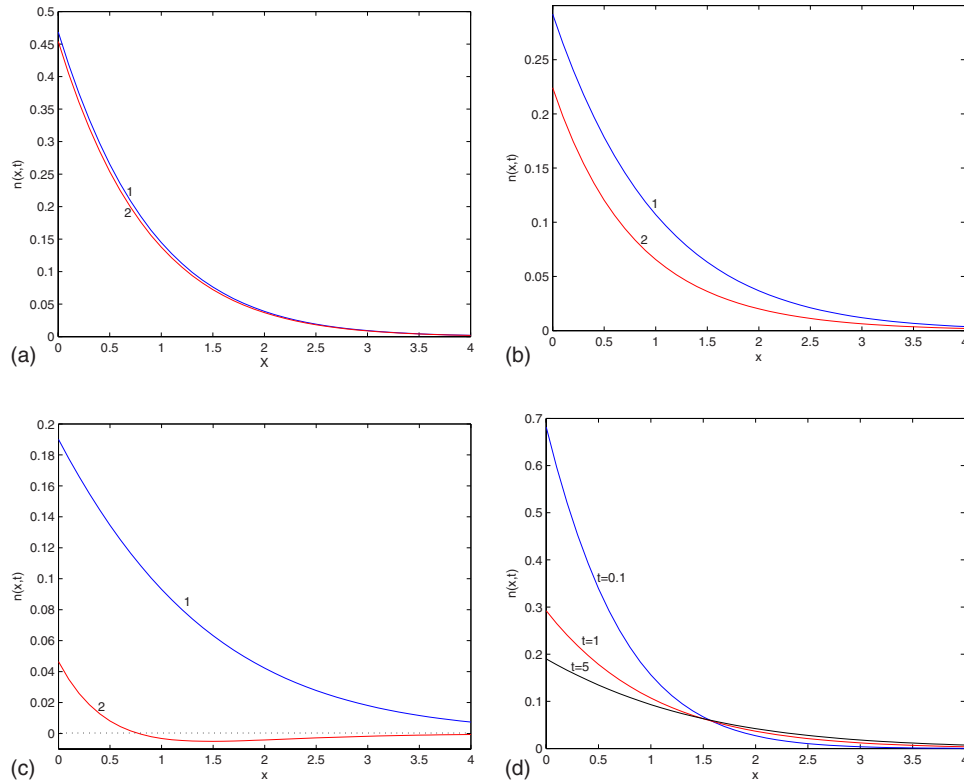


FIG. 1. (Color online) The infinite solution of Eqs. (3)–(5) with $K=-1$, $D=1$, $\lambda=1$, $\alpha=0.5$, and $\beta=1$. Curves 1 denote the concentration of the diffusive substance with reversible reaction ($\mu=1$) while curves 2 denote the irreversible one ($\mu=0$). (a) $t=0.3$, (b) $t=1$, and (c) $t=5$; (d) is a full picture of the concentration profiles at various times in the case $\mu=1$.

$$\begin{aligned} & \frac{1}{\sqrt{4\pi D}} \sum_{k=0}^{\infty} C_1(k,j) \frac{1}{2\pi i} \int_L \mathcal{H}_{1,2}^{2,0}(s) \left(\frac{x^2}{4D}\right)^{-s} \int_0^t \tau^{\beta(j+k)-1} (t-\tau)^{-\alpha/2+\alpha s} d\tau ds \\ &= \frac{t^{\beta j}}{\sqrt{4\pi D} t^\alpha} \sum_{k=0}^{\infty} C_2(k,j) (-\mu t^\beta)^k \frac{1}{2\pi i} \int_L \overline{\mathcal{H}}_{1,2}^{2,0}(s) \left(\frac{x^2}{4Dt^\alpha}\right)^{-s} ds, \end{aligned} \tag{28}$$

where

$$C_1(k,j) = \frac{(\lambda K)^j (-\mu)^k \Gamma(k+j)}{j! \Gamma(j) k! \Gamma(\beta(j+k))},$$

$$C_2(k,j) = \frac{(\lambda K)^j \Gamma(k+j)}{j! \Gamma(j) k!},$$

and

$$\overline{\mathcal{H}}_{1,2}^{2,0}(s) = \frac{\Gamma(s) \Gamma\left(\frac{1}{2} + j + s\right)}{\Gamma\left(1 - \frac{\alpha}{2} + \beta(j+k) + \alpha s\right)}.$$

During the above process, the definition of the *beta* function

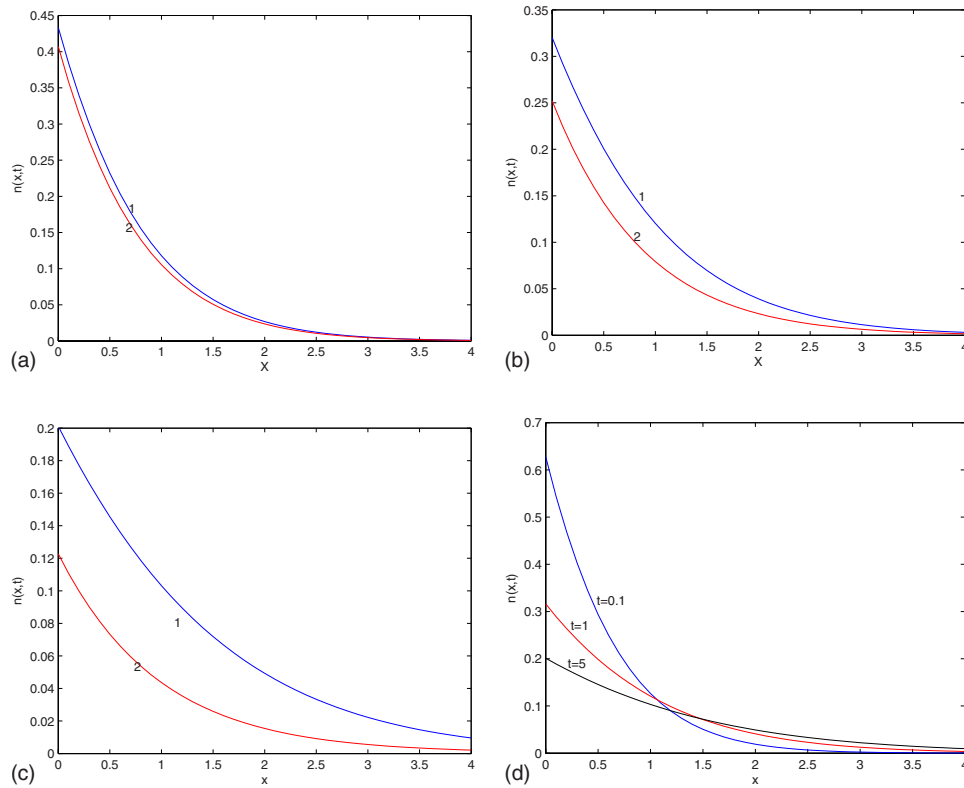


FIG. 2. (Color online) The infinite solution of Eqs. (3)–(5) with $K=-1$, $D=1$, $\lambda=1$, $\alpha=0.5$, and $\beta=0.5$. Curves 1 denote the concentration of the diffusive with reversible reaction ($\mu=1$) while curves 2 denote the irreversible one ($\mu=0$). (a) $t=0.3$, (b) $t=1$, and (c) $t=5$; (d) is a full picture of the concentration profiles at various times in the case $\mu=1$.

$$B(m,n) = \int_0^1 (1-\tau)^{m-1} \tau^{n-1} d\tau = \frac{\Gamma(m)\Gamma(n)}{\Gamma(m+n)} \tag{29}$$

is used. Employing the notation of H function and formula (28), we can obtain another form of the solution (22) as

$$n = \frac{1}{\sqrt{4\pi Dt^\alpha}} \left\{ H_{1,2}^{2,0} \left[\frac{x^2}{4Dt^\alpha} \middle| \begin{matrix} (1-\alpha/2, \alpha) \\ (0,1)(1/2,1) \end{matrix} \right] + \sum_{j=1}^{\infty} \sum_{k=0}^{\infty} \frac{(\lambda K)^j t^{\beta j}}{j! \Gamma(j)} \frac{\Gamma(k+j)(-\mu t^\beta)^k}{k!} H_{1,2}^{2,0} \left[\frac{x^2}{4Dt^\alpha} \middle| \begin{matrix} (1-\alpha/2+\beta(j+k), \alpha) \\ (0,1)(1/2+j,1) \end{matrix} \right] \right\}. \tag{30}$$

Though there are double summations in the above expression, each series can be tackled by computer conveniently. Furthermore, when $\mu=0$, it is obvious that the solution expressed by (30) becomes the solution by Henry *et al.*¹⁰ immediately. However, the solution (22) is not so obvious. The computations of S and $\partial S/\partial t$ can be treated by the same procedure.

B. Discussions on special cases

In Figs. 1 and 2, under different cases, the infinite domain solutions of Eq. (3) are shown at various times. The parameters we chosen here are $\alpha=1/2$, $K=-1$, $\lambda=1$, and $D=1$. In Figs. 1(a)–1(c) and 2(a)–2(c), curves 1 show the concentration of the diffusive substance in the case $\mu=1$, i.e., when backward reaction occurs. From Ref. 10 or Fig. 1(c), we can see that when $\mu=0$ and $\beta=1$, the solution would become negative at $t=5$ and is unrealistic. However, both for

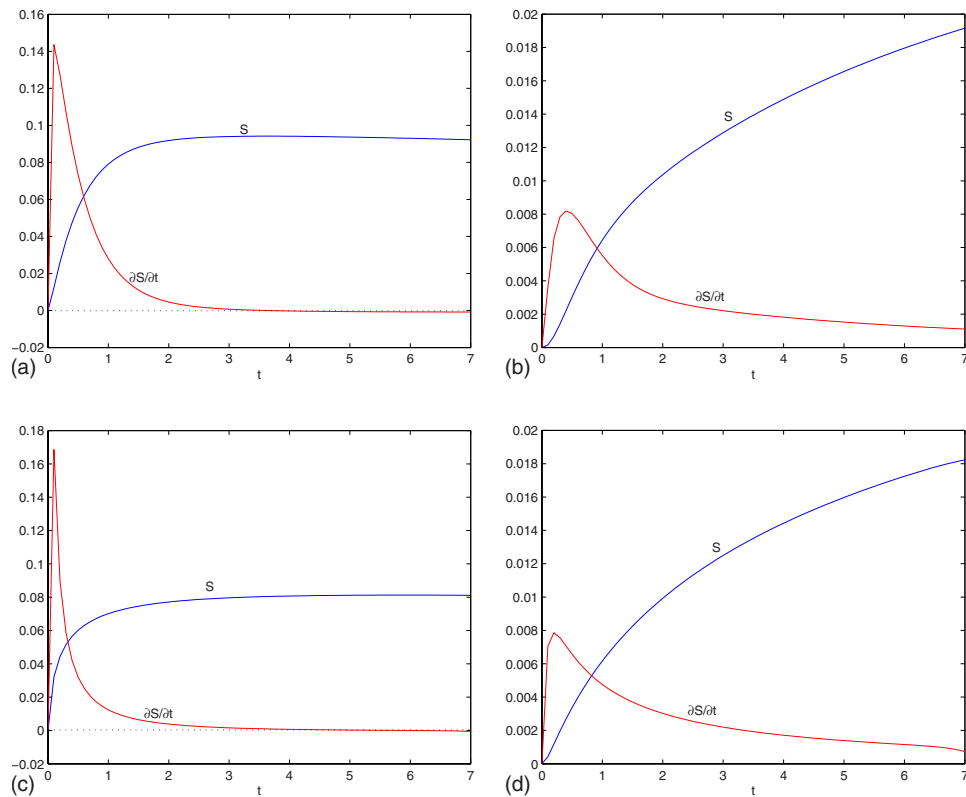


FIG. 3. (Color online) Profiles of S and $\partial S/\partial t$ at various positions with $K=-1$, $D=1$, $\lambda=1$, $\mu=1$, and $\alpha=0.5$. (a) and (b) for the case $\beta=1$ while (c) and (d) for the case $\beta=0.5$. [(a) and (c)] $x=1$; [(b) and (d)] $x=3$.

$\beta=1$ and $\beta=0.5$, the solutions of the model in the case $\mu=1$ for various times are always positive. It is obvious that the distance between curve 1 and curve 2 represents the influence of the backward reaction. Because of the assumption $S(0)=0$, the influence should be more and more profound as time progress. Both Figs. 1 and 2 can show the property.

In Fig. 3, the concentration of the product S and its rate of change $\partial S/\partial t$ are shown at $x=1$ and $x=3$. The figures show that the concentration of the product increases first and then decrease but the rate of decrease (dominated by the values of $\partial S/\partial t$) is rather slow. Comparisons between Figs. 3(a) and 3(b) and Figs. 3(c) and 3(d) show the conclusion that the operator $\mathcal{D}^{1-\beta}$ before the reaction term $\partial S/\partial t$ when $\beta=0.5$ delays the reaction process.

V. SUMMARY AND CONCLUSION

In the study, we present a heuristic model for reversible reactions in the subdiffusive regime by generalizing the reaction term in the model by Henry *et al.*¹⁰ to a reversible one (5). Figures 1–3 show that the model performs well even in the case the reaction term is local ($\beta=1$). Figure 1(c) shows that the breaking down of the irreversible reaction-diffusion equation ($\beta=1$, $\mu=0$, $\lambda=1$) is conquered.

Though some effects of introduction of the fractional operator $\mathcal{D}^{1-\beta}$ can be confirmed, the physical interpretation of the model is not clear. Furthermore, the assumption of infinite domain must be improved and the influence of the boundary condition must be considered.¹⁸ These problems are also the ones that must be solved in the development of applications of fractional calculus.

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