

First-passage time for the g -subdiffusion process of vanishing particlesTadeusz Kosztolowicz ^{*}*Institute of Physics, Jan Kochanowski University, Uniwersytecka 7, 25-406 Kielce, Poland*

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Subdiffusion equation and molecule survival equation, both with Caputo fractional time derivatives with respect to other functions g_1 and g_2 , respectively, are used to describe diffusion of a molecule that can disappear at any time with a constant probability. The process can be interpreted as an “ordinary” subdiffusion and “ordinary” molecule survival process in which timescales are changed by the functions g_1 and g_2 . We derive the first-passage time distribution for the process. The mutual influence of subdiffusion and molecule-vanishing processes can be included in the model when the functions g_1 and g_2 are related to each other. As an example, we consider the processes in which subdiffusion and molecule survival are highly related, which corresponds to the case of $g_1 \equiv g_2$.

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Introduction. In media where diffusion of molecules is very hindered, subdiffusion can occur. When modeling the subdiffusion process, it is usually assumed that the mean waiting time for a molecule to jump is infinite. The distribution of this time $\tilde{\psi}$ has a heavy tail, $\tilde{\psi}(\tau) \sim 1/\tau^{\alpha+1}$, where $\alpha \in (0, 1)$ is a subdiffusion parameter [1–3]. In a homogeneous medium, where the structure does not change with time, subdiffusion can be described by an “ordinary” subdiffusion equation with the “ordinary” fractional Caputo time derivative of the order α ; see Eq. (2), presented later. A changing timescale may cause subdiffusion to be accelerated or delayed. An example is the subordinate method, in which a stochastic process changing this scale is involved [4–7]. Recently a subdiffusion equation with a fractional time Caputo derivative with respect to another function g (g -subdiffusion equation) has been proposed [8–10]. The deterministic function g rescales the time variable in the “ordinary” subdiffusion model. For example, the waiting time for the molecule to jump in the g subdiffusion model is described by the ψ distribution, which is related to $\tilde{\psi}$ as $\psi(\tau) = \tilde{\psi}(g(\tau))$ [10].

We study the subdiffusion of a molecule that can be eliminated at any moment from further diffusion, and the molecule can be retained permanently or decayed; we treat both processes as “vanishing” of the molecule. We assume that the probability density of a molecule disappearing does not depend on a molecule position. Molecule elimination and diffusion processes can be related to each other. An example is diffusion of an antibiotic through a bacterial biofilm. Bacteria have different defense mechanisms against the action of the antibiotic; see Refs. [11,12] and the references cited in [13,14]. One of them is biofilm compaction, which makes it difficult for antibiotic molecules to diffuse and even stops them. The permanent arrest of the molecule can be treated as its “disappearance,” because it is eliminated from further diffusion. Since the antibiotic mainly attacks rapidly growing

bacteria, another defense mechanism is to temporarily stop the bacteria from multiplying. This mechanism even facilitates diffusion of the antibiotic through the biofilm. Another example is subdiffusion of antibiotic molecules in a medium having a “plum pudding” structure. This process can occur when a person infected with cystic fibrosis is also infected with *Pseudomonas aeruginosa* bacteria [14]. The “pudding” represents the cystic fibrosis mucus, and the “plums” represent the *Pseudomonas aeruginosa* biofilm. The purpose of the antibiotic is to kill the *Pseudomonas aeruginosa* bacteria whose defense mechanisms can destroy the antibiotic molecules. In this case, slowing subdiffusion in the “pudding” may increase the chance of avoiding the decay of the antibiotic molecules. These examples show that the processes of diffusion and decay of molecules can be related to each other. Slowing down molecule diffusion can cause an increase as well as a decrease of their disappearance probability. Thus, relations between diffusion and processes that lead to a molecule’s disappearance may be unique to a given process. Later we will pay attention to the process in which subdiffusion and the molecule survival are highly related, and transport properties of a medium change periodically. Below we mention a few processes that motivate the involvement of periodic functions in the model. The oscillating changes in the structure of the biofilm may be caused by the interdependence between the outer and inner cells. Outer cells can periodically change the availability of nutrients to the inner cells. Such periodic changes of biofilm structure increase the effectiveness of the biofilm defense against the action of the antibiotic [15]. Mechanical oscillations during the ultrasonication, which lead to periodic changes in the biofilm structure, can partially destroy the basic structure of biofilm and increase diffusion of antibiotics into the deeper layer of biofilm [16]. Subinhibitory levels of antibiotics can alter the structure of the biofilm, leading to an increase and then a decrease in the mass of biofilm, which may be followed by an increase in the mass [17]. Such a process can be interpreted as periodic with a time-dependent period.

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We consider the following problem. A molecule is located at point x_0 at the initial time $t = 0$ in a one-dimensional homogeneous system. We find the probability distribution of time that a molecule reaches the point x_M for the first time. This problem is potentially of great importance in modeling the transport of antibiotics in bacterial biofilms, and of an epidemic spreading when the arrival of an infected individual to some point may create a new source for the spread of the epidemic.

Model. We assume that the disappearance of a molecule is independent of its position. Let $P(x, t|x_0)$ be a probability density (Green’s function) that the molecule is at the point x at time t when particle decay would be “turned off,” where x_0 is the initial molecule position, and let $\rho(t)$ be a probability that the molecule still exists at time t . Then the Green’s function P_ρ for the vanishing molecule reads

$$P_\rho(x, t|x_0) = \rho(t)P(x, t|x_0). \tag{1}$$

We also assume that in some initial time interval a diffusion process is “ordinary” subdiffusion and the molecule-vanishing process is described by the “ordinary” decay equation. The “ordinary” subdiffusion is described by the following equation:

$$\frac{{}^C\partial^\alpha P(x, t|x_0)}{\partial t^\alpha} = D \frac{\partial^2 P(x, t|x_0)}{\partial x^2}, \tag{2}$$

where ${}^C\partial^\alpha f(t)/\partial t^\alpha = \int_0^t (t-u)^{-\alpha} f'(u) du / \Gamma(1-\alpha)$ is the “ordinary” Caputo fractional derivative, $0 < \alpha < 1$, $f'(t) = df(t)/dt$, and D is a subdiffusion coefficient given in units of m^2/s^α .

A frequently used equation to describe the disappearance of a molecule is $d\phi(t)/dt = -\lambda\phi(t)$, where ϕ is a probability density that molecule decays at time t and λ is a decay rate. The solution to this equation reads $\phi(t) = \lambda e^{-\lambda t}$. The probability that the molecule still exists at time t is $\rho(t) = 1 - \int_0^t \phi(u) du = e^{-\lambda t}$. This function fulfils the molecule survival equation (MSE) $d\rho(t)/dt = -\lambda\rho(t)$, with the initial condition $\rho(0) = 1$. However, if the function ρ has a heavy tail, $\rho(t) \sim 1/t^\beta$ with $\beta \in (0, 1)$ when $t \rightarrow \infty$, the MSE involves “ordinary” fractional Caputo derivative of the order β ,

$$\frac{d^\beta \rho(t)}{dt^\beta} = -\lambda\rho(t), \tag{3}$$

and the unit of λ is $1/s^\beta$.

We assume that at some initial stage the process is described by Eqs. (2) and (3). In the further course of the process, subdiffusion and the molecule-vanishing process may change due to the influence of external factors and/or due to the interaction of the processes. In order to describe the processes in the whole time domain, we assume that they are described by equations in which “ordinary” Caputo derivatives are replaced by g -Caputo derivatives, see Eq. (4) presented below. However, for short time the new equations should take the form of Eqs. (2) and (3).

The g -Caputo fractional derivative ${}^C d_g^\alpha f(t)/dt^\alpha$ of the order α with respect another function g is defined for $0 < \alpha < 1$ as [18–20]

$$\frac{{}^C d_g^\alpha f(t)}{dt^\alpha} = \frac{1}{\Gamma(1-\alpha)} \int_0^t [g(t) - g(u)]^{-\alpha} f'(u) du, \tag{4}$$

where $f'(t) = df(t)/dt$, the function g fulfils the conditions $g(0) = 0$, $g(\infty) = \infty$, and $g'(t) > 0$ for $t > 0$. The values of function g are given in a time unit. When $g(t) = t$, the g -Caputo fractional derivative takes the form of the “ordinary” Caputo derivative. For $\alpha = 1$, the g -Caputo derivative takes the form of the Riemann-Stieltjes derivative ${}^C d_g^1 f(t)/dt^\alpha \equiv f_g^{[1]} = f'(t)/g'(t)$.

We involve the g -Caputo fractional time derivative in both the subdiffusion equation and the molecular survival equation. We assume that g subdiffusion is controlled by a function g_1 , and the g -molecule survival process by a function g_2 :

$$\frac{{}^C d_{g_1}^\alpha P(x, t|x_0)}{\partial t^\alpha} = D \frac{\partial^2 P(x, t|x_0)}{\partial x^2}, \tag{5}$$

$$\frac{{}^C d_{g_2}^\beta \rho(t)}{dt^\beta} = -\lambda\rho(t), \tag{6}$$

$0 < \alpha, \beta \leq 1$. As we have assumed, Eqs. (5) and (6) describe the process over the entire time domain, but in the short time these equations take the form of Eqs. (2) and (3), respectively. The latter condition is fulfilled if $g_i(t) \rightarrow t$ when $t \rightarrow 0$. We assume that $g_i(t) = t + \xi_i(t)$ with ξ_i satisfying $\xi_i(t) \rightarrow 0$ when $t \rightarrow 0$, $i = 1, 2$. The functions g_1 and g_2 satisfy the general conditions $g_1(0) = g_2(0) = 0$, $g_1(\infty) = g_2(\infty) = \infty$, and $g_1(t), g_2(t), g_1'(t), g_2'(t) > 0$, for $t > 0$.

Solutions to Eqs. (5) and (6). The Green’s function P is a solution to Eq. (5) for the initial condition $P(x, 0|x_0) = \delta(x - x_0)$, where δ is the delta-Dirac function, and for the boundary conditions $P(\pm\infty, t|x_0) = 0$,

$$P(x, t|x_0) = \frac{1}{2\sqrt{D}} f_{-1+\alpha/2, \alpha/2} \left(g_1(t); \frac{|x - x_0|}{\sqrt{D}} \right), \tag{7}$$

where

$$f_{\nu, \mu}(z; a) = \frac{1}{z^{1+\nu}} \sum_{k=0}^{\infty} \frac{1}{k! \Gamma(-\nu - \mu k)} \left(-\frac{a}{z^\mu} \right)^k, \tag{8}$$

$a, \mu > 0$, see [10,21]. The solution to Eq. (6) for the initial condition $\rho(0) = 1$ is

$$\rho(t) = E_\beta(-\lambda g_2^\beta(t)), \tag{9}$$

where $E_\beta(z) = \sum_{n=0}^{\infty} z^n / \Gamma(\beta n + 1)$ is the Mittag-Leffler function, $0 < \beta \leq 1$; see Supplemental Material [22]. We note that for $\beta = 1$ we have $\rho(t) = e^{-\lambda g_2(t)}$. From Eqs. (1), (7), and (9) we get

$$P_\rho(x, t|x_0) = \frac{E_\beta(-\lambda g_2^\beta(t))}{2\sqrt{D}} \times f_{-1+\alpha/2, \alpha/2} \left(g_1(t); \frac{|x - x_0|}{\sqrt{D}} \right). \tag{10}$$

First passage time. The process of molecule vanishing is independent of its position. We assume that the correlation of the functions g_1 and g_2 , if any, is not local and does not depend on the variable x . Let the system consist of two parts, M_1 and M_2 , separated by a point x_M , $M_1 = (-\infty, x_M)$ and $M_2 = (x_M, \infty)$. At the initial moment $t = 0$ the molecule is at the point x_0 located in region M_1 .

The probability that the molecule leaves the region $M_1 = (-\infty, x_M)$ first in the time interval $(t, t + \Delta t)$, where Δt is

assumed to be small, is $F(t; x_0, x_M)\Delta t = \rho(t)[R(t; x_0, x_M) - R(t + \Delta t; x_0, x_M)]$. R is a probability that the molecule did not leave the M_1 region by time t , $R(t; x_0, x_M) = \int_{-\infty}^{x_M} P_{abs}(x, t|x_0)dx$, where $P_{abs}(x, t|x_0)$ is the probability of finding the molecule in the region M_1 in a system with a fully absorbing wall located at x_M [23]. The commonly used boundary condition at the absorbing wall is $P_{abs}(x_M, t|x_0) = 0$. The Green's function for a system with a fully absorbing wall can be found by means of the method of images, which for $x, x_0 < x_M$ gives $P_{abs}(x, t|x_0) = P(x, t|x_0) - P(x, t|2x_M - x_0)$ [24,25]. After calculations we get

$$R(t; x_0, x_M) = 1 - f_{-1, \alpha/2} \left(g_1(t); \frac{x_M - x_0}{\sqrt{D}} \right). \quad (11)$$

Taking the limit of $\Delta t \rightarrow 0$, we obtain

$$F(t; x_0, x_M) = -\rho(t) \frac{dR(t; x_0, x_M)}{dt}. \quad (12)$$

Eqs. (8), (11), and (12) provide

$$F(t; x_0, x_M) = g'_1(t) E_\beta(-\lambda g_2^\beta(t)) \times f_{0, \alpha/2} \left(g_1(t); \frac{x_M - x_0}{\sqrt{D}} \right). \quad (13)$$

For $t \leq 0$ we put $F(t; x_0, x_M) \equiv 0$.

The probability $Q(t; x_0, x_M)$ that the molecule has passed the point x_M until time t is calculated by means of the formula

$$Q(t; x_0, x_M) = \int_0^t F(u; x_0, x_M) du. \quad (14)$$

The function Q will be calculated numerically. Another function describing spreading of vanishing molecules is the mean-square displacement of a particle $\sigma_\rho^2(t) = \int_{-\infty}^{\infty} (x - \bar{x})^2 P(x, t|x_0) dx$, where \bar{x} is a mean value of x . For the Green's function Eq. (10) we get

$$\sigma_\rho^2(t) = \rho(g_2(t)) \frac{2D}{\Gamma(1 + \alpha)} g_1^\alpha(t). \quad (15)$$

When $g_1(t) \equiv t$ and $\rho(t) \equiv 1$, the function $\sigma_{\rho=1}^2(t) \sim t^\alpha$ is frequently used to define a kind of diffusion; subdiffusion is when $0 < \alpha < 1$, normal diffusion is for $\alpha = 1$, and superdiffusion for $\alpha > 1$.

Examples. As examples we consider the case of fully correlated subdiffusion and molecule-vanishing processes. We assume that $g_1(t) \equiv g_2(t) \equiv g(t)$ and that the function ξ_i depends on a periodic function. As g we consider the following functions: $g_0(t) = t$, $g_A(t) = t[1 + a \sin(\tilde{\omega}(t)t + \phi)]$, $g_B(t) = t + a[(\omega t)(\omega t - \sin(2\omega t)) + (1 - \cos(2\omega t))]/2$, and $g_C(t) = t + a(\sin(\omega t + \phi) - \sin(\phi))$, where a and ω are positive parameters. Since $g(t), g'(t) > 0$ we assume $a < 1$ in the cases of A and B , and $a < 1, a\omega < 1$ in the case of C . In the case of A the function $\tilde{\omega}$ depends on t , and for the numerical calculations we put $\tilde{\omega}(t) = t/(\tau_0 + t)$, where τ_0 is a parameter given in a time unit.

Figure 1 shows the plots of the first-passage time distribution Eq. (13). In Fig. 2 there is presented the time evolution of the probability that the molecule will reach the region M_2 by time t , which means that it has passed the point x_M at least once. The function Q has been numerically calculated from Eq. (14). Examples of the functions σ_ρ^2 Eq. (15) and ρ

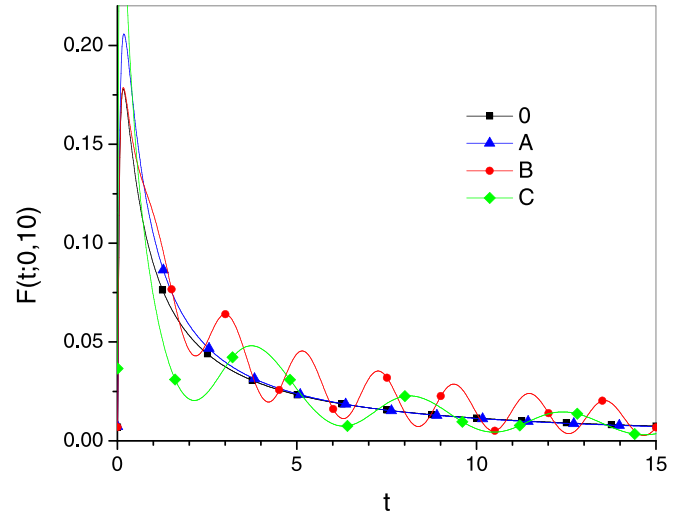


FIG. 1. Time evolution of the first-passage time distribution for g subdiffusion with molecule-vanishing processes controlled by the functions $g_1(t) \equiv g_2(t) \equiv g(t)$, where $g = g_0$ (symbol O in the legend), $g = g_A$ (symbol A), $g = g_B$ (symbol B), and $g = g_C$ (symbol C). The parameters are $\alpha = 0.6$, $\beta = 0.5$, $\lambda = 0.01$, $\omega = 1.5$, $\phi = 0$, $a = 0.4$, $x_0 = 0$, and $x_M = 10$, $\tilde{\omega} = 1.5t/(1+t)$ for g_B , $\tau_0 = 1$; all quantities are given in arbitrarily chosen units.

Eq. (9) are shown in Figs. 3 and 4. All plots are made for the parameters given in the Fig. 1 caption.

The function g_0 does not change the timescale. In the cases A , B , and C , the timescale is changed and periodic functions are involved in the function g . In the case of A , g_A contains $\sin(\tilde{\omega}(t))$, and the period of this function decreases with time and goes to zero when $t \rightarrow \infty$. For the parameters given in the caption of Fig. 1, g_A does not generate noticeable oscillating effects, see Figs. 1–4. For processes B and C oscillation effects are observed in the figures. In process B , the amplitude of

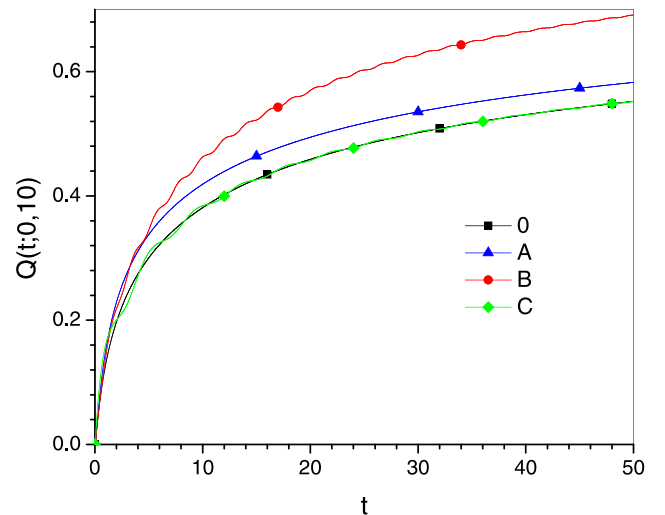


FIG. 2. Time evolution of the probability Q that a molecule has passed the point x_M until time t , Eq. (14). The oscillation effect for cases B and C is weaker here than for the first-passage time distributions presented in Fig. 1.

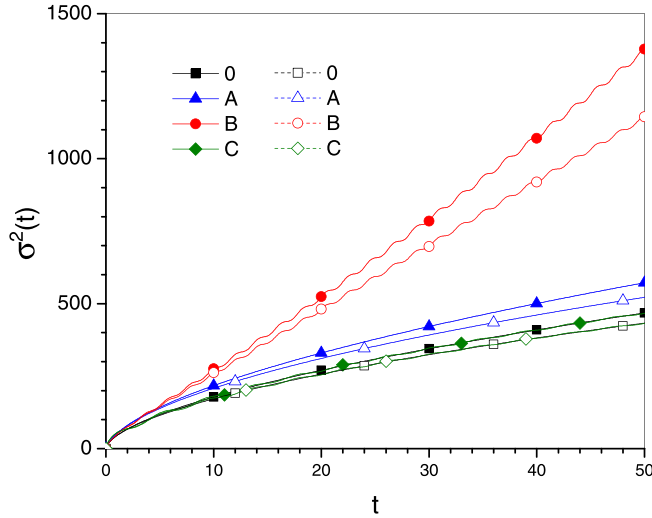


FIG. 3. The plots of the time evolution of mean-square displacement σ_ρ^2 , Eq. (15), for $\rho(t) \equiv 1$ (filled symbols) and for $\rho(t)$ varying in time (open symbols) are compared. Lines with filled symbols describe diffusion when particle vanishing is “turned off,” and lines with open symbols correspond to diffusion of vanishing particles; in both cases the parameters α and D are the same. We consider the case of a full correlation of diffusion and molecule vanishing; therefore the oscillations of both functions B are consistent.

timescale oscillations increases with time and is proportional to t . The change in the timescale is relatively large; the rate of change is described by the function $g'_B(t) = 1 + at \sin^2(\omega t)$, which results in $g_B \sim t^{2\alpha}$ in the long time limit. Thus, assuming $\alpha > 0.5$, the oscillation effect is superimposed here on the superdiffusion effect. In the case of C , the amplitudes of oscillations are constant, and the oscillation effect decreases with time. Here we have $\sigma_\rho^2(t) \sim t^\alpha$ when $t \rightarrow \infty$, and there is the “ordinary” subdiffusion effect in the long time limit.

Final remarks. Subdiffusion and a molecule-vanishing process are described by Eqs. (5) and (6). In general, by involving g -Caputo fractional time derivatives in a model instead of “ordinary” Caputo derivatives, we can describe changes of the course of the processes caused by external factors (e.g., temperature change causing the temporal evolution of parameters) and by the influence of different processes on each other. It has already been shown that the g -subdiffusion equation describes subdiffusion with a change of parameters α and D [8] and a transition from “ordinary” subdiffusion to slow subdiffusion (ultraslow diffusion) [9]. In this paper we use the equations with g -Caputo fractional derivatives to model the subdiffusion and molecule-vanishing processes, which can be related to each other.

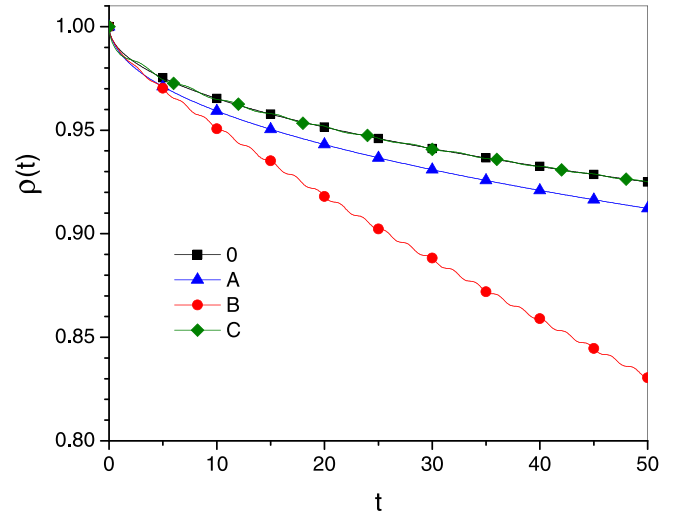


FIG. 4. Time evolution of the probability $\rho(t)$, see Eq. (9), that a particle still exists. Unlike in the other functions, the oscillation amplitude of the g_B function increases with time, and thus the probability ρ decreases fastest for function B .

We have assumed the molecule vanishing has a constant rate λ and does not depend on the molecule position. This process can be interpreted as a subdiffusion with $A \rightarrow \emptyset$ reaction. In [26] it was shown that the form of a subdiffusion-reaction equation depends on whether a diffusing molecule A can decay with a probability independent of its position or whether it must meet a B molecule, with a certain probability, for a reaction to occur. Subdiffusion-reaction equations are qualitatively different in both cases [26,27]. If the function ρ also depended on the variable x , then the process could be described by the following g -subdiffusion-reaction equation with a reaction $nA + mB \rightarrow \emptyset$ (n and m are positive parameters):

$$\frac{{}^C \partial_{g_1}^\alpha P_\rho(x, t|x_0)}{\partial t^\alpha} = D \frac{\partial^2 P_\rho(x, t|x_0)}{\partial x^2} - \Xi(P_\rho, C_A(x, t), C_B(x, t)), \quad (16)$$

where P_ρ describes the subdiffusion of the A molecule, Ξ is a reaction term which can be controlled by both functions g_1 and g_2 , and C_A , C_B denote concentrations of A and B molecules, respectively. We note that if $n \neq 1$, a probability of the reaction depends on the concentration C_A . The reaction term may be nonlinear, and the analysis of a general g -subdiffusion-reaction equation will be presented elsewhere. G subdiffusion with an $A \rightarrow \emptyset$ reaction can be described by the subdiffusion equation and the molecule survival equation using the model presented in this paper.

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