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A + B → 0 REACTION UNDER NON-MARKOVIAN SUBDIFFUSIVE KINETICS: EQUATIONS AND STATIONARY SOLUTIONS*

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We consider an A + B → 0 reaction in a flat subdiffusive medium in contact with two well mixed reservoirs of particles of both types on the sides. We show that the behaviour of the stationary concentration and reaction intensity profiles in subdiffusion differs strikingly from that observed in simple diffusion. The most marked differences correspond to accumulation peaks and depletion zones in the concentration profile. The height of these peaks as well as the height of the reaction zones exhibit a non-monotonic behaviour with respect to the reactant's concentrations at the boundaries. These characteristics are due to an effectively nonlinear transport under reaction which emerges from the non-Markovian property of the subdiffusion process.

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1. Introduction

Many phenomena in systems out of equilibrium corresponding to spatial dispersion and interaction of individuals can be modelled by reaction-diffusion equations. Examples cover a wide range of disciplines, such as trapping and annihilation of excitons and electron-hole recombination of charge carriers in physics, predator-prey-relationships in ecology or reaction of molecules in chemistry. In the case of normal diffusion, the corresponding reaction-diffusion equations are obtained by adding a classical kinetic rate term to the right hand side of a diffusion equation:

$$\dot{C}_j(\boldsymbol{r},t) = D_j \Delta C_j(\boldsymbol{r},t) \pm \kappa_j C_1^{n_1}(\boldsymbol{r},t) C_2^{n_2}(\boldsymbol{r},t) \ldots C_N^{n_N}(\boldsymbol{r},t)\,, \tag{1}$$

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where D_j is the diffusivity of component j, n_j are the stoichiometry factors and the κ_j denotes the reaction rate coefficient of of each of the reacting components. These equations hold whenever there is a strong scale separation between the characteristic size of the inhomogeneities of the system and the typical reaction scale. The stationary concentrations of particles in the system constantly fed by reactants is given by the same equations (under corresponding boundary conditions) with the left hand sides put to zero.

However, in many cases the diffusion process is anomalous [2], as recently found *e.g.* for motion in bacterial cytoplasm [3] or in gels [4]. Whereas in normal diffusion the mean square displacement is proportional to the time t, anomalous diffusion exhibits a mean square displacement that goes as $\left\langle r^2 \right\rangle \propto t^\alpha$, where $\alpha > 1$ corresponds to the superdiffusive case and $0 < \alpha < 1$ to the subdiffusive case. In recent years, subdiffusion has been modelled successfully within the framework of continuous time random walks (CTRW) with a heavy-tailed waiting time density function of the form $\psi(t) \propto t^{-(1+\alpha)}$, which yields a fractional diffusion equation instead of a Fickian one [5].

Although reaction-diffusion systems with anomalous transport have been extensively studied, a full theory of reaction-subdiffusion is still missing. Fractional reaction-diffusion equations of different types have been proposed on a phenomenological basis [6–10]. Analogous to reaction-diffusion, a fractional derivative was either posited in front of the spatial Laplacian or in front of both the Laplacian and the reaction term of Eq. (1) [8]. The latter variant holds for situations where the reaction on small scales is subdiffusion-controlled so that particles are able to react only instantaneously at the beginning (or end, respectively) of a waiting time period, as discussed in [9, 10] using a microscopic approach. However, it is not quite clear whether the former proposal corresponds to any physical reaction-diffusion setting.

In this paper we adhere to the situation where the reaction on small scales is diffusion controlled, corresponding *e.g.* to reaction-diffusion in porous media. From a mesoscopic point of view, non-Markovian transport is here induced by trapping of the particles in cavities on all length scales. The stagnant particles can still react and the reaction locally follows classical kinetics. Using a CTRW-approach, Ref. [11] have shown for linear reaction kinetics that reaction-subdiffusion equations in this case do not follow by simply changing the normal diffusion operator for a fractional one, since the reaction affects the transport term. Ref. [12] proposed a rather general approach to reaction-subdiffusion; its application to Turing instability [13, 14] as well as to front propagation in systems with Fisher-type kinetics [15] showed only quantitative differences with reaction-diffusion.

In what follows we discuss the simplest problem in which nontrivial spatial structures appear, namely the irreversible A + B → 0 reaction, and show that stationary structures formed under subdiffusion differ strikingly from those in reaction-diffusion, or are even absent under certain conditions where their reaction-diffusion analogues exist. The A + B → 0 reaction equation corresponds to a bimolecular irreversible reaction between the particles of two different sorts, where the product of the reaction is either inert (like in the chemical examples of Refs. [16–18]) or immediately leaves the system (as it is the case in the recombination reactions when this product is a photon). The above situation can be realized in an experimental setting proposed in [4] or in a porous medium fed by a well mixed reservoir on either side. Here we assume the particle concentrations to depend only on the x-coordinate.

2. Reaction-subdiffusion equations

We first derive the reaction-subdiffusion-equations following the scheme put forward in [11]. In a CTRW, a particle arriving at site i stays there for a sojourn time t drawn at random according to a probability density function (pdf) $\psi(t)$. Leaving a site it makes a random step in either direction. For an unbiased random walk, the probabilities of going to the left and to the right are equal to 1/2. The generalized reaction-diffusion equations are based on two balance conditions; a local one corresponding to the balance between probability loss and gain at one site, and another according to transitions between two neighbouring sites, *i.e.* particle conservation during a jump. The balance equation for A-particles at each site reads:

$$\begin{aligned} \dot{A}_i(t) &= j_i^+(t) - j_i^-(t) + R_i\{A,B\} && (2)\\ &= \tfrac{1}{2} j_{i-1}^-(t) + \tfrac{1}{2} j_{i+1}^-(t) - j_i^-(t) + R_i\{A,B\}\,. && (3) \end{aligned}$$

where $j_i^-(t)$ is the loss flux of A-particles at site i, *i.e.* the probability for an A-particle to leave i per unit time, $j_i^+(t)$ is the gain flux at the site, and $R_i\{A,B\} = -\kappa A_i B_i$ is the reaction term, describing loss of particles due to reaction. Since in our case the equations for A- and B-particles are symmetric, we concentrate on the equations for A_i. A reaction-subdiffusion equation is a combination of the continuity equation (3) and the equation for the loss fluxes $j^-(t)$ following from the waiting time distribution $\psi(t)$ and the survival probability $P(t, t_0)$. The loss flux for site i at time t is connected to the gain flux at the site at all previous times and to the survival probability. That is to say, the particles leaving site i at time t were either there from the very beginning and survived until t, or arrived there at some time $t' < t$ and survived until t. The probability density to make a step at t having arrived at t' is given by the waiting time pdf $\psi(t-t')$. We have then

$$j_i^-(t) = \psi(t)P_i(t,0)A_i(0) + \int\limits_0^t \psi(t-t')P_i(t,t')j_i^+(t')dt' \,. \tag{4}$$

Rewritten by means of Eq. (2), this yields

$$\begin{aligned} j_i^-(t) &= \psi(t)P_i(t,0)A_i(0) + \int\limits_0^t \psi(t-t')P_i(t,t') \\ &\times \left[\dot{A}_i(t') + j_i^-(t') + \kappa A_i(t')B_i(t')\right] dt' \,. \end{aligned} \tag{5}$$

The survival probability of A at site i is given by the classical kinetic rate equation

$$\frac{d}{dt}P_i(t) = -\kappa B_i(t)P_i(t) \tag{6}$$

and depends on $B_i(t)$:

$$P_i(t,t_0) = \exp\left(-\kappa \int\limits_{t_0}^t B_i(t')dt'\right) \,. \tag{7}$$

The equations (3), (5) and (7) and the corresponding equations for B represent the full system of equations for the time dependent concentrations. At this stage we also can assume the relative change in concentration to be small compared to the lattice spacing a, and change to a continuous coordinate $x = ai$. Our system of equations finally reads

$$\begin{aligned} \dot{A}(x,t) &= \frac{a^2}{2}\Delta j^-(x,t) - \kappa A(x,t)B(x,t) \,, \\ j^-(x,t) &= \psi(t)P(x,t,0)A_(x,0) + \int\limits_0^t \psi(t-t')P(x,t,t') \\ &\times \left[\dot{A}(x,t') + j^-(x,t') + \kappa A(x,t')B(x,t')\right] dt' \\ P(x,t,t') &= \exp\left(-\kappa \int\limits_{t'}^t B(x,t'')dt''\right) \,, \end{aligned} \tag{8}$$

together with the corresponding system for the B-concentrations.

3. Stationary equations for the Dirichlet boundary value problem

In our previous discussion we considered an initial condition problem, where the particles were introduced into the system at time $t = 0$ and we followed the subsequent evolution of their concentrations. In the case of reaction-diffusion, a steady state can be obtained by fixing the particle concentrations at the boundaries of the system, *e.g.* by external sources. The corresponding equations remain the same as in the initial condition problem, with the time derivatives at the left hand side put to zero. For reaction-subdiffusion, it is not quite evident that this assumption still holds.

Let us assume that a steady state characterized by constant concentrations $A(x)$ and $B(x)$ exists. Let us furthermore assume that no sources of A- and B-particles are present in the interior of the system and that the steady state is maintained through the sources at the boundaries. In the present boundary value problem, new particles are permanently fed into the system. Accordingly, the equations for the initial value problem have to be modified. The particles now are labelled according to the time t_0 they were introduced, so that $A(x,t|t_0)dt_0$ is the concentration at x at time t of A-particles that were introduced between t_0 and $t + dt_0$. The partial concentration $A(x,t_0|t_0)$ of newly introduced particles is zero everywhere in the interior of the system, and the overall concentration is given by the integral

$$A(x) = \int\limits_{-\infty}^{t} A(x,t|t_0)dt_0 \,. \tag{9}$$

Due to translation invariance in time, in a steady state $A(x,t|t_0)$ can only be a function of the elapsed time $t_e = t - t_0$, so that $A(x,t|t_0) = A(x,t-t_0)$ and $A(x) = \int_0^\infty A(x,t_e)dt_e$. We have

$$\dot{A}(x,t_e) = \frac{a^2}{2}\Delta j^-(x,t_e) - \kappa A(x,t_e)B(x)\,. \tag{10}$$

for all $A(x,t|t_0)$. With constant $A(x)$ and $B(x)$, also the survival probability becomes a function of the difference of its time arguments,

$$P(x,t,t') = \exp\left[-\kappa B(x)(t-t')\right]\,, \tag{11}$$

and the integral in the equation for the flux takes the form of a convolution:

$$\begin{aligned} j^-(x,t|t_0) \;&=\; \psi(t)P(x,t-t_0)A(x,t_0|t_0) + \int\limits_{t_0}^{t} \psi(t-t')P(x,t-t') \\ &\times \left[\dot{A}(x,t'|t_0) + j^-(x,t'|t_0) + \kappa A(x,t'|t_0)B(x)\right] dt' \end{aligned} \tag{12}$$

with $j^-(x,t'|t_0)$ being the loss fluxes for those A-particles which were introduced into the system at time t_0. Passing to the Laplace domain with respect to t_e and denoting $\tilde{A}(x,u) = \int_{t_0}^{\infty} A(x,t|t_0)\exp[-u(t-t_0)]dt$, we obtain

$$\tilde{j}^-(x,u) = \frac{[u+\kappa B(x)]\tilde{\psi}(u+\kappa B(x))}{1-\tilde{\psi}(u+\kappa B(x))}\tilde{A}(x,u)\,, \tag{13}$$

where the Laplace transform of the product $\Psi(t,x) = \psi(t)\exp[-\kappa B(x)t]$ is given by the shift theorem with $\tilde{\Psi}(u,x) = \tilde{\psi}(u+\kappa B(x))$. Inserting this into the equations for the partial concentrations (10), we get in the Laplace domain:

$$\begin{aligned} &u\tilde{A}(x,u) - A(x,t_0|t_0) \\ &= \frac{a^2}{2}\Delta\frac{[u+\kappa B(x)]\tilde{\psi}(u+\kappa B(x))}{1-\tilde{\psi}(u+\kappa B(x))}\tilde{A}(x,u) - \kappa\tilde{A}(x,u)B(x)\,. \end{aligned} \tag{14}$$

Since there are only sources at the boundaries, $A(x,t_0|t_0)$ vanishes in the interior of the system. Taking the limit $u \to 0$ and putting $A(x) = \tilde{A}(x,0)$, the stationary concentration of A-particles in the interior of the system yields

$$\frac{a^2}{2}\Delta\frac{\kappa B(x)\tilde{\psi}(\kappa B(x))}{1-\tilde{\psi}(\kappa B(x))}A(x) - \kappa A(x)B(x) = 0\,, \tag{15}$$

together with the appropriate boundary conditions. For the case of Markovian transport, *i.e.* for waiting time pdfs of the form $\psi(t) = \frac{1}{\tau}\exp[-\frac{t}{\tau}]$, we get $\tilde{\psi}(u) = 1/(1+u\tau)$ and correspondingly:

$$\frac{a^2}{2\tau}\Delta A(x) - \kappa A(x)B(x) = 0\,, \tag{16}$$

the stationary reaction-diffusion equation for A with the diffusion coefficient $D = a^2/(2\tau)$. In the non-Markovian case, the waiting time pdf in the Laplace domain can be approximated by $\tilde{\psi}(u) \simeq 1-(\tau u)^\alpha \Gamma(1-\alpha)$ for small u, provided that the cut-off parameter $\kappa B(x)\tau \ll 1$. Eq. (15) becomes

$$\frac{a^2}{2}\frac{1}{\tau^\alpha \Gamma(1-\alpha)}\Delta B(x)^{1-\alpha}A(x) - \kappa^\alpha A(x)B(x) = 0\,, \tag{17}$$

where $D_\alpha = a^2/2\tau^\alpha\Gamma(1-\alpha)$ is the generalized diffusion coefficient. The system of equations with additional temporal operator acting on the Laplacian in the case of an initial-condition problem turns to a system of reaction-diffusion equations with a *nonlinear* diffusion term for a stationary state with given boundary conditions. For $\alpha = 1$, Eq. (17) reduces to the Markovian stationary reaction-diffusion equation. Eq. (15) together with the corresponding equation for B constitutes the full system of steady state equations.

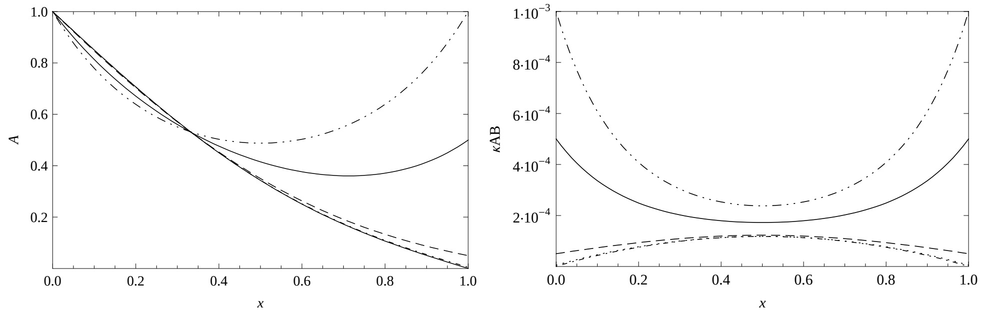
4. Stationary particle concentrations and stationary reaction intensities

We consider a system on an interval $(0,1)$ with given concentrations of reactants on the boundaries, *e.g.* a subdiffusive gel reactor in contact with two well mixed reservoirs on both sides. These reservoirs contain reacting mixtures of both species at different concentrations. To simplify matters, we examine a symmetric situation with $B(x) = A(1-x)$ and put $A(0) = B(1) = 1$. In what follows, we denote the left boundary of the medium as the major source of A, whereas the left boundary will be referred to as to the minor source (except for $A(1) = B(0) = 1$). To solve Eq. (17), we first change to new variables $A^*(x) = B^{1-\alpha}(x)A(x)$ and $B^*(x) = A^{1-\alpha}(x)B(x)$. They are considered to be formally dependent on the new time variable ξ. The new equation

$$\frac{\delta}{\delta\xi}A^*(x,\xi) = D_\alpha \Delta A^*(x,\xi) - \kappa^\alpha (A^*(x,\xi)B^*(x,\xi))^{1/(2-\alpha)} \tag{18}$$

with $B^*(1-x,\xi) = A^*(x,\xi)$ was integrated numerically from the initial condition $A(x,0) = (A(1)-1)x + 1$ using the Crank–Nicholson algorithm until the solution reaches stationarity.

The concentration profiles for subdiffusion ($\alpha < 1$) are compared to those of normal diffusion ($\alpha = 1$), Figs. 1(a) and 2. The $A(0) = B(1)$ concentration at the major source is fixed to be $A(0) = 1$, the other concentration varies from $B(0) = A(1) = 1$, corresponding to the symmetric case, when the CTRW-reactor separates two stoichiometric reacting mixtures, to $B(0) = A(1) = 10^{-4}$.



(a) (b)

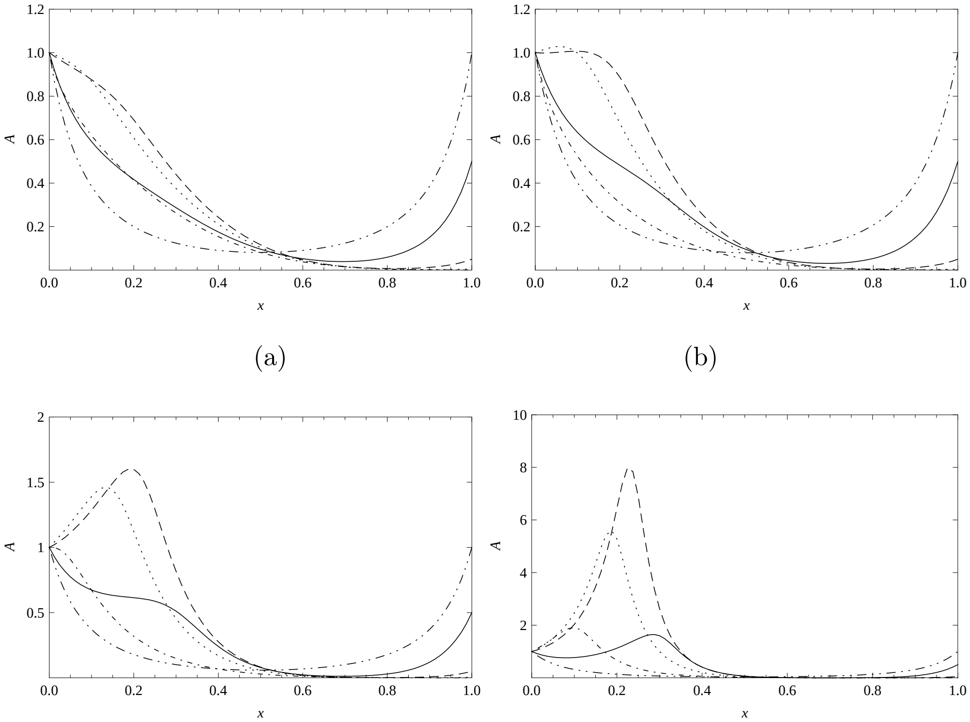
Fig. 1. (a) Stationary particle concentration $A(x)$ and (b) stationary reaction intensity $R(x) = \kappa A(x)B(x)$ for $\alpha = 1$; different boundary values: $A(1) = 1$ (dash-dot-dotted), 0.5 (solid line), 5×10^{-2} (dashed), 5×10^{-3} (dotted), and 1×10^{-4} (dash-dotted); $\kappa = 0.001$, $D_\alpha = 1/2$.

In the symmetric case, reaction-diffusion and reaction-subdiffusion concentration profiles behave similarly with maximal concentrations in the regions close to the boundaries where the system is fed by reactants. Under asymmetric conditions the behaviours of the concentrations differ considerably. The most marked difference corresponds to accumulation of A-particles in the interior of the system close to the major source in the subdiffusive case, the counterpart on the other side of the system is a depletion zone corresponding to the symmetric accumulation zone for B. The height of the accumulation peak exhibits a *nonmonotonic* dependence on the strength of the minor source. The reduction of the minor source strength $A(1) = B(0)$ leads first to its growth, and then to its outwards motion accompanied by decay. For equally strong $A(0)$ and $B(0) = A(1)$, the particles react in the vicinity of the boundaries, where their concentratioin is high, before they could cover a considerable distance. For smaller $B(0)$, some of the A-particles can travel further into the medium without reaction. In the case of subdiffusive transport, the effective mobility of the particles decays in the course of time, the number of performed steps goes as $t^{\alpha-1}$. The particles accumulate inside the system which leads to the formation of a peak. For very weak minor sources the peak moves closer to the boundary and eventually dissappears. Low α amplify the effects of subdiffusive transport on the concentration profiles. The smaller is the α, the more pronounced get the peak and the depletion zone. Fig. 2 shows the profiles for different boundary conditions for $\alpha = 0.9$, 0.8, 0.7 and 0.6.

Whereas in reaction-diffusion $A(x)$ reaches a limiting form for $A(1) \to 0$, no stationary concentration profile exists in the subdiffusive case. The effective diffusion coefficient vanishes at the corresponding side of the system preventing the inflow of reactants from their major sources into the system. To explain this long time behaviour, one can also consider the time evolution of concentrations by discussing the inverse Laplace transform of Eq. (15). We put $\tilde{\psi}(u) \simeq 1 - (\tau u)^\alpha \Gamma(1-\alpha)$:

$$\frac{\partial A(x,t)}{\partial t} = \frac{a^2}{2} \Delta \int_0^t \Phi(t-t') A(x,t') dt' - \kappa B(x) A(x,t) \,, \tag{19}$$

where the Laplace transform of the integral kernel is $\tilde{\Phi}(u) = [\tau^\alpha \Gamma(1-\alpha)]^{-1} (u + \kappa B(x))^{1-\alpha}$. This yields:



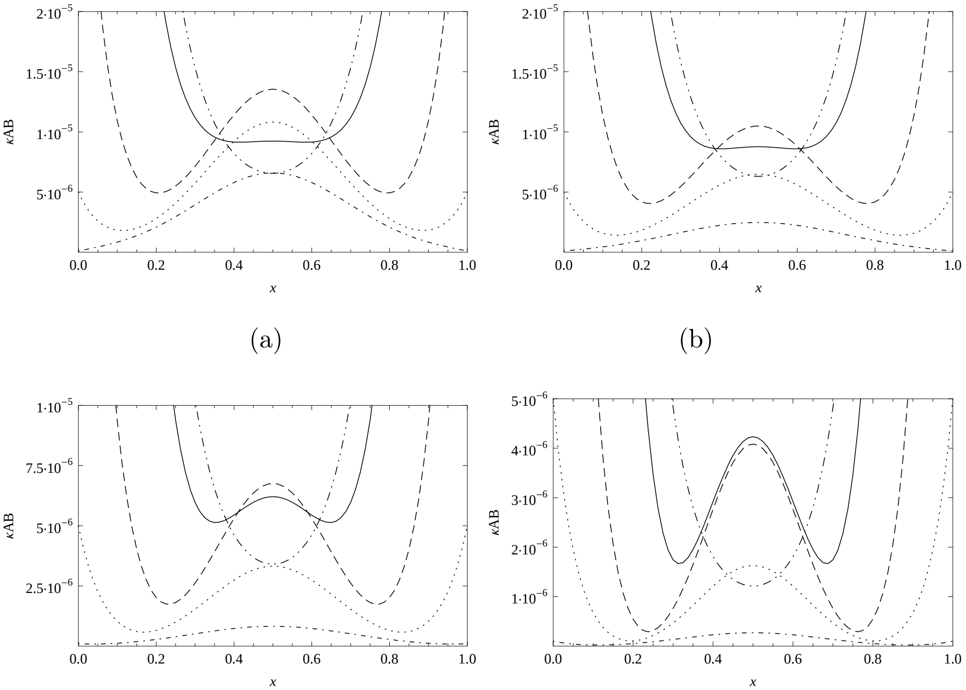
(a) (b)

(c) (d)

Fig. 2. Stationary particle concentrations $A(x)$ for a) $\alpha = 0.9$, (b) $\alpha = 0.8$, (c) $\alpha = 0.7$, (d) $\alpha = 0.6$; $A(1)$ see Fig. 1; $\kappa = 0{,}001$, $D_\alpha = 1/2\Gamma(1-\alpha)$. The smaller the α, the more pronounced get the peak and the depletion zone.

$$\begin{aligned}\frac{\partial A(x,t)}{\partial t} &= \frac{a^2}{2\tau^\alpha \Gamma(1-\alpha)} \Delta \Bigg[\frac{d}{dt} \int_0^t \frac{e^{-\kappa B(x)(t-t')}}{(t-t')^{1-\alpha}} A(x,t')dt' \\ &\quad + \kappa B(x) \int_0^t \frac{e^{-\kappa B(x)(t-t')}}{(t-t')^{1-\alpha}} A(x,t')dt' \Bigg] \\ &\quad - \kappa B(x) A(x,t) \,. \end{aligned} \tag{20}$$

As in the case of linear reaction dynamics [11], the reaction affects the transport term, which however becomes nonlinear due to its dependence on $B(x)$. The factor $\exp[-\kappa B(x)t]$ imposes a cut-off upon the long tail of the waiting time pdf $\psi(t)$. The effective diffusivity in a stationary state remains constant in time but varies in space, depending on $B(x)$. In the case of $B(x) = 0$, the effective diffusivity close to the boundary where the B-concentration is kept at zero, decreases permanently as $t^{\alpha-1}$. This permanent change in the effective diffusivity is mirrored by the fact that the stationary concentration profile does not exist.



(a) (b)

(c) (d)

Fig. 3. Stationary reaction intensity $\kappa A(x)B(x)$ for (a) $\alpha = 0.9$, (b) $\alpha = 0.8$, (c) $\alpha = 0.7$, (d) $\alpha = 0.6$; $A(1)$ see Fig. 1; $\kappa = 0,001$, $D_\alpha = 1/2\Gamma(1-\alpha)$. The smaller the α, the smaller becomes the reaction intensity. Smaller α require a smaller $A(1)$ for a reaction zone to evolve between both reacting mixtures (note the solid curve for $A(1) = 0.5$).

The reaction intensity profiles under subdiffusion show striking differences in comparison to the normal situation, Figs. 1(b) and 3. For decreasing $A(1)$ the reaction zone starts to form in the middle of the system. In the normal case, for small $A(1)$ the form of the reaction zone depends only marginally on $A(1)$, and there exists a clear limiting form for $A(1) = 0$. In reaction-subdiffusion, the dependence of the height of the reaction zone on the strength of the minor source is nonmonotonic. When lowering $A(1)$, the maximum of the reaction intensity first gets higher and then starts to lower; the distribution as a whole broadens. There is no limiting form for $A(1) \to 0$. A stationary reaction zone can only exist if a permanent current of reacting particles from the corresponding sides is maintained. The reaction zone is the higher and the narrower, the larger is the particles' inflow into the reaction area. In the subdiffusive case, this inflow depends on the effective diffusion coefficient and hence on the concentration of the reacting counterpart. For $A(1) = B(0) = 0$ the effective diffusion coefficient vanishes and no stationary front exists, the reaction zone blurs and fades out. Fig. (refbildRa shows stationary reaction intensity profiles in dependence of the concentrations at the boundaries for $\alpha = 0.9$, 0.8, 0.7 and 0.6.

5. Conclusions

We considered a subdiffusive medium fed by reacting mixtures on its both sides. Stationary particle concentration profiles and reaction intensity profiles formed under subdiffusion differ vastly from those in reaction-diffusion. Due to the non-Markovian character of subdiffusive motion, the corresponding stationary equations possess a nonlinear coupling not only in the reaction- but also in the transport term. Accumulation and depletion zones emerge close to the boundaries, and a non-monotonic behavior both of the reaction intensity and of the particle concentration with respect to the strength of the minor source can be observed in the anomalous case. If one of the A- or B-particle concentrations is zero at the boundaries, no stationary solution exists.

REFERENCES

[1] D. Froemberg, I.M. Sokolov, *Phys. Rev. Lett.* **100**, 108304 (2008).

[2] I.M. Sokolov, J. Klafter, A. Blumen, *Phys. Today* **55** (11), 48 (2002).

[3] I. Golding, E.C. Cox, *Phys. Rev. Lett.* **96**, 098102 (2006).

[4] T. Kosztołowicz, K. Dworecki, St. Mrówczyński, *Phys. Rev. Lett.* **94**, 170602 (2005).

[5] R. Metzler, J. Klafter, *Phys. Rep.* **339**, 1 (2000).

[6] T. Kosztołowicz, K.D. Lewandowska, *Acta Phys. Pol. B* **37**, 1571 (2006).

[7] S.B. Yuste, L. Acedo, K. Lindenberg, *Phys. Rev.* **E69**, 036126 (2004).

[8] B.J. Henry, S.L. Wearne, *Physica A* **276**, 448 (2000); *SIAM J. Appl. Math.* **62**, 870 (2002); B.I. Henry, T.A.M. Langlands, S.L. Wearne, *Phys. Rev.* **E72** (2), 026101 (2005).

[9] K. Seki, M. Wojcik, M. Tachiya, *J. Chem. Phys.* **119**, 2165 (2003).

[10] J.Y. Sung, R.J. Silbey, *Phys. Rev. Lett.* **91**, 160601 (2003).

[11] I.M. Sokolov, M.G.W. Schmidt, F. Sagués, *Phys. Rev.* **E73**, 031102 (2006).

[12] M.O. Vlad, J. Ross, *Phys. Rev.* **E66**, 061908 (2002).

[13] A. Yadav, W. Horsthemke, *Phys. Rev.* **E74**, 066118 (2006).

[14] Y. Nec, A.A. Nepomnyashchy, *J. Phys.* **A40**, 14687 (2007).

[15] A. Yadav, S. Fedotov, V. Mendez, W. Horsthemke, *Phys. Lett.* **A371**, 374 (2007).

[16] A. Yen, Y.E.L. Koo, R. Kopelman, *Phys. Rev.* **E54**, 2447 (1996).

[17] S.H. Park, S. Parus, R. Kopelman, H. Taitelbaum, *Phys. Rev.* **E64**, 055102(R) (2001).

[18] E. Monson, R. Kopelman *Phys. Rev.* **E69**, 021103 (2004).

[19] R. Balescu, *Statistical Dynamics — Matter out of Equilibrium*, Imperial College Press, London (1997).