Diffusion-Limited Coalescence and Annihilation in Random Media

Catalin Mandache and Daniel ben-Avraham

Physics Department, and Clarkson Institute for Statistical Physics (CISP),
Clarkson University, Potsdam, NY 13699-5820

We study the kinetics of diffusion-limited coalescence, $A + A \rightarrow A$, and annihilation, $A + A \rightarrow 0$, in random media consisting of disconnected domains of reaction. Examples include excitons fusion and annihilation in porous matrices and along polymer chains. We begin with an exact analysis of $A + A \rightarrow A$ in a finite segment. This result is applied to coalescence in a random distribution of segment lengths, and the implications for coalescence and annihilation in percolation cluster and other confined geometries are then derived by means of scaling techniques.

I. INTRODUCTION

Diffusion-limited reactions are far less understood than their reaction-limited counterpart. Simple reaction models such as diffusion-limited coalescence, $A + A \rightarrow A$, and annihilation, $A + A \rightarrow 0$, have attracted much recent attention [1–14], since their kinetics in one-dimensional space may be analyzed exactly, and they shed light on other less tractable reaction schemes. Moreover, coalescence and annihilation serve as models of exciton dynamics in several real systems. Examples include photoexcited solitons and polarons in MX chain compounds, such as $\text{[Pt(en)2]}\,[\text{Pt(en)2Cl}_2]\,[\text{BF}_4]_4$ ((en) denotes ethylenediamine) [15–17], and fusion of photogenerated excitons in tetramethylammonium manganese trichloride (TMMC) chains [17]. Kroon and Sprik [17] review many other systems which are aptly modeled by diffusion-limited coalescence or annihilation in one dimension.

Most theoretical studies to date assume an infinite medium, however, rich kinetic behavior arises from the finite size of the substrate in question. For times that exceed the typical diffusion time needed to span the size of the sample, finite-size effects set in and dramatically affect the kinetics. We argue that when the system consists of a collection of reaction-domains of varying sizes, this might lead to a stretched exponential decay in the concentration of particles, as opposed to the usual power-law decay. For example, disorder in a system of polymers might effectively fracture the medium into separate reaction zones. Indeed, stretched exponential decay of photogenerated excitons in MX compound chains [15–16] and in TMMC chains [17] is a well documented fact.

The problem generalizes to systems of higher dimensionality. Triplet exciton annihilation in naphtalene-$d_8$ embedded in porous matrices of napthalene-$d_8$ has been studied by Kopelman [18]. Here the geometry of the reaction zones and their characteristic sizes might be well described by percolation theory. Kopelman [18] has studied exciton dynamics in several systems, spanning an impressive range of different geometries.

In this paper we deal with diffusion-limited coalescence, or annihilation, in random media — media consisting of disconnected reaction domains with a given distribution of sizes. We begin with an exact derivation of the kinetics of coalescence, $A + A \rightarrow A$, in a finite segment of length $L$, in section II. At early times, $t \ll 1/Dc_0^2$ ($D$ is the diffusion coefficient of the particles, and $c_0$ is their initial concentration), few reactions take place and the concentration is nearly constant; at the intermediate time regime of $1/Dc_0^2 \ll t \ll L^2/D$ the concentration decays in power-law fashion, $c \sim 1/\sqrt{Dt}$; and in late times, $L^2/D \ll t$, the decay is exponential, $c \sim \exp[-(2\pi^2 D/L^2)t]$. In section III, we use these results to derive the kinetics of diffusion-limited coalescence of particles confined to a set of segments characterized by the distribution of their lengths, $\psi(L)$, as might be the case for exciton dynamics in polymer compounds. For a completely random distribution, $\psi(L) = \rho^{-1}\exp(-\rho L)$, we find that the long time asymptotic decay is stretched-exponential, $c \sim \exp(-t^{1/3})$, similar to the long time behavior in the famous problem of trapping [19]. The earlier time regime is dominated by a somewhat faster stretched-exponential decay, $c \sim \exp(-t^{1/2})$. Coalescence and annihilation in percolation clusters is addressed in section IV. Here the situation is more complicated, for the reaction domains are fractal and diffusion is then anomalous [20]. Our analysis consists of scaling arguments, motivated by the more firmly grounded results in one dimension. We conclude with a summary and discussion, in section V.

II. COALESCENCE IN A SEGMENT

Consider the diffusion-limited coalescence process, $A + A \rightarrow A$, where the particles diffuse with diffusion coefficient $D$ and merge instantaneously upon encounter, taking place in a segment of length $L$. An exact analysis is possible through the method of Empty Intervals, known also as the method of Inter-Particle Distribution Functions (IPDF) [13]. The key parameter is $E(x,t)$ — the probability that an arbitrary interval of length $x$ is empty (contains no particles), at time $t$. $E(x,t)$ satisfies the diffusion equation [13]:

$$\frac{\partial}{\partial t}E(x,t) = 2D\frac{\partial^2}{\partial x^2}E(x,t) \quad . \quad (1)$$

\[\text{arXiv:cond-mat/9911041v1 [cond-mat.stat-mech]} \quad 3 \text{ Nov} 1999.\]
The coalescence reaction imposes the boundary condition

\[ E(0, t) = 1 , \]

and, because \( E \) is a probability, also

\[ E(x, t) \geq 0 . \]

Finally, if there are any particles in the segment then

\[ E(L, t) = 0 . \]

From \( E(x, t) \) one can compute various useful quantities. For example, the particle concentration is

\[ c(t) = -\frac{\partial E(x, t)}{\partial x} \bigg|_{x = 0} , \]

and the conditional probability that the next nearest particle to a given particle is at distance \( x \) (the IPDF), is

\[ p(x, t) = c(t)^{-1} \frac{\partial^2}{\partial x^2} E(x, t) . \]

The steady state solution of eq. (1) (i.e., \( \partial E/\partial t = 0 \)), with the boundary conditions (2), (3), and (4), is

\[ E_s(x) = 1 - \frac{x}{L} . \]

Clearly, if there is initially at least one particle present, a single particle would remain at the end of the process, unable to undergo any further reactions. This is confirmed by eq. (7), and using (3):

\[ c_s = \frac{1}{L} . \]

In systems consisting of various reaction domains, this gives rise to a residual concentration at the end of the process (Appendix A). Our goal is to describe the approach of the concentration to the residual limit.

For the full time analysis, we rewrite the empty interval probability as a sum

\[ E(x, t) = E_s(x) + E_{\text{trans}}(x, t) , \]

where \( E_{\text{trans}} \) symbolizes the transient behavior of interest. \( E_{\text{trans}} \) satisfies the same equation as \( E \), eq. (1), but the boundary condition (2) is replaced by

\[ E_{\text{trans}}(0, t) = 0 . \]

A spectral decomposition then yields

\[ E_{\text{trans}} = \sum_{n=1}^{\infty} a_n \sin \left( \frac{n\pi x}{L} \right) e^{-\lambda_n t} ; \quad \lambda_n = \frac{2n^2\pi^2 D}{L^2} , \]

where the \( \{a_n\} \) are determined from initial conditions.

For practical purposes, we consider the case where the segment is initially filled with particles at concentration \( c_0 \). We assume that the particles are randomly (Poisson) distributed, and hence the initial empty interval probability, given that the segment contains at least one particle, is

\[ E(x, 0) = \frac{e^{-c_0 x} - e^{-c_0 L}}{1 - e^{-c_0 L}} , \]

and

\[ E_{\text{trans}}(x, 0) = \frac{e^{-c_0 x} - e^{-c_0 L}}{1 - e^{-c_0 L}} - \frac{L - x}{L} . \]

Performing a Fourier analysis and comparing to eq. (11), we find

\[ a_n = \frac{2c_0^2 L^2(\cos n\pi - e^{c_0 L})}{(-1 + e^{c_0 L})\pi(c_0^2 L^2 + n^2\pi^2)} . \]

Finally, from this and eqs. (9) and (11) we conclude that

\[ c_L(t) = \frac{2}{L} \sum_{n=1}^{\infty} \frac{e^{c_0 L} - (-1)^n}{(e^{c_0 L} - 1)(1 + n^2\pi^2/c_0^2 L^2)} e^{-2n^2\pi^2 D t} . \]

The residual concentration \( 1/L \) has been eliminated, so that \( c_L(t) \) represents the approach to the asymptotic limit (in a single segment of length \( L \), and given that there is at least one particle at the start).

It is easily verified that the limit \( L \to \infty \) yields the known result for the infinite line [13]:

\[ c_\infty(t) = c_0 e^{c_0 t} \text{erfc}(\sqrt{t/t_0}) , \quad t_0 = \frac{1}{2c_0^2 D} . \]

If \( L \) is finite, however, at long times, \( t \gg t_1 = L^2/(2\pi^2 D) \), the sum in (13) is dominated by the slowest decaying exponential:

\[ c_L \sim a(L)e^{-t/t_1}, \quad a(L) = \frac{2}{L(1 + \pi^2/c_0^2 L^2)} e^{c_0 L} - 1 . \]

III. SYSTEMS CONSISTING OF SEGMENTS OF RANDOM LENGTH

We now have all the tools for the analysis of diffusion-limited coalescence in a system consisting of a collection of segments of various lengths, taken from the distribution \( \psi(L) \). The approach to the steady state is given by

\[ c(t) = \langle L \rangle^{-1} \int_0^\infty c_L(t) L\psi(L) dL , \]

where \( \langle L \rangle = \int_0^\infty L\psi(L) dL \) is the average length of the segments. Here and henceforth we write \( c(t) \) instead of
\[ c(t) - c(\infty), \] for the sake of brevity. The residual concentration of particles at the end of the process, \( c(\infty) \), is analyzed in Appendix A.

Eq. (18), in conjunction with eq. (13), gives the exact concentration decay, however, some approximations are necessary in order to reveal the asymptotic trends. Thus, we replace the exact result for \( c_L(t) \) with the limiting behavior of eqs. (10) and (17), but expressing the crossover between the two regimes in terms of the length \( \Lambda = \sqrt{2\pi^2Dt} \) rather than the time \( t_1 \):

\[
 c_L(t) \sim \begin{cases} 
 c_\infty(t) & L \gg \Lambda, \\
 a(L)e^{-t/t_1} & L \ll \Lambda. 
\end{cases}
\]

Then,

\[ c(t) = \langle L \rangle^{-1} \left\{ \int_0^\Lambda a(L)e^{-t/t_1} L\psi(L) dL + c_\infty(\Lambda) \int_\Lambda^\infty L\psi(L) dL \right\} \equiv c_<(t) + c_>(t), \]

where \( c_<(t) \) and \( c_>(t) \) represent the contribution to \( c(t) \) from short (\( L < \Lambda \)) and long (\( L > \Lambda \)) segments respectively. Furthermore, a reasonable approximation to (19) is:

\[ c_\infty(t) \sim \frac{1}{1/c_0 + \sqrt{2\pi D\tau}}. \]

Essentially, at times \( t \gg t_0 = (2c_0 D)^{-1} \), the initial concentration has no effect and \( c_\infty \sim 1/\sqrt{2\pi D\tau} \).

A. Exponential distribution of segment lengths

Consider first a completely random distribution of segment lengths, \( \psi(L) = pe^{-pL} \). This might be the case in chain compounds when the concentration of disorder (defects along the polymers which discourage diffusion and effectively terminate the chains) is \( p \), and the average segment length is \( \langle L \rangle = 1/p \). We also assume that \( c_0 \gg \rho \), that is, the initial average number of particles per segment is large. The contribution from short segments can then be estimated using a saddle point approximation:

\[ c_<(t) \sim 4\sqrt{\frac{3}{\pi}} p^{1/6} t^{1/3} e^{-3^{1/3}t}, \quad \tau \gtrsim 1, \]

where \( \tau = t/(2/\pi^2 Dp^2) \) is a reduced (dimensionless) time. \( c_<(t) \) is essentially the same as the survival probability in the famous trapping problem [19], and for similar reasons: the stretched exponential decay results from the longer survival of particles in very long segments, weighed against the relative scarcity of such segments.

The contribution from long segments can be computed analytically:

\[ c_>(t) = c_\infty(t)[1 + \rho \Lambda(t)]e^{-\rho \Lambda(t)}. \]

Then, with \( c_0 \gg \rho \), and using the approximation (21), we have

\[ c_>(t) \sim \sqrt{\pi \rho}(1 + \frac{1}{2\sqrt{\tau}})e^{-2\sqrt{\tau}}, \quad \tau \gtrsim 1. \]

The two contributions, \( c_< \) and \( c_> \), become equal at \( \tau \approx 3.5 \). Thus, a crossover is expected between the stretched exponential decay with exponent 1/2 at early times (but greater than \( 2/\pi^2 Dp^2 \)) and a stretched exponential decay with exponent 1/3 at later times. In reality, the concentration decay is stretched-exponential, with a slowly-varying exponent; from roughly 1 at \( \tau \approx 1 \), to 1/2 at \( \tau \approx 10 \), to 1/3 at \( \tau \gtrsim 100 \) (figure 1). At the very beginning of the process, \( t \ll 2/\pi^2 Dp^2 \) (\( \tau \ll 1 \)), the survival probability is practically the same as in an infinite system: \( c(t) \approx c_\infty(t) \) of eq. (14), or (22).

B. Power-law distribution of segment lengths

Consider next a power-law distribution of the segment lengths:

\[ \psi(L) = \begin{cases} 
 0 & L < b, \\
 (\gamma - 1)b^{\gamma - 1}L^{-\gamma} & L > b, 
\end{cases} \]

where \( b \) is a microscopic cutoff length, and \( \gamma > 1 \). This is an example of a long-tailed distribution, and it is also characteristic of percolation clusters at criticality. If \( \gamma < 2 \) then the average segment length diverges (in the thermodynamic limit, when the size of the system is infinite). In this case the system is dominated by the largest segment, which then coincides with the overall size of the system, \( L \), and \( c(t) \sim c_L(t) \) of Section II.

For \( \gamma > 2 \) the average segment length is finite: \( \langle L \rangle = [(\gamma - 1)/(\gamma - 2)]b \). The contribution from short segments can be written as
\[ c_<(t) = (\gamma - 2)b^{\gamma - 2} \int_1^\Lambda a(\frac{\Lambda}{u})e^{-u^2(\frac{\Lambda}{u})^1-\gamma} \frac{\Lambda}{u^2} du \] (26)

Since \( b \) is a microscopic cutoff lengthscale, we assume that \( cb \leq 1 \). In this case \( t \gg t_0 = 1/(2\sqrt{D}) \) implies \( \Lambda \gg b \). Due to the term \( e^{-u^2} \), the main contribution to the integral in (26) comes from \( u \approx 1 \), in which case \( a(\Lambda/u) \approx 2u/\Lambda \), and the upper integration limit may be extended to \(+\infty\). Thus,

\[ c_<(t) \sim (\gamma - 2)(\Lambda + 1)b^{\gamma - 2}(2\pi^2 D t)^{(1-\gamma)/2}, \] (27)

where \( \Gamma(a, x) = \int_x^\infty z^{a-1}e^{-z} dz \) is the incomplete gamma function [21].

The contribution from the large segments, for \( t \gg t_0 \) (and \( cb \ll 1 \)), is

\[ c_>(t) = c_\infty(t) \left( \frac{b}{\Lambda} \right)^{\gamma - 2} \sim \sqrt{\pi} b^{\gamma - 2}(2\pi^2 D t)^{(1-\gamma)/2}. \] (28)

Thus, the contributions from short and long segments are of the same order of magnitude at all times \( t \gg t_0 \), and the overall concentration decays in power-law fashion, \( c(t) \sim (1/b)(\beta^2 / D t)^{(\gamma - 1)/2} \), faster than the \( 1/\sqrt{Dt} \)-decay of coalescence in the infinite line.

IV. REACTIONS IN PERCOLATION CLUSTERS

The physical properties of three-dimensional systems with disorder are well represented by the percolation model of second-order phase transitions [23]. A relevant example is triplet exciton annihilation in naphtalene-hs embedded in porous matrices of naphtalene-ds [13]. The domains of reaction of naphtalene-hs can then be modeled by percolation clusters. Suppose that a reaction domain, i.e., a percolation cluster, has mass \( s \). Then the concentration of particles in the coalescence (or annihilation) process is [21, 26, 27]:

\[ c_s(t) \sim \begin{cases} \frac{t^{-d_s/2}}{s} & s > \sigma, \\ \frac{t^{-d_s/2}}{t_1} & s \leq \sigma, \end{cases} \] (29)

where \( d_s \approx 4/3 \) is the spectral (or fracton) dimension of the clusters, and \( \sigma \) is the crossover mass between algebraic and exponential decay.

Consider first percolation well below the critical threshold, when the distribution of cluster masses is \( n(s) = pe^{-ps} \). The typical cluster mass \( 1/p \) is smaller the farther one is from the percolation transition. Then, following a reasoning similar to the one of Section III.A, we find

\[ c(t) \sim \rho t^{-d_s/2}e^{-\tau d_s/2} + \rho(1 + \tau^{-d_s/2})e^{-\tau d_s/2}, \] (30)

where \( \tau = \rho^{2/d_s}t \) is the reduced time, and the first and second terms on the r.h.s. represent \( c_< \) and \( c_> \), respectively. We note that the largest (and rarest) clusters in percolation below criticality — which are the ones that actually determine the asymptotic behavior of \( c_< \) — are different from the typical clusters. They are known as “lattice animals”, and their spectral dimension \( d_s' \approx 1.18 \) (in \( d = 3 \)) is somewhat smaller than \( d_s \). Thus, the exponent characterizing the stretched exponential decay of \( c(t) \) varies slowly from \( d_s/2 \approx 0.67 \) at \( \tau \approx 1 \), to \( d_s'/ (2 + d_s') \approx 0.37 \) at later times.

Exactly at criticality, the distribution of the largest (dominant) clusters is given by \( n(s) \sim s^{-\tau} \), with \( \tau \) slightly larger than 2 (\( \tau \approx 2.186 \), in three-dimensional percolation). Following the argument for power-law distributions we then find

\[ c(t) \sim t^{-(\tau-1)d_s/2}, \] (31)

where once again the contributions from small and large clusters are comparable. Using the known values of the various exponents in three dimensions we predict an algebraic decay: \( c(t) \sim t^{-0.79} \).

Well above criticality the system is dominated by the spanning (infinite) cluster. This cluster has fractal structure, and is characterized by the spectral dimension \( d_s \approx 4/3 \), only for lengthscales shorter than \( 2 (\tau \approx 2.186) \), in three-dimensional percolation). Following the argument for power-law distributions we find

\[ c(t) \sim \begin{cases} t^{-d_s/2} & t < \xi_d, \\ t^{-1} & t > \xi_d, \end{cases} \] (32)

where \( d_w \approx 3.88 \) is the walk dimension of diffusion in the fractal clusters. The \( 1/t \)-concentration decay at lengthscales larger than \( \xi \) takes place because the upper critical dimension of diffusion-limited coalescence (and annihilation) is \( d = 2 \), so that in the effectively three-dimensional cluster the kinetics is reaction-limited.

V. SUMMARY AND DISCUSSION

Starting from the exact solution of diffusion-limited coalescence \( (A + A \rightarrow A) \) in a segment of finite length \( L \), we have computed the approach of the concentration to its steady-state limit, in a system consisting of disjoint segments characterized by the distribution of their lengths, \( \psi(L) \). For an exponential distribution, \( \psi(L) = pe^{-\rho L} \), as might be the case in polymer chains with an element of disorder, we find three different regimes: (a) an early time regime where the concentration decay is similar to that in the infinite line, \( c \sim 1/\sqrt{Dt} \), (b) an intermediate time regime, dominated by a stretched-exponential
decay, \( c \sim e^{-\alpha t^{1/2}} \), and (c) a late time regime where \( c \sim e^{-\beta t^{1/3}} \) (\( \alpha \) and \( \beta \) are known constants). In practice, the exponent characterizing the stretched-exponential decay in regimes (b) and (c) varies slowly over time, due to crossover effects and amplitude corrections (figure 1). It would be interesting to compare our theoretical results to the anomalous decay of photogenerated excitons observed in TMCCM chains and MX compounds. We note that the theory contains no free parameters beyond the diffusion coefficient of the excitons, \( D \), and the concentration of disorder (defects), \( \rho \). We have also considered a long-tailed distribution of segment lengths, \( \psi(L) \sim L^{-\gamma} (\gamma > 2) \). In this case the concentration decay is \( c \sim t^{-\gamma/2} \), faster than in the infinite line.

The above results were extended to systems in higher dimensions, where the reaction zones could be modeled by percolation theory (e.g., annihilation of triplet-excitons in naphtalene-h\(_8\) embedded in porous matrices of napthalene-d\(_8\)). The results are similar to those in one dimension, but depend crucially on whether the system is above, below, or at the critical threshold of percolation, and \( d_s \) (the spectral dimension of the clusters) affects the actual value of the various decay exponents. Below criticality, we find an early time decay of \( c \sim t^{-d_s/2} \sim t^{-0.66} \), followed by a stretched-exponential decay, \( c \sim \exp[(\text{const})t^{1/2}] \), with a slowly varying exponent: from \( x = d_s/2 \approx 0.66 \) to \( x = d_s'/2(2 + d_s') \approx 0.37 \) (\( d_s' \) is the spectral dimension of lattice animals, and all numerical values cited pertain to the physical case of three-dimensional percolation) [23]. At criticality there is a faster power-law decay, \( c \sim t^{-(d_s/2)} \approx t^{-0.79} \), and above criticality there is a crossover from \( c \sim t^{-d_s/2} \approx t^{-0.66} \) at early times to a reaction-limited decay, \( c \sim t^{-1} \), thereafter.

As a final observation, we note that the scaling of the concentration decay for diffusion-limited annihilation \( (A + A \rightarrow 0) \) is exactly the same as that of coalescence, and hence the results presented here, in spite of being grounded on the exactly solvable case of coalescence in a segment, are valid also for annihilation. Likewise, the concentration decay at all times, and in particular the long-time exponential decay of eq. (13), remains essentially the same when the particles are absorbed at the segment edges, rather than reflected. Thus, our conclusions are pretty independent of the fate of the particles as they hit the boundaries of the reaction domains.

**ACKNOWLEDGMENTS**

We thank Prof. Harry L. Frisch for introducing us to the problem, and we gratefully acknowledge support from the National Science Foundation (PHY-9820569).

**APPENDIX A: RESIDUAL PARTICLES**

Consider a finite domain of volume \( V \) where coalescence or annihilation might take place. At the end of the process, when no further reactions are possible, there would be left either one or zero particles. For coalescence, \( A + A \rightarrow A \), the residual number of particles is one, unless the volume had been empty to begin with. In the case of annihilation, \( A + A \rightarrow 0 \), the parity of the initial number of particles determines the residue: zero when the initial number is even, and one when it is odd.

Suppose that the initial distribution of particles is completely random, at concentration \( c_0 \). The probability that the domain \( V \) contains exactly \( n \) particles is given by the Poisson distribution:

\[
 p_n = \frac{(c_0 V)^n}{n!} e^{-c_0 V} . \tag{A1}
\]

Thus, for coalescence, the probability that one particle is left over is

\[
 p_{\text{coal}} = 1 - p_0 = 1 - e^{-c_0 V} , \tag{A2}
\]

while for annihilation it is

\[
 p_{\text{annil}} = \sum_{n \text{ odd}} p_n = \frac{1 - e^{-2c_0 V}}{2} . \tag{A3}
\]

If the system consists of disconnected domains of reaction, characterized by a distribution \( \psi(V) \) of their volumes, and the initial concentration of particles is \( c_0 \), then the concentration of residual particles at the end of the process is

\[
 c_{\text{res}} = \left\langle V \right\rangle^{-1} \int p_{\text{res}}(c_0, V) \psi(V) dV , \tag{A4}
\]

where \( \left\langle V \right\rangle = \int V \psi(V) dV \) is the average volume of the reaction domains. For example, if the distribution of reaction-domain sizes is exponential, \( \psi(V) = \rho e^{-\rho V} \), we obtain \( c_{\text{res}}^{\text{coal}} = c_0 \rho/(c_0 + \rho) \) and \( c_{\text{res}}^{\text{annil}} = c_0 \rho/(2c_0 + \rho) \). De Albuquerque and Lyra [4] performed a lattice computation for the residual concentration of particles for annihilation in two-dimensional percolation, under the assumption that there is initially one particle per site.

[1] Hinrichsen, K. Krebs, and M. P. Pfannmüller, “Finite-size scaling of one-dimensional reaction-diffusion systems. Part I: analytic results”, J. Stat. Phys. 78, 1429 (1995); B. Wehefritz, K. Krebs, and M. P. Pfannmüller, “Finite-size scaling of one-dimensional reaction-diffusion systems. Part II: Numerical methods”, J. Stat. Phys. 78, 1471 (1995); H. Hinrichsen, K. Krebs, and I. Peschel, “Solution of a one-dimensional reaction-diffusion model with spatial asymmetry”, Z. Phys. B 100, 105 (1996).
H. Takayasu, I. Nishikawa and H. Tasaki, “Power-law
V. Privman, “Exact results for diffusion-limited reac-
A. A. Lushnikov, “Binary reaction 1 + 1
M. Bramson and D. Griffeath, “Clustering and dispersion
J. L. Spouge, “Exact solutions for a diffusion-reaction
L. Peliti, “Path integral approach to birth-death pro-
Z. Rácz, “Diffusion-controlled annihilation in the pres-
F. K. Fang, ed., (Springer-Verlag, Heidelberg, 1976);
R. Kopelman, “Exciton percolation in molecular al-
S. Havlin and D. ben-Avraham, “Diffusion in disordered
M. Abramowitz and I. M. Stegun, eds., Handbook of
M. Sahimi, Applications of Percolation Theory, (Taylor & Francis, London, 1994).
P. Meakin and H. E. Stanley, “Novel dimension-
M. Hoyuelos and H. O. Martín, “Annihilation and co-
D. C. Torney and H. M. McConnell, “Diffusion-limited
D. Balboni, P.-A. Rey, and M. Droz, “Universality of
M. Henkel, E. Orlandini, and G. M. Schütz, “Equivalence
D. Ben-Avraham, M. A. Burschka, and C. R. Doering,
J. Simon, “Concentration for one and two species one-
M. Henkel, E. Orlandini, and J. Santos, “Reaction-diffusion processes from equivalent integrable quantum
D. C. Torney and H. M. McConnell, “Diffusion-limited
L. Peliti, “Path integral approach to birth-death processes on a lattice”, J. Phys., Paris 46, 1469 (1985); J. Phys. A 19, L365 (1985).
Z. Rácz and M. Plischke, “Correlations in a nonequilibrium steady state: exact results for a generalized kinetic Ising model”, Acta Physica Hungarica 62 203 (1987).
A. A. Lushnikov, “Binary reaction 1 + 1 → 0 in one dimension”, Phys. Lett. A 120, 135 (1987).
H. Takayasu, I. Nishikawa and H. Tasaki, “Power-law mass distribution of aggregation systems with injection”, Phys. Rev. A 37, 3110 (1988).
V. Privman, “Exact results for diffusion-limited reactions with synchronous dynamics”, Phys. Rev. E 50, 50 (1994); “Exact results for 1D conserved order parameter model”, Mod. Phys. Lett. B 8, 143 (1994); V. Privman, A.M.R. Cadilhe, and M.L. Lawrence, “Exact solutions of anisotropic diffusion-limited reactions with coagulation and annihilation”, J. Stat. Phys. 81, 881 (1995); “Anisotropic diffusion-limited reactions with coagulation and annihilation”, Phys. Rev. E 53, 739 (1996).
D. ben-Avraham, M. A. Burschka, and C. R. Doering, “Statics and dynamics of a diffusion-limited reaction: Anomalous kinetics, nonequilibrium self-ordering, and a dynamic transition”, J. Stat. Phys. 60, 695 (1990).
E. L. de Albuquerque and M. L. Lyra, “Scaling behavior of diffusion limited reactions on random media”, J. Chem. Phys. 105, 5945 (1996).
N. Kuroda, Y. Wakabayashi, M. Nishida, N. Wakabayashi, M. Yamashita, and N. Matsushita, “Decay kinetics of long-lived photogenerated kinks in an MX chain compound”, Phys. Rev. Lett. 79, 2510 (1997).
H. Okamoto, Y. Oka, T. Mitani, and M. Yamashita, “Dynamics of solitons and polarons in the quasi-one-dimensional MX chain compound [Pt(en)_2][Pt(en)_2I_2](ClO_4)_2”, Phys. Rev. B 55, 6330 (1997).

R. Kroon and R. Sprik, “Diffusion-limited exciton kinetics in one-dimensional systems”, in Nonequilibrium Statistical Mechanics in One Dimension, V. Privman, ed., (Cambridge University Press, 1997).
R. Kopelman, “Exciton percolation in molecular alloys and aggregates”, in Topics in Applied Physics 15, F. K. Fang, ed., (Springer-Verlag, Heidelberg, 1976); “Rate processes on fractals: Theory, simulations, and experiments”, J. Stat. Phys. 42, 185 (1986); “Low-dimensional exciton reactions”, Phil. Mag. B 56, 717 (1987); “Fractal reaction-kinetics”, Science 241, 1620 (1988).
N. D. Donsker and S. R. S. Varadhan, “On the number of distinct sites visited by a random walk”, Comm. Pure Appl. Math. 32, 721 (1979).
S. Havlin and D. ben-Avraham, “Diffusion in disordered media”, Adv. Phys. 36, 695 (1987).
M. Abramowitz and I. M. Stegun, eds., Handbook of Mathematical Functions, (Dover Publications, New York, 1964).
M. Sahimi, Applications of Percolation Theory, (Taylor & Francis, London, 1994).
P. Meakin and H. E. Stanley, “Novel dimension-independent behavior for diffusive annihilation on percolation fractals”, J. Phys. A 17, L173 (1984).
M. Hoyuelos and H. O. Martín, “Annihilation and coagulation reactions in low-dimensional substrata: Effects of probability of reaction and short-range interactions”, Langmuir 12, 61 (1996).
Our results are quite different from [14]. We note that their numerical simulations for two-dimensional percolation were for τ ≲ 20, not long enough to study the long-time asymptotic behavior.
