

The Long-Time Behavior of Initially Separated $A + B \rightarrow 0$ Reaction-Diffusion Systems with Arbitrary Diffusion Constants

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We examine the long-time behavior of $A + B \rightarrow 0$ reaction-diffusion systems with initially segregated species A and B. All of our analysis is carried out for arbitrary (positive) values of the diffusion constants D_A and D_B and initial concentrations a_0 and b_0 of A's and B's. We divide the domain of the partial differential equations describing the problem into several regions in which they can be reduced to simpler, solvable equations, and we merge the solutions. Thus we derive general formulas for the concentration profiles outside the reaction zone, the location of the reaction zone center, and the total reaction rate. An asymptotic condition for the reaction front to be stationary is also derived. The properties of the reaction layer are studied in the mean-field approximation, and we show that not only the scaling exponents, but also the scaling functions are independent of D_A , D_B , a_0 and b_0 .

KEY WORDS: Reaction kinetics; diffusion; segregation; partial differential equations.

1. INTRODUCTION

The study of the interfacial region formed in diffusion-limited $A + B \rightarrow 0$ type reactions between domains of unlike species has attracted much current interest.⁽¹⁻²¹⁾ A natural way to examine this problem is to prepare a system with the components initially segregated along the plane $x=0$, and then investigate the spatiotemporal evolution of their concentrations ρ_A and ρ_B , and the reaction rate R . Such a geometry, first studied by Gálfi and Rácz⁽²⁾, has been investigated by means of various methods, including experiments,⁽³⁻⁵⁾ numerical simulations,⁽⁶⁻⁹⁾ the renormalization group

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approach,⁽¹⁰⁻¹²⁾ analytical computations,⁽¹³⁻¹⁵⁾ and scaling^(2, 9, 16, 17) and dimensional⁽¹⁶⁻¹⁸⁾ analyses.

A standard way to treat the initially separated problem analytically is to solve the partial differential equations⁽²⁾

$$\left. \begin{aligned} \frac{\partial \rho_A}{\partial t} &= D_A \frac{\partial^2 \rho_A}{\partial x^2} - R \\ \frac{\partial \rho_B}{\partial t} &= D_B \frac{\partial^2 \rho_B}{\partial x^2} - R \end{aligned} \right\} \quad (1)$$

with the initial state given by

$$\left. \begin{aligned} \rho_A(x, t=0) &= a_0 H(-x) \\ \rho_B(x, t=0) &= b_0 H(x) \end{aligned} \right\} \quad (2)$$

where $\rho_A(x, t)$ and $\rho_B(x, t)$ are the local concentrations of A's and B's, respectively, R is the reaction rate, $H(x)$ denotes the Heaviside step function, and a_0 , b_0 , D_A and D_B are some positive constants related to the initial concentrations of species A and B and their diffusion coefficients, respectively. It is customary^(2, 8, 9, 11, 15-20) to assume $D_A = D_B \equiv D$, which leads to the conclusion that $u(x, t) \equiv \rho_A - \rho_B$ obeys the readily solvable diffusion equation $\partial_t u = D \partial_x^2 u$ irrespective of R . Finally, some form of R must be assumed, and in most cases either the mean-field approximation $R \propto \rho_A \rho_B$ (refs. 2, 13, 15, 19) or its generalization $R \propto \rho_A^m \rho_B^n$ (refs. 8, 9, 16, and 17) was adopted.

With these assumptions, two fundamental concepts were developed, both referring to the long-time limit. According to the first one,⁽²⁾ the long time behavior of the system inside the reaction layer can be described with a help of some scaling functions S_A , S_B and S_R through

$$\rho_A(x, t) \propto t^{-\gamma} S_A \left(\frac{x - x_f(t)}{t^\alpha} \right) \quad (3)$$

$$\rho_B(x, t) \propto t^{-\gamma} S_B \left(\frac{x - x_f(t)}{t^\alpha} \right) \quad (4)$$

$$R(x, t) \propto t^{-\beta} S_R \left(\frac{x - x_f(t)}{t^\alpha} \right) \quad (5)$$

where $x_f(t)$ denotes the point at which the reaction rate R attains its maximal value, and exponents α , β , and γ are some positive constants given, for

$R \propto \rho_A^m \rho_B^n$, by $\gamma = 1/(m+n+1)$, $\alpha = 1/2 - \gamma$ and $\beta = 1 - \gamma$.⁽⁹⁾ The scaling ansatz is based on the assumption that the width $w(t)$ of the reaction layer grows with time as t^α with $\alpha < 1/2$, so that in addition to the diffusion length scale $\lambda_D \sim \sqrt{Dt}$, the problem possesses also another relevant length scale $w \propto t^\alpha$.

According to the second theory, called the quasistationary approximation^(16, 19) the currents $J_A(t)$ and $J_B(t)$ of particles A and B arriving at the interface layer from the two densely occupied domains are changing so slowly that the relatively narrow interface has enough time to equilibrate. To equilibrate means here to reach a state completely determined by the current boundary conditions, i.e., by J_A and J_B . Mathematically this is equivalent to the assumption that the state of the reaction zone is entirely given by equations obtained from (1) by replacing their left sides, or the time derivatives, with zero. This leads to much simpler equations

$$\left. \begin{aligned} D_A \frac{\partial^2 \rho_A}{\partial x^2} &= R \\ D_B \frac{\partial^2 \rho_B}{\partial x^2} &= R \end{aligned} \right\} \quad (6)$$

which are to be solved with the boundary conditions $\partial \rho_A / \partial x \rightarrow -J_A(t)$ and $\rho_B \rightarrow 0$ as $x \rightarrow -\infty$, and $\rho_A \rightarrow 0$, $\partial \rho_B / \partial x \rightarrow J_B(t)$ as $x \rightarrow +\infty$. The most important feature of the quasistationary equations (6) is that they depend only on x , with time t being a parameter entering their solutions $\rho_A(x, t)$ and $\rho_B(x, t)$ only through the time dependent boundary currents J_A and J_B .

It was conjectured by Gálfi and Rácz⁽²⁾ that the first of the above assumptions, $D_A = D_B$, is irrelevant with regard to the long-time behavior of the system, the ratio D_A/D_B affecting perhaps the form of the scaling functions S_A , S_B and S_R , but not the values of exponents α , β , and γ . This hypothesis was generally accepted after numerical⁽⁶⁾ and experimental⁽³⁾ verification. Alas, this verification encompassed only the case where the ratio D_A/D_B was of order 1, whereas it is known^(16, 20) that if one of the diffusion constants is equal to zero, the mean-field exponents assume values entirely different from those predicted by Gálfi and Rácz, namely $\alpha = 0$, $\beta = 1/2$, and $\gamma = 1/4$.

This situation improved when Lee and Cardy^(10, 12) presented a renormalization group analysis of the general case of initially separated A + B → 0 systems with arbitrary positive values of diffusion constants D_A and D_B . In particular, they confirmed that the values of exponents α , β , and γ do

not depend on the ratio D_A/D_B ; they also conjectured that the form of the scaling functions S_A , S_B , and S_R should be the same for any nonzero D_A and D_B . The aim of our paper is to present another approach to the general theory comprising the case of any positive diffusion constants D_A and D_B and initial concentrations a_0 and b_0 .

Unfortunately, we know of only three successful attempts to derive the macroscopic form of R from the microscopic properties of the system. One of them is the 1D system with $D_B = 0$.⁽¹⁴⁾ Dimensional^(9, 16, 18) and renormalization group⁽¹⁰⁻¹²⁾ analyses lead to another important conclusion that the mean-field approximation is valid in spaces of dimension higher than $d_c = 2$. The latter approach also enabled Howard and Cardy⁽¹¹⁾ to derive the asymptotic ($|x| \rightarrow \infty$) form of R in the general case of any dimension d and with fluctuation effects taken into account. Therefore our basic equation (1) might seem useful only for these three types of systems for which the form of R is known. In our approach, however, we will not need to impose any special restriction on the form of R . Instead, we will require that the solutions of (1) satisfy a few physically justifiable relations. Therefore our theory can be applied even to systems for which the form of R remains unknown, including experiments and microscopic models. In such cases verification of our postulates should be far easier than the task of finding the exact form of R , let alone solving (1) afterward.

The paper is organized as follows. In the next section we present the assumptions upon which our theory is founded, as well as their brief physical justification. The general theory is formulated in Section 3. In Section 4 we use it to derive and discuss the scaling ansatz in the mean-field approximation. Section 5 is devoted to conclusions.

2. ASSUMPTIONS

We will consider systems which can be described with the Gálfi and Rácz equations (1) and the boundary conditions (2). We will assume that D_A , D_B , a_0 and b_0 are some known positive constants. Our analysis will be based on a few physical assumptions:

(i) At any time $t > 0$ there exists a unique point $x_f(t)$ at which the reaction term R attains its maximal value, and a unique point $x_0(t)$ at which $D_A \rho_A(x_0, t) - D_B \rho_B(x_0, t) = 0$.

(ii) The reaction is concentrated in a region $|x - x_f| \sim w(t) \sim t^\alpha$ with $0 < \alpha < 1/2$. Outside this region, for $x \ll x_f - w$ there is $\rho_A \gg \rho_B$, and for $x \gg x_f + w$ we have $\rho_A \ll \rho_B$.

(iii) The evolution of ρ_A in the region $x \ll x_f - w$ can be approximated by

$$\rho_A(x, t) = a_0 - C_A [\operatorname{erf}(x/\sqrt{4D_A t}) + 1] \quad (7)$$

where C_A is a constant, and $\operatorname{erf}(x) \equiv 2\pi^{-1/2} \int_0^x \exp(-\eta^2) d\eta$ is the error function.⁽²²⁾

Similarly, for $x \gg x_f + w$, the evolution of ρ_B can be estimated by

$$\rho_B(x, t) = b_0 + C_B [\operatorname{erf}(x/\sqrt{4D_B t}) - 1] \quad (8)$$

where C_B denotes another constant. Both C_A and C_B depend on the initial parameters a_0 , b_0 , D_A , and D_B .

(iv) The quasistatic approximation is valid in the region $-(D_A t)^{1/2} \ll x \ll (D_B t)^{1/2}$.

The first assumption introduces two functions $x_f(t)$ and $x_0(t)$, restricting the considerations to the cases where they are uniquely defined. Function x_f identifies directly the location of the reaction layer at time t , and x_0 is an auxiliary mathematical object helpful in examining the behavior of x_f . That x_0 exists for any $t > 0$ stems from the initial conditions (2). As for the second postulate, it is satisfied by all the A + B → 0 interfacial systems examined so far. The third assumption comes from the observation that, due to postulate (ii), in the region $x \ll x_f - w$ the concentration of particles A is expected to be much greater than that of B's, the latter having to cross the whole reaction layer to get there. Therefore, the evolution of A's is practically unaffected by B's, and so it should be governed by the standard diffusion equation $\partial_t \rho_A = D_A \partial_x^2 \rho_A$. The particular, based on the error function form (7) of its solution was predicted and experimentally confirmed by Koo and Kopelman.⁽³⁾ Notice also that for any time t such a form of ρ_A guarantees that the relation $\lim_{x \rightarrow -\infty} \rho_A = a_0$ implied by the initial conditions (2) is also fulfilled. A similar argument leads to (8). As for the last postulate, the quasistationary approximation is based on the following observation.⁽¹⁶⁾ The diffusion current of particles arriving at the reaction layer is $J \propto t^{1/2}$, so the characteristic time scale on which this current changes is $\tau_J \propto (d \log J/dt)^{-1} \propto t$, whereas the equilibration time of the reaction front is $\tau_F \propto \tilde{w}^2 \propto t^{2\alpha}$; therefore $\alpha < 1/2$ implies that as time goes to infinity, the ratio τ_F/τ_J goes to 0, validating the quasistatic approximation.

As we mentioned above, we will not impose any explicit restrictions on the form of the macroscopic reaction rate R , requiring only that it be consistent with the above postulates. However, to investigate the behavior

of the $A + B \rightarrow 0$ system inside the reaction zone we will need more detailed information about R . Therefore in Section 4 we will concentrate on the mean-field approximation $R \propto \rho_A \rho_B$.

3. ANALYSIS

The following observation constitutes the basis of the analysis of our model. For sufficiently long time t , at any point x we can employ either assumption (iii) or (iv) or both of them (see Fig. 1). Therefore we can divide the x axis into several regions, and in each of them the initial problem of solving (1) can be reduced to a much simpler one. Then, the overlapping of the domains of applicability of (iii) and (iv) will enable us to merge the solutions.

Consider first the region $-\sqrt{D_A t} \ll x \ll \sqrt{D_B t}$. By assumption (iv) the system is governed here by the quasistationary equations (6). They imply that $\Psi(x, t) \equiv D_B \rho_B - D_A \rho_A$ satisfies $\partial^2 \Psi / \partial x^2 = 0$. Therefore Ψ is linear in x . Let $J(t)$ denote its slope. By definition of x_0 we have $\Psi(x_0, t) = 0$. Thus we arrive at the conclusion that at sufficiently long time t , for $-\sqrt{D_A t} \ll x \ll \sqrt{D_B t}$, we have

$$D_B \rho_B - D_A \rho_A \approx J(t)(x - x_0(t)) \quad (9)$$

and so $J_A(t) = J_B(t) = J(t)$. The notation $f(t) \approx g(t)$ means $\lim_{t \rightarrow \infty} f(t)/g(t) = 1$.

Consider now the region $-\sqrt{D_A t} \ll x \ll x_f - w$, so that $\varepsilon \equiv x_f - x$ satisfies $t^\alpha \ll \varepsilon \ll t^{1/2}$. Applying assumption (ii) to (9), we can approximate the form of ρ_A by

$$\rho_A(x, t) \approx -D_A^{-1} J(t)(x - x_0(t)) \quad (10)$$

On the other hand, however, by assumption (iii), here ρ_A can be as well expressed by Eq. (7). So we have

$$a_0 - C_A \left[\operatorname{erf} \left(\frac{x_f(t) - \varepsilon}{\sqrt{4D_A t}} \right) + 1 \right] \approx -D_A^{-1} J(t)(x_f(t) - x_0(t) - \varepsilon) \quad (11)$$

and

$$\left. \frac{\partial}{\partial x} \left(a_0 - C_A \left[\operatorname{erf} \left(\frac{x}{\sqrt{4D_A t}} \right) + 1 \right] \right) \right|_{x_f - \varepsilon} \approx -D_A^{-1} J(t) \quad (12)$$

By assumption (ii), for any x located outside the reaction layer, the ratio ρ_A/ρ_B will either converge to zero, or diverge to infinity as $t \rightarrow \infty$.

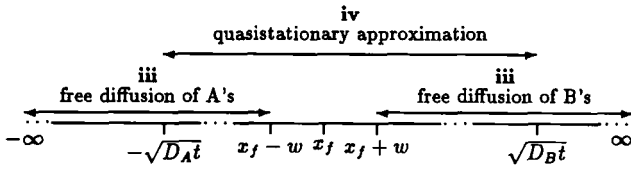


Fig. 1. Schematic diagram of the regions of applicability of postulates (iii) and (iv). Asymptotically $w(t) \propto t^2 \ll t^{1/2}$.

However, by definition of x_0 , this ratio assumes the constant value D_B/D_A at $x = x_0$. So x_0 must lie inside the reaction layer. As its width grows as t^2 , we conclude that there must exist a number θ such that $|x_f(t) - x_0(t)| \leq \theta t^\alpha$. We can see now that in the long-time limit $|x_f - x_0|$ becomes negligibly small compared to ε , which, in turn, gets negligibly small compared to $t^{1/2}$. Therefore we can drop ε on the left hand side of (11) and (12) and $x_f - x_0$ on the rhs of (11). After these transformations the asymptotic value of the lhs of (11) turns out to be independent of ε , whereas the rhs of (11) becomes proportional to $\varepsilon J(t)$. As ε can vary between t^α and $t^{1/2}$, we conclude that $J(t) \varepsilon(t)$ goes either to 0 or to ∞ . The latter case is impossible because (11) approximates the value of ρ_A , which must be finite. In the long-time limit we therefore have

$$J(t) \varepsilon(t) \rightarrow 0 \tag{13}$$

$$a_0 - C_A \left[\operatorname{erf} \left(\frac{x_f(t)}{\sqrt{4D_A t}} \right) + 1 \right] \rightarrow 0 \tag{14}$$

and

$$J(t) \sqrt{t} \rightarrow C_A \left(\frac{D_A}{\pi} \right)^{1/2} \exp \left(-\frac{x_f^2(t)}{4D_A t} \right) \tag{15}$$

Similar arguments applied to the region $x_f + w \ll x \ll \sqrt{D_B t}$ lead to

$$b_0 + C_B \left[\operatorname{erf} \left(\frac{x_f(t)}{\sqrt{4D_B t}} \right) - 1 \right] \rightarrow 0 \tag{16}$$

and

$$J(t) \sqrt{t} \rightarrow C_B \left(\frac{D_B}{\pi} \right)^{1/2} \exp \left(-\frac{x_f^2(t)}{4D_B t} \right) \tag{17}$$

It follows from (14) and (16) that in the long-time limit

$$x_f(t)/\sqrt{t} \rightarrow C_f \quad (18)$$

where C_f is a constant given either by

$$C_f = 2\sqrt{D_A} \operatorname{erf}^{-1}[(a_0 - C_A)/C_A] \quad (19)$$

or

$$C_f = 2\sqrt{D_B} \operatorname{erf}^{-1}[(C_B - b_0)/C_B] \quad (20)$$

Now (15), (17), and (18) imply that as time goes to infinity we have

$$J(t) \sqrt{t} \rightarrow C_J \quad (21)$$

where C_J is another constant given either by

$$C_J = C_A \sqrt{D_A/\pi} \exp(-C_f^2/4D_A) \quad (22)$$

or

$$C_J = C_B \sqrt{D_B/\pi} \exp(-C_f^2/4D_B) \quad (23)$$

Notice that (21) is consistent with (13).

So far we have introduced four constants C_A , C_B , C_f and C_J . The first two of them, C_A and C_B , control the asymptotic profile of the majority species outside the reaction flayer. The third constant, C_f , governs the location of the reaction layer center. Finally, through the formula $J(t) \approx \int R(x, t) dx \approx C_J/t^{1/2}$, parameter C_J is related to the magnitude of the current $J(t)$ of particles entering the reaction layer, or, equivalently, the total reaction rate at time t . Due to the form of the initial state (2) we expect $\partial_x \rho_A \leq 0$ and $\partial_x \rho_B \geq 0$, which implies $C_A > 0$, $C_B > 0$, and $C_J > 0$.

Equations (19), (20), (22), and (23) can be reduced to

$$\Phi\left(\frac{-C_f}{2\sqrt{D_A}}\right) = \frac{a_0\sqrt{D_A}}{b_0\sqrt{D_B}} \Phi\left(\frac{C_f}{2\sqrt{D_B}}\right) \quad (24)$$

where

$$\Phi(x) \equiv [1 - \operatorname{erf}(x)] \exp(x^2) \quad (25)$$

An important feature of $\Phi(x)$ is that it diminishes monotonically from ∞ to 0 as x grows from $-\infty$ to ∞ . This property guarantees that Eq. (24) always has a unique solution $C_f = C_f(a_0/b_0, D_A, D_B)$, which, moreover, can be readily found numerically. The only problem that can appear while

solving (24) numerically is that when x is positive, $\Phi(x)$ is a product of a very small and a very big number. For this reason, if x is greater than 5, we suggest using the asymptotic form $\Phi(x) \approx 1/(\sqrt{\pi}x)$, which comes from the asymptotic properties of the error function erf.⁽²²⁾

With C_f computed from (24), the values of C_A , C_B and C_f can now be calculated from (19), (20), and (22). The opposite statement is also true: if we know (e.g., from an experiment) the values of C_A , C_B , C_f and C_f , our equations determine uniquely the values of a_0 , b_0 , D_A , and D_B .

The immediate consequence of (24) is that the sign of C_f is determined by the sign of $a_0\sqrt{D_A}/(b_0\sqrt{D_B}) - 1$. In particular, we conclude that

$$C_f = 0 \Leftrightarrow a_0\sqrt{D_A} = b_0\sqrt{D_B} \quad (26)$$

This formula is important for planning experiments, as it clarifies the way the initial concentrations of the species should be chosen in order to have the reaction layer move asymptotically as slowly as possible. Condition (26) is consistent with that of Jiang and Ebner,⁽⁶⁾ who, by numerical examination of the mean-field approximation $R \propto \rho_A\rho_B$, found a stronger relation $x_f = 0 \Leftrightarrow a_0\sqrt{D_A} = b_0\sqrt{D_B}$. Our general formula, derived for any reaction term R , implies only that with this particular choice of the initial parameters can the function x_f not be changing as fast as $t^{1/2}$. An example of a system where $C_f = 0$ and $x_f(t) \propto t^\alpha$ was investigated in ref. 17.

Equation (24) enables us also to observe a striking similarity between the long- and short-time behavior of x_f . According to ref. 13, in the short-time limit the reaction term does not affect the solutions of (1), and so ρ_A and ρ_B assume the same forms as in the readily solvable case $R = 0$. The point x_f can be then found as the point at which $\partial R/\partial x = 0$. For $R \propto \rho_A^m\rho_B^n$ such a procedure yields $\lim_{t \rightarrow 0} x_f/\sqrt{t} = C_0$, where C_0 can be found from a relation very similar to that of (24),

$$\Phi\left(\frac{C_0}{2\sqrt{D_A}}\right) = \frac{m\sqrt{D_B}}{n\sqrt{D_A}}\Phi\left(\frac{-C_0}{2\sqrt{D_B}}\right) \quad (27)$$

4. THE REACTION LAYER

In the previous section we carried out our analysis without imposing any restrictions on the form of the macroscopic reaction term R . As we now proceed to examine the asymptotic properties of the reaction layer, we will obviously need more specific information about R . Therefore we will concentrate on the mean-field approximation $R = k\rho_A\rho_B$, $k = \text{const}$, still allowing a_0 , b_0 , D_A , and D_B to take any positive values.

By assumption (iv) we expect that in the region $-(D_A t)^{1/2} \ll x \ll (D_B t)^{1/2}$ we can apply the quasistatic approximation equations (6). Let $\rho_A(x, t)$ and $\rho_B(x, t)$ denote their solutions for some values of D_A , D_B , $x_0(t)$, and $J(t)$. By the following linear transformation we introduce two new functions of a single variable $\tilde{\rho}_A(z)$ and $\tilde{\rho}_B(z)$:

$$\begin{aligned}\rho_A(x, t) &= \eta_A(t) \tilde{\rho}_A[(x - x_0(t))/w(t)] \\ \rho_B(x, t) &= \eta_B(t) \tilde{\rho}_B[(x - x_0(t))/w(t)]\end{aligned}\quad (28)$$

where

$$w(t) \equiv \left(\frac{D_A D_B}{k J(t)}\right)^{1/3} = \left(\frac{D_A D_B}{k C_J}\right)^{1/3} t^{1/6} \quad (29)$$

$$\eta_A(t) \equiv \frac{J(t) w(t)}{D_A} = \left(\frac{D_B}{k}\right)^{1/3} \left(\frac{C_J}{D_A}\right)^{2/3} t^{-1/3} \quad (30)$$

$$\eta_B(t) \equiv \frac{J(t) w(t)}{D_B} = \left(\frac{D_A}{k}\right)^{1/3} \left(\frac{C_J}{D_B}\right)^{2/3} t^{-1/3} \quad (31)$$

Denoting $\tilde{R}(x) \equiv \tilde{\rho}_A(x) \tilde{\rho}_B(x)$, we have also

$$R(x, t) = k \rho_A \rho_B = C_J^{4/3} (D_A D_B)^{-1/3} k^{1/3} t^{-2/3} \tilde{R}[(x - x_0)/w(t)] \quad (32)$$

The essential property of $\tilde{\rho}_A(x)$ and $\tilde{\rho}_B(x)$ is that they constitute the particular solution to Eq. (6) with $D_A = D_B = J = k = 1$ and $x_0 = 0$. Therefore, by symmetry, $\tilde{R}(x)$ assumes its maximal value for $x = 0$, and so Eq. (32) implies that R attains the maximal value at $x = x_0$. In the long-time limit we can therefore identify x_f with x_0 . Comparing now (28) and (32) with the scaling ansatz (3)–(5), we see that we can also identify $\tilde{\rho}_A$ with S_A , $\tilde{\rho}_B$ with S_B , and \tilde{R} with S_R . Therefore not only are the above formulae consistent with Gálfi and Rácz's scaling ansatz, but through $C_J(a_0, b_0, D_A, D_B)$ they also *exactly* relate the quantities of physical importance [e.g., $w(t)$] to the parameters of the system (D_A , D_B , a_0 , b_0 , and k).

Because (28) can be applied to systems with any positive values of "external" parameters a_0 , b_0 , D_A , D_B and k , we arrive at the conclusion that the long-time evolution of initially segregated $A + B \rightarrow 0$ systems is even more universal than was predicted by Gálfi and Rácz; namely, not only the scaling exponents, but also the form of the scaling functions does not depend on the external parameters. Therefore, to find the scaling properties of the reaction layer it is sufficient to concentrate on the simplest, symmetric case $D_A = D_B$ and $a_0 = b_0$.

Notice that we have achieved these results by means of a simple linear transformation (28). In this way we took advantage of the very feature of Eqs. (1) and (6) that prevents them from being solved analytically—nonlinearity.

The above analysis is straightforward and can be easily generalized for many other reaction terms R . In particular, for $R = k\rho_A^m\rho_B^n$, with $k = \text{const}$ and m, n being any (positive) real numbers, the following relation should be used instead of (29):

$$w^{m+n+1} \equiv D_A^m D_B^n k^{-1} J^{1-m-n} \quad (33)$$

This formula, together with (21), (30), and (31), generalizes the scaling theory of Cornell *et al.*⁽⁹⁾ for the case of any positive a_0, b_0, D_A, D_B, m , and n .

5. CONCLUSIONS

We have investigated the long-time behavior of the concentrations ρ_A and ρ_B of phases A and B in the Gálfi and Rácz problem. Our analysis is the first analytical attempt to consider it in the general case of arbitrary positive initial concentrations a_0 and b_0 and diffusion constants D_A and D_B of A's and B's.

Our approach is very general, as it does not impose any restrictions on the form of the macroscopic reaction rate R . Instead, it is based on the assumption that in the long time limit ρ_A and ρ_B satisfy a few physically justifiable relations. Therefore our theory can be applied to various systems, including those for which the form of the macroscopic reaction rate R remains unknown. Another peculiar feature of our theory is that, unlike most previous studies, it does not concentrate on the investigation of the reaction layer only, but takes into account the properties of the whole, infinite system.

In this way we managed to derive general formulas for the concentration profiles of the majority species outside the reaction layer, the location of the layer, and the total reaction rate. It is interesting to notice that these quantities turned out to be independent of R . We also derived analytically Jiang and Ebner's condition for the reaction front to be asymptotically stationary. This relation also turned out to be independent of R . These results correspond to the recent findings based on dimensional analysis,^(9, 16, 18) according to which the scaling properties of the reaction layer are independent of the form of R .

Next we derived the general scaling ansatz for the mean-field approximation. We gave the formulas which exactly relate some quantities of physical importance, (e.g., the width w of the reaction layer) to the external parameters of the system a_0, b_0, D_A, D_B and k . It turned out that not only

the scaling exponents, but also the forms of the scaling functions are independent of the values of these parameters. This justifies the customary approach of examining the properties of the reaction layer only in the simplest, symmetric case $a_0 = b_0$ and $D_A = D_B$.

Our work suggests also that the behavior of the reaction-diffusion system can be understood as a subtle interplay between two scaling regimes. The first one is valid far from the reaction zone, where the densities of particles A and B assume the scaling forms typical of purely diffusive systems: $\rho_A(x, t) \approx \Psi_A(x/t^{1/2})$ and $\rho_B(x, t) \approx \Psi_B(x/t^{1/2})$. These scaling laws determine also the location of the point $x_f(t)$ of the maximal reaction and the magnitude of the current $J(t)$ of the particles entering the reaction zone. However, at x_f the spatial derivatives of Ψ_A and Ψ_B suffer discontinuity. Therefore in the vicinity of x_f a new form of scaling develops, and ρ_A and ρ_B assume the form $\rho_A(x, t) = S_A(x/t^\alpha)$ and $\rho_B(x, t) = S_B(x/t^\alpha)$ with $\alpha < 1/2$.

Although we confined our considerations to the long-time limit, it would be interesting to combine our results with those of Taitelbaum *et al.*⁽¹³⁾ for short and intermediate times. We believe that the striking similarity between Eqs. (24) and (27) is not accidental and should lead to a general theory comprising the short-, intermediate-, and long-time limits. The first attempt in this direction has already been made.⁽²¹⁾

Notice also that the quasistationary approximation leads to new definitions of "short"-, "intermediate"-, and "long"-time regimes. Namely, we can define them as the time intervals in which the reaction term in the vicinity of x_f is vanishingly small compared to the time derivative ("short time"); or the interval in which they are of similar magnitude ("intermediate time"); or the interval in which it is the time derivative that can be neglected ("long time").

Another interesting problem concerns the limit $D_A \rightarrow 0$ with other external parameters fixed. In this limit the scaling exponents (in the mean-field approximation) are expected to change from $\alpha = 1/6$, $\beta = 2/3$ to $\alpha = 0$, $\beta = 1/2$. A paper in which this problem is examined within the framework of the theory presented here is under preparation. We mention here only that as D_A goes to 0, the time at which the mean-field system reaches the long-time regime can be shown to go to infinity, so the case $D_A = 0$ can be considered as the case where the system always remains in the "intermediate"-time regime.

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