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Properties of the asymptotic nA + mB → C reaction-diffusion fronts

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Abstract. We discuss, at the mean-field level, the asymptotic shape of the reaction fronts in the general $nA + mB \to C$ reaction-diffusion processes with initially separated reactants, thereby generalizing to arbitrary reaction-order kinetics the work done by Gálfi and Rácz for the case $n = m = 1$. The obtained information allows us to calculate the asymptotic density of C particles deposited by the moving reaction front, a quantity that plays an important role in the theories of Liesegang patterns formation.

PACS. 82.20.Mj Nonequilibrium kinetics – 82.20.Db Statistical theories (including transition state) – 66.30.Ny Chemical interdiffusion; diffusion barriers

1 Introduction

Consider two reactants, initially separated, which are put in contact at time $t = 0$ and start to mix one into each other by diffusion. A *reaction zone* (*i.e.* a region where the reaction rate is high) will develop at their interface. The mathematical function describing the variation, in space and time, of the amplitude of the reaction rate is usually called a *reaction front* $R(\mathbf{x}, t)$.

Dynamical properties of reaction fronts in (purely or effectively) one-dimensional reaction-diffusion systems have been the object of numerous studies [1–12]. In general, it is observed that these fronts obey *asymptotic scaling*, characterized by a scaling function G and scaling exponents α, γ:

$$R \sim t^{-\gamma} \Phi \left(\frac{x - x_{\rm f}(t)}{t^{\alpha}} \right). \qquad (1)$$

In the previous expression, $x_{\rm f}(t)$ locates the position of the front (usually defined as its first moment), which generally obeys

$$x_{\rm f}(t) \propto \sqrt{t}, \qquad (2)$$

accounting for the diffusive origin of the front's dynamics.

In the framework of a mean-field approximation (which is ours from now on), scaling hypothesis, together with balance considerations, can lead quite directly to the values of the dynamical scaling exponents (see [1,2]). Accessing the structure (*i.e.* the *shape*) of the scaling function itself requires however to go one level deeper into the analysis of the process in consideration.

In addition to satisfying a purely theoretical curiosity, knowing better Φ itself can provide a practical advantage. For example, in studying phenomena involving reaction-diffusion processes, like Liesegang patterns formation, it may be useful to dispose of an explicit, approximated analytical form for Φ. This allows, for example, to bypass the dynamical generation of this front in a numerical simulation and save computation time [14]. Finding such an analytical approximation requires evidently to gain sufficient information about the scaling function.

Both tasks (derivation of the scaling exponents *and* of the scaling function) were accomplished in the pioneering paper by Gálfi and Rácz [1], where they studied the reaction front in the $A + B \xrightarrow{k} C$ process with initially segregated A-s and B-s in mean-field approximation.

In the present paper, we provide a generalization of their work to the case of arbitrary reaction-order kinetics, $nA{+}mB \xrightarrow{k} C$, and explain how to calculate the associated C density profile in the asymptotic regime.

An important motivation for this generalization is the following: in the case of Liesegang patterns, the primary chemical reaction leading (through several complex coarsening processes) to the formation of precipitate turns out to be most often of the types $A + 2B$ or $2A + B$, and not $A + B$, as usually considered for simplicity in theoretical models [13–15].

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2 Scaling analysis

2.1 Definition and notations

The case of mean-field, general reaction-order kinetics in the initially segregated reactants case has already been addressed in [2]. Using scaling analysis, the authors showed that the exponents controlling the asymptotic behaviour of the reaction front are given, in terms of the reaction-order constants, by:

$$\alpha(n,m) = \frac{n+m-1}{2(n+m+1)}\ , \ \gamma(n,m) = \frac{1}{n+m+1} \tag{3}$$

where α, γ are the same as in (1), and (n,m) are integers both ≥ 1.

It is important to note the following properties of these exponents:

1. $\alpha(n,m) = \alpha(n+m), \quad \gamma(n,m) = \gamma(n+m)$.
2. $\alpha(n,m) < 1/2 \ \forall\, (n,m), \quad \alpha$ increases monotonically from $1/6$ $(n=m=1)$ to $1/2$ $(n+m \to \infty)$.
3. $\alpha(n,m) + \gamma(n,m) = 1/2$.

We can start from the above results to formulate a general derivation that will lead us to the family of ordinary differential equations defining the asymptotic shape of the reaction fronts $R_{(n,m)}$[1]. To this goal, let us consider the following one dimensional initial-value problem, describing a reaction-diffusion process between initially separated A and B particles in the mean-field approximation:

$$\partial_T A(X,T) = D_A \partial_X^2 A(X,T) - kn(A^n B^m)(X,T) \tag{4a}$$
$$\partial_T B(X,T) = D_B \partial_X^2 B(X,T) - km(A^n B^m)(X,T) \tag{4b}$$
$$\partial_T C(X,T) = k(A^n B^m)(X,T), \tag{4c}$$

with

$$A(X,T=0) = a_0 \theta(-X), \tag{5a}$$
$$B(X,T=0) = b_0 \theta(X), \tag{5b}$$
$$C(X,T=0) \equiv 0. \tag{5c}$$

In the above equations,

- θ denotes the Heaviside step function $[\,\theta(X<0) = 0, \quad \theta(X \geq 0) = 1\,]$.
- A, B and C are concentrations with dimensions $[A,B,C] = [X^{-1}]$.
- D_A and D_B are diffusion coefficients $([D_A, D_B] = [X^2 T^{-1}])$.
- k is the reaction rate $([k] = [X^{n+m-1} T^{-1}])$.

In the following, we will only consider the case of equal diffusion coefficients, $D_A = D_B \equiv D$, since the method we are going to use requires this strong condition to be satisfied. The asymmetric case $D_A \neq D_B$ reveals to be several orders of magnitude higher in difficulty. Interesting results have been obtained in the case $n = m = 1$, in connection to the front's dynamics [7,8], but a derivation of the shape of the scaling functions for arbitrary D_A/D_B seems to be still out of reach for the moment.

The first step in our calculation is to render the problem adimensional. This can be done through the following change of variables:

$$x \equiv \sqrt{\frac{k a_0^{n+m-1}}{D}}\, X, \tag{6a}$$
$$t \equiv k a_0^{n+m-1} T, \tag{6b}$$
$$a, b, c \equiv A/a_0, B/b_0, C/c_0. \tag{6c}$$

The equations then read:

$$\partial_t a(x,t) = \partial_x^2 a(x,t) - n a^n(x,t) b^m(x,t), \tag{7a}$$
$$\partial_t b(x,t) = \partial_x^2 b(x,t) - m a^n(x,t) b^m(x,t), \tag{7b}$$
$$\partial_t c(x,t) = a^n(x,t) b^m(x,t), \tag{7c}$$

with

$$a(x,t=0) = \theta(-x), \tag{8a}$$
$$b(x,t=0) = \frac{b_0}{a_0}\theta(x), \tag{8b}$$
$$c(x,t=0) \equiv 0. \tag{8c}$$

2.2 Solution for a − (n/m)b

We define:

$$u(x,t) \equiv \left(a - \frac{n}{m} b\right)(x,t). \tag{9}$$

This function obeys the diffusion equation:

$$\partial_t u(x,t) = \partial_x^2 u(x,t), \tag{10a}$$
$$u(x<0, t=0) = 1, \tag{10b}$$
$$u(x>0, t=0) = -\frac{n}{m}\frac{b_0}{a_0} \equiv -\frac{n}{m} q, \tag{10c}$$

whose solution reads

$$u(x,t) = \frac{1}{2}\left((1 - \frac{n}{m} q) - (1 + \frac{n}{m} q) \mathrm{erf}\left(\frac{x}{2\sqrt{t}}\right)\right). \tag{11}$$

In the above equation, erf denotes the error function, $\mathrm{erf}(x) \equiv (2/\sqrt{\pi}) \int_0^x \exp(-w^2) \mathrm{d}w$.

Let $x_\mathrm{f}(t)$ be such that $u(x_\mathrm{f}(t), t) = 0$. One can check that

$$x_\mathrm{f}(t) = \sqrt{2 D_\mathrm{f} t}, \tag{12}$$

with $D_\mathrm{f} = D_\mathrm{f}(q)$ given by:

$$\mathrm{erf}\left(\sqrt{\frac{D_\mathrm{f}}{2}}\right) = \frac{1 - \frac{n}{m} q}{1 + \frac{n}{m} q}\,. \tag{13}$$

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[1] This derivation follows closely the steps and notation in [1], and the reader should refer to it for further details and justifications.

2.3 Equation for a in the reaction zone

We write now $b = \frac{m}{n}(a-u)$ and plug it into (7a), thereby getting an equation for a involving only a and the known function u:

$$\partial_t a(x,t) = \partial_x^2 a(x,t) - n\left(\frac{m}{n}\right)^m \Big[a^n(a-u)^m\Big](x,t). \quad (14)$$

We are interested in the solution of this equation in the reactive region $|x - x_\mathrm{f}| \approx t^{\alpha(n,m)}$. As the latter is believed to widen with a time exponent $\alpha(n,m) < 1/2$, this allows us to expand u around x_f to the lowest-order in $x/\sqrt{t}$, since the neglected terms will vanish as $t \to \infty$:

$$u(x,t) \approx -K\frac{x - x_\mathrm{f}}{\sqrt{t}} \qquad |x - x_\mathrm{f}| \approx t^{\alpha(n,m)}, \quad (15)$$

with K given by:

$$K = \frac{1 + \frac{n}{m}q}{2\sqrt{\pi}}\exp(-D_\mathrm{f}/2). \quad (16)$$

The boundary conditions that the solution to (14) must satisfy in the reactive region are:

$$a(x \to -\infty, t) = -K\frac{x - x_\mathrm{f}}{\sqrt{t}}, \quad (17a)$$

$$a(x \to +\infty, t) = 0. \quad (17b)$$

2.4 Scaling hypothesis

We shall now assume that asymptotically (*i.e.* when $t \to \infty$), the solution to (14) adopts the following scaling form:

$$a(x,t) \approx t^{-\gamma(n,m)} G_{(n,m)}\left(\frac{x - x_\mathrm{f}(t)}{t^{\alpha(n,m)}}\right), \quad (18)$$

where $\{G_{(n,m)}\}_{n,m\geq 1}$ are a family of scaling functions remaining to be characterized. The scaling exponents are given by (3).

2.5 Differential equation for $\mathbf{G_{(n,m)}}$

Let's define first the reaction zone coordinate z:

$$z \equiv \frac{x - x_\mathrm{f}}{t^{\alpha(n,m)}}\cdot \quad (19)$$

Inside the reaction zone, u and $b = \frac{m}{n}(a-u)$ write

$$u(z) = -Kt^{\alpha(n,m)-1/2}z, \quad (20)$$

$$b(z) = \frac{m}{n}(t^{-\gamma(n,m)}[G_{(n,m)}(z) + Kz]). \quad (21)$$

Using (18), equation (14) becomes:

$$\begin{aligned} &t^{2\alpha(n,m)-1}[-\gamma(n,m)G_{(n,m)} - \alpha(n,m)z\partial_z G_{(n,m)}] \\ &- \sqrt{\frac{D_\mathrm{f}}{2}}t^{\alpha(n,m)-1/2}\partial_z G_{(n,m)} \\ &= \partial_z^2 G_{(n,m)} - n\left(\frac{m}{n}\right)^m G^n_{(n,m)}[G_{(n,m)} + Kz]^m. \end{aligned} \quad (22)$$

We now take the asymptotic limit inside the reaction zone, *i.e.* we let $t \to \infty$, keeping z fixed. The two terms on the left-hand side vanish (remember that $\alpha_{(n,m)} < 1/2$!) and we remain with the following ordinary, non-linear second-order differential equation for the scaling functions $G_{(n,m)}$:

$$G''_{(n,m)}(z) = n\left(\frac{m}{n}\right)^m G^n_{(n,m)}(z)[G_{(n,m)}(z) + Kz]^m. \quad (23)$$

The boundary conditions (17a, 17b) imply the following asymptotics for $G_{(n,m)}$ [1]:

$$G_{(n,m)}(z) \to -Kz, \quad z \to -\infty, \quad (24a)$$

$$G_{(n,m)}(z) \to 0, \quad z \to \infty. \quad (24b)$$

We are now left with a boundary value problem (23, 24) that can be solved numerically.

2.6 Solving the equation for $\mathbf{G_{(n,m)}}$

We can make the problem K-independent by rescaling G and z:

$$G \equiv K^{\mu(n,m)}\tilde{G}, \quad (25a)$$

$$z \equiv K^{\nu(n,m)}\tilde{z}, \quad (25b)$$

and by using a suitable choice for μ and ν. Inserting these scaled forms into (23) and imposing that K drops out leads to:

$$\mu(n,m) = \frac{2}{n+m+1} = \mu(n+m), \quad (26a)$$

$$\nu(n,m) = \frac{-(n+m-1)}{n+m+1} = \nu(n+m). \quad (26b)$$

The problem we are left to treat is now:

$$\tilde{G}''_{n,m}(\tilde{z}) = n\left(\frac{m}{n}\right)^m \tilde{G}^n_{n,m}(\tilde{z})\Big[\tilde{G}_{n,m}(\tilde{z}) + \tilde{z}\Big]^m, \quad (27a)$$

$$\tilde{G}_{n,m}(\tilde{z}) \to -\tilde{z} \qquad \tilde{z} \to \infty, \quad (27b)$$

$$\tilde{G}_{n,m}(\tilde{z}) \to 0 \qquad \tilde{z} \to \infty. \quad (27c)$$

The reader should keep in mind, from now on, that the "tilde" sign stands for quantities expressed in terms of the rescaled K-independent version of the scaling function $G_{(n,m)}$ and reactive coordinate z.

2.7 The dimensionless reaction front

By definition:

$$\begin{aligned} R_{(n,m)}(x,t) &= a^n(x,t)b^m(x,t) \\ &= \left(\frac{m}{n}\right)^m t^{-\frac{n+m}{n+m+1}} G^n_{(n,m)}[G_{(n,m)} + Kz]^m \\ &\equiv t^{-\beta(n,m)}F_{(n,m)}(z), \end{aligned} \quad (28)$$

the last equality defining both the reaction rate amplitude exponent β and the asymptotic reaction front scaling function $F_{(n,m)}$.

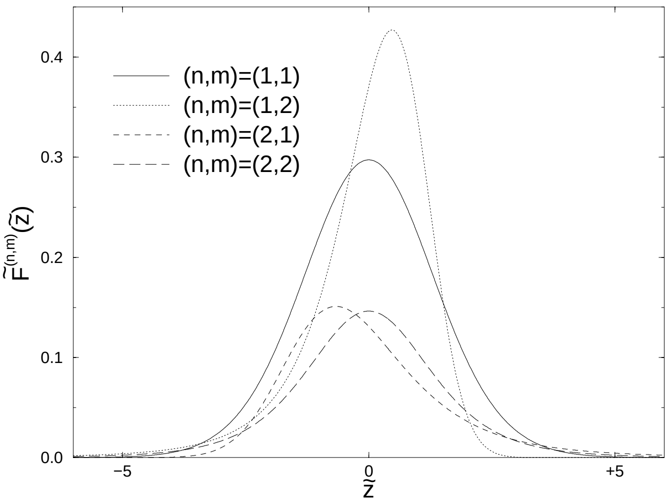

Fig. 1. Numerical solutions to (27) for $(1 \le n, m \le 2)$.

Figure 1 shows the result of the numerical computation of the fronts $\tilde{F}_{(n,m)} \equiv F(\tilde{G}, \tilde{z})$ for the cases $2 \le n+m \le 4$ (only such low values of $n+m$ are relevant in connection to experiments). The reader should not be surprised by the asymmetry between the $(n,m) = (1,2)$ and $(n,m) = (2,1)$ cases, as it is due to the passage from the variables $\{G_{(n,m)}, z\}$ to $\{\tilde{G}_{(n,m)}, \tilde{z}\}$. Both quantities are namely rescaled by a K-dependent factor which is *not* (n,m)-symmetric !

For $n = m$, the front is naturally a symmetric function of z. The particular case $(n,m) = (1,1)$ exhibits a front that is essentially localized: the dominant contribution to both tails at $z \to \pm\infty$ has been shown to be proportional to $z^{3/4} \exp(-z^{3/2})$ [9]. For the higher-order cases $n+m > 2$, one can make use of (23) to obtain a rough idea of the dominant contribution to the tails' decay. Approximating $Kz + G(z)$ by Kz for $z \to \pm\infty$, we can distinguish two qualitatively different cases: when $n = 1, m > 1$, the decay is dominated by $\exp(-z^{1+m/2})$ at $z \to \infty$, but remain algebraic for $z \to -\infty$: $R(z \to -\infty) \approx z^{-(2m+1)/(m-1)}$. When $n > 1, m > 1$, both tails are algebraic, with $R(z \to \infty) \approx z^{-(2m+n)/(n-1)}$. It is also worth noting that in the asymmetric cases $n \ne m$, x_f does not coincide with the maximum of the front.

As seen, a breaking of the fronts' symmetry can be induced by the asymmetry in the reaction-order coefficients n and m, but not by the asymmetry of either the initial reactant concentrations, or the diffusion coefficients [7]. Interestingly however, the singular case where one reactant is completely immobile ($D_b = 0$) falls into the class of the asymmetric and fast-decreasing fronts [8].

3 The C concentration profile

3.1 Derivation of the asymptotic profile

We are interested now in estimating the (possibly x-dependent) density $c_0^{(n,m)}(x)$ of C particles left behind by the fronts $R_{(n,m)}$ which travel diffusively through the system. This quantity is, for example, of great importance in the theories of Liesegang pattern formation [13,14].

In dimensionless units, $c_0^{(n,m)}$ is formally given by

$$c_0^{(n,m)}(x) \equiv \int\limits_0^\infty R_{(n,m)}(x,t)\,\mathrm{d}t. \tag{29}$$

Due to the several timescales dependence of $R_{(n,m)}$, this integral is difficult to handle. The estimation of $c_0^{(n,m)}(x)$ turns out however to be possible by making use of:

a) the precious algebraic relation $\alpha + \gamma = 1/2$ between the scaling exponents, and
b) the particular structure of the solutions to (27).

Let's consider first a narrow slice $\delta F_{(n,m)}(z_0, \delta w)$ of the scaling function $F_{(n,m)}$, centered on $z = z_0$, of width $\delta w \ll 1$. In the spirit of the Riemann integral, we can approximate the amplitude of δF inside $[z_0 - \delta w/2, z_0 + \delta w/2]$ by its value $F_{(n,m)}(z_0)$ at the center.

We can estimate the contribution $\delta C^{(n,m)}(z_0,x)/\delta x$ of this slice to the asymptotic local C density inside $[x, x+\delta x]$ as follows: the fraction of the front we are considering will reach x at a certain time $t(z_0,x)$. The quantity of C particles deposited in the interval $[x, x+\delta x]$ will be proportional to the amplitude, the width and inversely proportional to the speed of the slice at $t = t(z_0,x)$:

$$\begin{aligned}
&\delta C^{(n,m)}(z_0,x) \\
&\quad\approx \frac{t(z_0,x)^{-\gamma(n,m)} F_{(n,m)}(z_0) t(z_0,x)^{\alpha(n,m)} \delta w}{\sqrt{D_\mathrm{f}/(2t(z_0,x))}}\, \delta x \\
&\quad= \sqrt{2/D_\mathrm{f}} F_{(n,m)}(z_0) \delta w \, \delta x.
\end{aligned} \tag{30}$$

In other words, the contribution of the slice to the density at x is proportional to its "mass" $F_{(n,m)}(z_0)\delta w$ but independent of $t(z_0,x)$, and hence of x. This indicates that the asymptotic density profile is flat. By superposition, our argument leads immediately to the result we are looking for:

$$\begin{aligned}
c_0^{(n,m)}(x) \equiv c_0^{(n,m)} &= \text{const.} \\
&\approx \sqrt{2/D_\mathrm{f}} \int_{-\infty}^{\infty} F_{(n,m)}(z)\,\mathrm{d}z.
\end{aligned} \tag{31}$$

Now our real fortune is that we are able to evaluate analytically $\int F_{(n,m)}$. Using (23) and (28), we have

$$\begin{aligned}
\int_{-\infty}^{\infty} F_{(n,m)}(z)\,\mathrm{d}z = \frac{1}{n}\Big[&\partial_z G_{(n,m)}(z \to \infty) \\
&- \partial_z G_{(n,m)}(z \to -\infty)\Big].
\end{aligned} \tag{32}$$

It is intuitively clear, from purely physical considerations, that the solution will converge to its values at $\pm\infty$ in such a way that:

$$\lim_{z \to -\infty} G'_{(n,m)}(z) = -K \ , \ \lim_{z \to +\infty} G'_{(n,m)}(z) = 0 \tag{33}$$

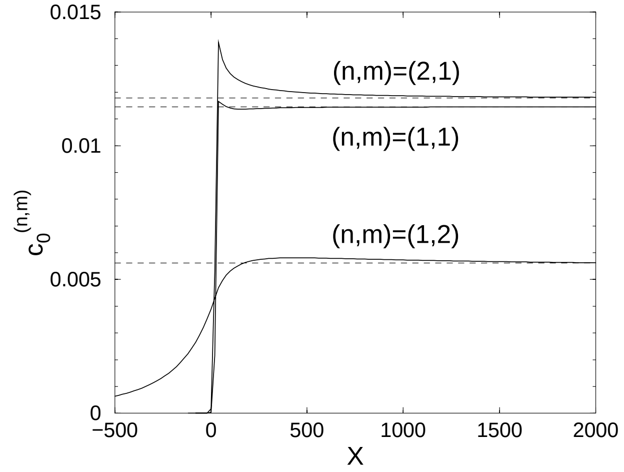

Fig. 2. Numerical solutions to (4) for $(2 \le n + m \le 3)$ (plain curves). The dashed lines indicate the asymptotic values as calculated from (13, 16, 36). The following values of the "free" parameters were chosen for all three cases: $D = 1$, $a_0 = 10^{-2} b_0 = 1$, $k = 0.1$. Units for X and k vary with (n, m) according to (6a), while $c_0^{(n,m)}$ is measured in units of $1/a_0$.

(see (27b, 27c)). So we finally have:

$$\int_{-\infty}^{\infty} F_{(n,m)}(z)\,\mathrm{d}z = \frac{K}{n}, \tag{34}$$

and we end up with the result:

$$c_0^{(n,m)} \approx \frac{K}{n}\sqrt{2/D_\mathrm{f}}. \tag{35}$$

Going back to the dimensional variables A, B, C, X and T, one can check that (35) writes:

$$c_0^{(n,m)} \approx \sqrt{\frac{2D}{D_\mathrm{f}}}\frac{Ka_0}{n}\,. \tag{36}$$

Figure 2 shows the $c_0^{(n,m)}$ profiles obtained by numerical integration of the reaction-diffusion equations (4), together with the asymptotic values predicted by (36).

3.2 Low q expansion

In the context of Liesegang patterns-forming experiments, which are generally based on the penetration of a highly concentrated solution into a dissolved one, it is useful to dispose of an expansion of this result for low $q \equiv b_0/a_0$ values. We recall that D_f is given by:

$$\mathrm{Erf}\left(\sqrt{\frac{D_\mathrm{f}}{2D}}\right) = \frac{1 - \frac{n}{m}q}{1 + \frac{n}{m}q} = 1 - 2\frac{n}{m}q \overset{q \ll 1, n \le 2}{\cong} 1. \tag{37}$$

The large x asymptotics of $\mathrm{Erf}(x)$ is given by [16]:

$$\mathrm{Erf}(x) \sim 1 - \frac{\mathrm{e}^{-x^2}}{\sqrt{\pi}x}\Big[1 - \frac{1}{2x^2} + \cdots\Big], \tag{38}$$

so we obtain from (37):

$$\sqrt{\frac{D}{D_\mathrm{f}}}\exp(-D_\mathrm{f}/2D) \cong \Big[1 - \frac{1}{2}\frac{D}{D_\mathrm{f}}\Big]^{-1} \times \sqrt{2\pi}\frac{\frac{n}{m}q}{1 + \frac{n}{m}q}, \tag{39}$$

which, together with the dimensional expression for K,

$$K = \frac{1 + \frac{n}{m}q}{2\sqrt{\pi}}\exp(-D_\mathrm{f}/2D), \tag{40}$$

gives finally:

$$c_0^{(n,m)} \cong \frac{b_0}{m}\left[1 + \frac{1}{2}\frac{D}{D_\mathrm{f}} + \mathcal{O}\left(\left(\frac{D}{D_\mathrm{f}}\right)^2\right)\right].$$

The physical meaning of this result is clear: if $D_A = D_B = D$ and $q \ll 1$, then $D_\mathrm{f} \gg D$ and the B particles appear as nearly immobile for the invading A-s. As m B-s are required to produce one C, the density equals b_0/m to a very good approximation.

However, in the typical conditions of a Liesegang experiment (where $D_A \cong D_B$ and $10^{-2} \le q \le 5 \times 10^{-2}$ typically), the first order correction in D/D_f to $c_0^{(n,m)}$ lies in the range $(0.1 - 0.2)(b_0/m)$, and should therefore, in principle, not be neglected as can be seen of Figure 2.

4 Summary

We have derived the family of ordinary differential equations defining the asymptotic shape of the reaction fronts in the $nA + mB \xrightarrow{k} C$ reaction-diffusion process with initially separated reactants (Eqs. (23, 24)). The four lowest-order cases in $n+m$ have been solved numerically (Fig. 1). We have also shown, and confirmed by numerical simulations, that the density $c_0^{(n,m)}$ of C particles deposited in the system by these traveling fronts is asymptotically constant (Fig. 2), and we have made explicit the dependence of this density on the reaction orders n, m, as well as on the material parameters D, a_0, k and b_0 entering the problem (Eq. (36)).

5 Conclusion

To conclude this study, we would like to comment on the interesting phenomenon shown by Figure 2. When, for the two $m = 1$ cases, no significant quantity of reaction product is created in the majority species subspace, the case where $m = 2$ exhibits, on contrary, an important deposit of C on the left hand side, up to far beyond the initial location of the interface. This fact must be evidently related to the details of the short-time dynamics of the reaction front. Some studies have already been carried on the early-time regime subject for the $n = m = 1$ case [17,18] in the past. They unveiled the existence of a surprisingly complex behaviour, including successive power-law regimes for the early front's dynamics, and even the possibility of a change in its direction of motion. Such nontrivial behaviour has also been observed numerically in the higher-order kinetics cases we have addressed in the present paper, and a detailed study of the dependence on (n, m) of the short-time dynamics should be worth being investigated.

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