Kinetic regimes and cross-over times in many-particle reacting systems

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Abstract. – We study kinetics of single-species reactions ("$A + A \to \emptyset$") for general local reactivity Q and dynamical exponent z (rms displacement $x_t \sim t^{1/z}$.) For small molecules $z = 2$, whilst $z = 4, 8$ for certain polymer systems. For dimensions d above the critical value $d_c = z$, kinetics are always mean field (MF). Below d_c, the density n_t initially follows MF decay, $n_0 - n_t \sim n_0^2 Q t$. A 2-body diffusion-controlled regime follows for strongly reactive systems ($Q > Q^* \sim n_0^{(z-d)/d}$) with $n_0 - n_t \approx n_0^2 x_t^d$. For $Q < Q^*$, MF kinetics persist, with $n_t \sim 1/Qt$. In all cases $n_t \approx 1/x_t^d$ at the longest times. Our analysis avoids decoupling approximations by instead postulating weak physically motivated bounds on correlation functions.

The kinetics of reactions between diffusing particles are anomalous in low spatial dimensions [1, 2]. Mean-field (MF) theory, according to which the reaction rate is proportional to the product of reactant densities, is only valid for dimensions d above a critical value d_c. In the most fundamental problem of single-species reactions into inert products, $A + A \to \emptyset$, for $d < d_c = 2$ the density decays asymptotically as $1/t^{d/2}$, independently of the magnitude of the local chemical reactivity Q and initial reactant density. That is, in low dimensions the $1/Qt$ decay predicted by MF theory is invalid. This is supported by numerical simulations [2-5] and renormalization group studies [6]. For the case of "infinitely" reactive particles for which $Q = \infty$ (probability of reaction per collision unity), rigorous bounds [7] on the asymptotic decay of density have been derived and in one dimension exact solutions exist [8-11]. Many other workers have employed approximate methods starting from the hierarchy of coupled dynamical equations for correlation functions of all orders which is then truncated by expressing higher-order correlation functions in terms of lower-order correlations [1, 12].

In this work we address the two major aspects of single-species reaction kinetics which remain poorly understood. 1) Although it is known that at the shortest timescales MF kinetics apply [13, 14], the crossover from the short-time MF kinetics to the asymptotic $1/t^{d/2}$ behavior has not been established. Some groups on the basis of numerical simulations [15-17]

have claimed an "intermediate" time regime during which density decays according to a non-universal Q-dependent power law. 2) No systematic theory has been able to predict reaction kinetics for general Q [6,13,14] across all time regimes. As a result numerical simulations have been fitted to empirical rate laws [18,19].

The present work will also consider general dynamics. The above results were for reactions between particles obeying simple Fickian diffusion for which the dynamical exponent z describing rms displacement x_t as a function of time, $x_t \approx a(t/t_a)^{1/z}$, is equal to 2. Here t_a is the time corresponding to diffusion distance of order the particle size a. For reactive groups attached to long polymer chains z can also be 4 or 8, depending on time and degree of entanglement [20]. In these cases the critical dimension is $d_c = z$ [21] below which the asymptotic density decay is $1/x_t^d$ [22].

In this letter we give a complete description of the sequence of kinetic regimes for the single-species reaction kinetics for arbitrary values of dynamical exponent z (where z is assumed a fixed number independent of dimension). Results are derived after postulating simple physical bounds on correlation functions without the need to resort to *ad hoc* decoupling approximations. In agreement with previous studies, we find that below the critical dimension $d_c = z$ the density n_t decays as $1/x_t^d$ at long times. We find two possible kinetic sequences for $d < d_c$, depending on the magnitude of Q with respect to a marginal value Q^*. For $Q < Q^*$ ("weak" systems) the crossover to $n_t \approx 1/x_t^d$ occurs after a $1/t$ MF regime. For $Q > Q^*$ ("strong" systems) the crossover to $1/x_t^d$ occurs before the $1/t$ regime has developed. In this case a short-time regime exists during which the number of reactions is proportional to the reactant exploration volume, $n_0 - n_t \approx n_0^2 x_t^d$.

We begin by noting that the average reaction rate $\dot{n}_t \equiv \frac{d}{dt} n_t$ at point $\mathbf{r}_1$ is proportional [23] to the 2-body correlation function $\rho_t(\mathbf{r}_1, \mathbf{r}_2)$ evaluated at $\mathbf{r}_1 = \mathbf{r}_2$. Due to translational invariance $\dot{n}_t = -\lambda \rho_t(0,0)$, where $\lambda \equiv Q a^d$. Now the dynamical equation for ρ involves the 3-body correlation function $\rho^{(3)}$. Following Doi [23] the exact equation is

$$\rho_t(\mathbf{r}_1, \mathbf{r}_2) = n_0^2 - \lambda \int d\mathbf{r}_1' \, d\mathbf{r}_2' \int_0^t dt' \, G_{t-t'}(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_1', \mathbf{r}_2') \, \rho_{t'}(\mathbf{r}_1', \mathbf{r}_2') \delta(\mathbf{r}_1' - \mathbf{r}_2')$$

$$- \lambda \int d\mathbf{r}_1' d\mathbf{r}_2' d\mathbf{r}_3' \int_0^t dt' G_{t-t'}(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_1', \mathbf{r}_2') \rho_{t'}^{(3)}(\mathbf{r}_1', \mathbf{r}_2', \mathbf{r}_3') \{\delta(\mathbf{r}_1' - \mathbf{r}_3') + \delta(\mathbf{r}_2' - \mathbf{r}_3')\}, \quad (1)$$

where $G_t(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_1', \mathbf{r}_2')$ is the net weighting for two particles to arrive at $\mathbf{r}_1, \mathbf{r}_2$ given starting points $\mathbf{r}_1', \mathbf{r}_2'$, in the absence of reactions. The sink terms on the right-hand side of eq. (1) describe the three ways in which reactions diminish ρ_t from its initial value n_0^2. The first *two-body* sink term subtracts off pairs which failed to reach $\mathbf{r}_1, \mathbf{r}_2$ because their members reacted with one another at $\mathbf{r}_1'$ at time t'. The remaining two sink terms subtract off pairs which would be at $\mathbf{r}_1, \mathbf{r}_2$ but only one member of which reacted at time t' at location $\mathbf{r}_3'$. Such a reaction involves a third particle, weighted by the appropriate 3-body correlation function. In eq. (1) we used a δ function as a reactive sink which is a coarse-grained description of the reaction process over a scale of order the particle size a.

Notice that eq. (1) is not closed in terms of ρ_t since it involves the unknown $\rho_t^{(3)}$. It is in fact impossible to write a closed exact equation for ρ_t since correlation functions of all orders are coupled in an infinite hierarchy of dynamical equations [23]. This complication may be resolved by approximating 3-body correlations as products of lower-order correlation functions [1]. However we are able to write a closed equation for ρ_t after assuming much less restrictive bounds on the magnitude of correlation functions. First we transform eq. (1) to an expression for the function $q_t(\mathbf{r}, 0) \equiv \rho_t(\mathbf{r}, 0)/n_t^2$. Notice that $q_t(0,0) = k_t/\lambda$, where the rate

constant k_t is defined by $\dot{n}_t = -k_t n_t^2$. We find that eq. (1) is transformed into

$$q_t(\mathbf{r}, 0) = 1 - \int_0^t \mathrm{d}t' \, G_{t-t'}^{\mathrm{sep}}(\mathbf{r}, 0) \, k_{t'} \; + \; \varphi_t(\mathbf{r}), \tag{2}$$

where $G_t^{\mathrm{sep}}(\mathbf{r}_1, \mathbf{r}_2) \equiv \int \mathrm{d}\mathbf{r}_2' \, G_t(\mathbf{r}_1', \mathbf{r}_1' + \mathbf{r}_1, \mathbf{r}_2', \mathbf{r}_2' + \mathbf{r}_2)$ is the probability density with which two particles are separated by $\mathbf{r}_1$ at time t given initial separation $\mathbf{r}_2$. The function φ_t is defined by

$$\varphi_t(\mathbf{r}) \equiv \int_0^t \mathrm{d}t' \int \mathrm{d}\mathbf{r}' \, G_{t-t'}^{\mathrm{sep}}(\mathbf{r}, \mathbf{r}') \, k_{t'} \, \mu_{t'}(\mathbf{r}'), \quad \mu_t(\mathbf{r}) \equiv 2 \left\{ \rho_t(\mathbf{r}|0) - \rho_t^{(3)}(\mathbf{r}|0, 0) \right\}. \tag{3}$$

Here $\rho_t(\mathbf{r}|0) \equiv \rho_t(\mathbf{r}, 0)/n_t$ and $\rho_t^{(3)}(\mathbf{r}|0, 0) \equiv \rho_t^{(3)}(\mathbf{r}, 0, 0)/\rho_t(0, 0)$ are the conditional densities at $\mathbf{r}$, given one and two particles, respectively, at the origin.

Now we would like to solve eq. (2) for $q_t(0, 0)$. This requires information on the properties of $\varphi_t(0)$ which involves unknown 2-body and 3-body conditional densities. To proceed, let us now make the following assumption: We assume that the more particles placed at the origin, the lower the conditional density. Chemical reactivity can only induce anticorrelations. Thus

$$\rho_t^{(3)}(\mathbf{r}|0, 0) \; \leq \; \rho_t(\mathbf{r}|0) \; \leq \; n_t \quad \text{(assumption)}. \tag{4}$$

Equation (4) implies the following constraints on μ:

$$0 \; \leq \; \mu_t(\mathbf{r}) \; \leq \; 2n_t, \quad \int_0^t \mathrm{d}t' k_{t'} \; \geq \; \int_0^t \mathrm{d}t' \int \mathrm{d}\mathbf{r}' \, k_{t'} \, \mu_{t'}(\mathbf{r}'). \tag{5}$$

The first constraint immediately follows from eq. (3) while the second is obtained by requiring that the magnitude of the second term on the right-hand side of eq. (2) exceeds that of the 3rd term (since $q_t \leq 1$) and then integrating over $\mathbf{r}$.

On the strength of the above constraints, eq. (5), we will argue below that the solution of eq. (2) for $q_t(0, 0)$, but with the term $\varphi_t(0)$ deleted, gives the correct power law solution for k_t to within a constant prefactor. Expressing $q_t(0, 0)$ in terms of k_t, setting $\mathbf{r} = 0$ and deleting $\varphi_t(0)$, eq. (2) becomes

$$k_t = \lambda - \lambda \int_0^t \mathrm{d}t' S_{t-t'} k_{t'}, \tag{6}$$

where we have introduced the return probability $S_t \equiv G_t^{\mathrm{sep}}(0, 0) \approx 1/x_t^d$. The validity of this approximation is justified by self-consistent arguments whose outline is as follows (full details will be published elsewhere [24]). If one accepts eq. (6) then one obtains a sequence of power law regimes in time for k_t and n_t (see below). Using these solutions in eq. (5) and in the expression for $\varphi_t(0)$ (eq. (3)), we have determined the function $\mu_{t'}^{\mathrm{max}}(\mathbf{r}')$ which maximizes $\varphi_t(0)$ (for a given time t) subject to the constraints of eq. (5). This in turn implies an upper bound $\varphi_t^{\mathrm{max}}(0)$ on $\varphi_t(0)$. Now for short times, we find that this bound is much less than unity: hence $\varphi_t(0)$ may be deleted in eq. (2) without error and even prefactors are expected to be correct. Meanwhile, for long times ($n_t \approx 1/x_t^d$) we find $\varphi_t^{\mathrm{max}}(0) = A$, where A is a constant of order unity. We then make a second simple assumption: we assume k_t remains a power law at long times. We have shown that the deletion of $\varphi_t(0)$ from eq. (2) is then valid insofar as it will generate the correct power laws for k_t (albeit with possibly incorrect prefactors).

It is now straightforward to solve eq. (6) for k_t. For $d > z$ the integral term in eq. (6) is negligible and one recovers the expected MF rate constant $k \approx \lambda$, true for all times. For $d \leq z$ one finds two regimes: for $t < t_2^*$ (see definition eq. (9) below) the integral term in eq. (6) is
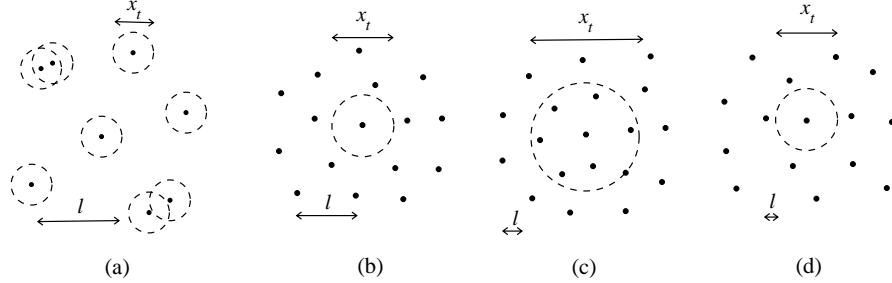
Fig. 1. – Snapshots of particle distributions during various kinetic regimes for $d < z$. Rms displacement is x_t and l is the typical initial separation among reactants. (a) Times $t \ll t_l$. Reactions are due to the few isolated pairs which happened to be initially close enough such that their exploration volumes (indicated by dashed lines) overlap by t. For strong systems most of these pairs will have reacted by t_2^*, leading to $n_0 - n_t \approx n_0^2 x_t^d$. (b) The situation at time $t = t_l$. A region of size x_t^d contained of order one particle initially. For strong systems reaction kinetics are already diffusion-controlled and a crossover occurs to $n_t \approx 1/x_t^d$. (c) Weak systems, $t_l \ll t \ll t_2^*$. For weak systems MF kinetics persist beyond t_l. Many reactants now exist within the exploration volume of a given particle. Reaction with the mean density field occurs at a timescale t_m^* leading to $n_t \sim 1/Qt$ for $t_m^* \ll t \ll t_2^*$. (d) For both weak and strong systems, at sufficiently long times only of order one particle survives within a volume of size x_t^d, implying $n_t \approx 1/x_t^d$.

small compared to λ and can be neglected. For $t > t_2^*$, the solution is obtained by balancing the integral term with λ. One finds

$$k_t \approx \begin{cases} \lambda & (t \ll t_2^*) \\ x_t^d/t & (t_2^* \ll t) \end{cases} \quad (d < z), \qquad k_t \approx \begin{cases} \lambda & (t \ll t_2^*) \\ a^d/[t_a \ln(t/t_a)] & (t_2^* \ll t) \end{cases} \quad (d = z), \quad (7)$$

as can be verified by direct substitution in eq. (6). Substituting k_t in $\dot{n}_t = -k_t n_t^2$, it is easy to show that for low dimensions two possible sequences of reaction kinetics may occur, depending on the magnitude of Q:

$$n_t \approx \begin{cases} n_0 - \lambda n_0^2 t \xrightarrow{t_2^*} n_0 - n_0^2 x_t^d \xrightarrow{t_l} 1/x_t^d & (Q > Q^*, \text{ strong}) \\ n_0 - \lambda n_0^2 t \xrightarrow{t_m^*} 1/\lambda t \xrightarrow{t_2^*} 1/x_t^d & (Q < Q^*, \text{ weak}) \end{cases} \quad (d < z), \qquad (8)$$

where

$$\frac{t_2^*}{t_a} \equiv (Qt_a)^{z/(d-z)}, \quad \frac{t_l}{t_a} \equiv (n_0 a^d)^{-z/d}, \quad t_m^* \equiv \frac{1}{\lambda n_0}, \quad Q^* t_a \equiv (n_0 a^d)^{(z-d)/d}. \qquad (9)$$

For the marginal case $(d = z)$ one simply replaces x_t^d in eq. (8) by $a^d(t/t_a)/\ln(t/t_a)$. The timescales t_2^* and t_l are then modified to $t_2^* \equiv t_a e^{1/(Qt_a)}$, $t_l \equiv t_a(n_0 a^d)^{-1} \ln[1/(n_0 a^d)]$, while $Q^* t_a \equiv 1/\ln(t_l/t_a)$.

These results for $d < z$ have a simple physical interpretation (see fig. 1). Now if one were to assume MF theory were valid, *i.e.* that particles are distributed at all times as in equilibrium and hence $\dot{n}_t = -\lambda n_t^2$, the density would decay as $n_0 - \lambda n_0^2 t$ initially, crossing over to $1/\lambda t$ for $t \gg t_m^*$. Thus t_m^* would be the timescale for a particle to react with the mean reaction field supplied by the other reactants. For dimensions smaller than the critical value $d_c = z$, however, reactions induce nonequilibrium correlations and MF kinetics break down beyond a certain timescale. Consider a pair which happened to be initially within diffusive range, *i.e.* within x_t, as in fig. 1(a). Since each reactant explores a volume of order x_t^d approximately uniformly, the number of collisions $\mathcal{N}_{\text{coll}}$ after t/t_a "steps" increases roughly as $(t/t_a)(a^d/x_t^d) \sim t^{1-d/z}$.

Thus for $d < d_c$ the net reaction probability $Q t_a \mathcal{N}_{coll}$ is an increasing function of time which becomes of order unity at the timescale t_2^* of eq. (9). For $t > t_2^*$ a depletion hole of size x_t thus develops in the 2-body correlation function, invalidating MF theory.

For $d < d_c$, the sequence of kinetic regimes after t_2^* depends on the relative magnitudes of t_2^* and t_m^*. Another relevant timescale is t_l, namely the time to diffuse a distance of order the typical initial particle separation. One can easily show that the magnitude of t_m^* always lies in between those of t_2^* and t_l. The condition $t_2^* = t_m^* = t_l$ defines the critical reactivity Q^* in eq. (9). Case 1: Strongly reactive species, $Q > Q^*$ (or equivalently $t_2^* < t_m^* < t_l$). For $t_2^* < t < t_l$ reactions are due to the few isolated pairs which happened to be initially within diffusive range (fig. 1(a)). Since most of these pairs will have reacted by t, the number of reactions per unit volume during this time regime is proportional to the number of pairs per unit volume initially within x_t, $n_0^2 x_t^d$. Now for times $t > t_l$, a region of volume x_t^d contained initially several particles. Since only of order one of these particles could have survived by t, this implies a $1/x_t^d$ density decay. Notice that t_m^* is irrelevant in strong systems. Case 2: Weakly reactive species, $Q < Q^*$ (or $t_l < t_m^* < t_2^*$). Now when nonequilibrium correlations develop after t_2^* the $1/\lambda t$ MF kinetics have already developed. Since $t_2^* > t_l$, the same reasoning as in the strong case for $t > t_l$ implies that a direct crossover occurs after t_2^* to $1/x_t^d$ decay.

To summarize, we argued that the time-dependent rate constant k_t in single-species reaction kinetics is given by a Smoluchowski-type expression (eq. (6)). Equivalent expressions have been the starting point of previous works [25, 26, 22]. Here we have justified eq. (6) based on simple bounds on correlation functions. We argued that the solution for the rate constant below the critical dimension will then be correct at short times, but there is evidence that at long times (during the $1/x_t^d$ density regime) the actual numerical prefactor will be larger than that predicted by eq. (6). Indeed, for infinitely reactive particles in one dimension for $z = 2$, Torney and McConnell [8] have proved that k_t as determined by eq. (6) is correct at short times, but is smaller by a factor of $\pi/2$ from its actual value at long times.

Our result for the density decay (eq. (8)) does not give evidence for a Q-dependent power law decay at intermediate times as suggested in refs. [15-17] for $z = 2$. We suggest these workers may have been observing intermediate kinetics between the $1/t$ and $1/t^{d/2}$ regimes of eq. (8).

We remark that the results here have been for general diffusion dynamics described by the dynamical exponent z. They apply for both subdiffusion ($z > 2$) and superdiffusion ($z < 2$) provided that reactants diffuse independently of one another, and that x_t and mean collision time t_a are well defined. Implicitly, we have assumed that all moments of displacement are characterized by the same scale, x_t. Systems in which reactants perform Lévy walks, for which the rms displacement does not exist [27, 28], are not covered. The results do not apply also to systems in which reactants perform random walks for which the probability distribution for the time between successive steps has a power law long-time tail and hence t_a does not exist [29, 30]. In order to describe systems of the latter type, eq. (1) should be modified to include an extra integration over all possible collision times.

To specialize to the most widely studied case of small Fickian molecules one sets $z = 2$. For example, at the critical dimension $d = 2$ we predict a short-time diffusion-controlled regime for high reactivity ($Q > Q^*$) in which $n_0 - n_t \sim n_0^2 t / \ln t$, and we predict that in all cases the long-time decay is $n_t \sim \ln t / t$.

Finally, it is worth discussing how the above results may generalize to the two-species reaction problem, $A + B \rightarrow \emptyset$, for which segregation of reactants into A-rich and B-rich regions occurs at long times in dimensions $d < 2z$. This leads to an asymptotic decay $n_t \approx (n_0 / x_t^d)^{1/2}$ [2, 3]. For $d < z$, assuming that these kinetics apply for times longer than a timescale t_s, we may estimate the magnitude of t_s by assuming that eq. (8) applies for $t < t_s$

and demanding continuity in n_t. Thus one finds $t_s \approx t_l$ for strong systems, while for weak systems $t_s \approx t_a [n_0 a^d (Q t_a)^2]^{z/(d-2z)}$ which satisfies $t_m^* < t_s < t_2^*$. This would suggest that at least for weak systems the $1/x_t^d$ regime does not arise in $A + B \to \emptyset$. Indeed, this is consistent with the numerical simulations of ref. [2], where no $1/x_t^d$ regime was found for the case $z = 2$, $d = 1$. These workers observed that for sufficiently short times the density remained close to n_0. For high Q values a crossover to $t^{-1/4}$ occurred at t_l. For small Q, they found that the $t^{-1/4}$ regime was preceded by a $1/t$ regime during $t_m^* < t < t_s$. (We interpret the onset time here to be t_m^*, not $t_l/(Q t_a)$ as interpreted by the authors of ref. [2]. The numerical values of these two timescales happen to be very close to one another in this simulation.) We hope future numerical simulations as well as experiments on reacting polymers where novel values of z are realized will test the validity of the above theoretical predictions.

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