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Influence of external steady source structure on particle distributions and kinetics of diffusion-limited reactions. II. A+B→0 simulations

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Monte Carlo simulations were performed to study the effect of a steady external source structure (i.e., particle correlation and vertical reactions) on diffusion-limited A+B→0 reactions at steady state. Several methods were developed to describe the spatial organization of the system. They are the distributions of aggregates and inter-particle distance ("gap"), and a parameter based on the number of boundaries between A-rich and B-rich domains. The correlation of the particles in the steady external source reduces the local fluctuation in the particle landing process. The vertical reaction restrains the organization of particles, and does not allow it to reach total segregation. The degree of segregation affects the steady state kinetic behavior. The simulation results are consistent with existing theoretical predictions for the reaction order, the correlation length, and the segregation size.

1. Introduction

In a previous paper [1], we have shown how correlation in a steady external source affects the pattern of particle distribution and the reaction rate in the case of A+A→0 reactions. Particle self-organization was observed, and a depletion zone was characterized, based on the inter-particle distribution (IPDD). It was found that the depletion zone and the reaction order increase with the pair correlation length, δ, in the external source. Thus higher correlation in the source (smaller δ) led to a less correlated (or more random) particle distribution at steady state.

The particle self-organization in A+B→0 reactions is described not only in terms of depletion zone, but also the segregation (i.e., A-rich and B-rich domains). Segregation, of course, reduces the rate of reaction. The effects of segregation on the macroscopic kinetic rate law have attracted much theoretical interest for both reactions with and without the steady external source [2–8]. The theoretical predictions [7,8] show that the structure of the external source, such as particle correlation and vertical reactions, plays an important role in the formation of segregation, and affects the macroscopic rate law for A+B→0 reactions.

In this report, we demonstrate by Monte Carlo simulations the effect of the steady external source on the segregation and the kinetics of diffusion-limited A+B→0 reactions occurring on one-dimensional lattices. To characterize spatial distributions of particles, the aggregation distributions and the inter-particle distance distributions (IPDD) are obtained. Parameters are developed to quantify the degree of segregation. The simulation results are compared with theory.

2. Method of computation

Our simulations were carried out on a one-dimensional lattice with a cyclic boundary condition. Only one particle is allowed to occupy one site (i.e., the excluded volume effect). A+B→0 reactions are simulated by removing two particles of opposite species from the system when they occupy the same site.

The simulation methods regarding particle landing and diffusion have been discussed in detail in ref. [1]. For clarity, we briefly review here the particle landing processes. Two components in a landing process were considered: the reaction during the landing and the spatial distribution. When a landing particle hits

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the site occupied by a particle of the same species, this landing particle tries again to land at another site. If a particle lands at a site occupied by a particle of the opposite species, either the particles react, or the landing particle tries to land at another site again immediately. The first case is called *landing without vertical reaction*, and the second case is called *landing with vertical reaction*. "Correlation" in the external source means a spatial relationship between the two particles of the same pair, such as geminate (the correlation length $\delta=0$), and correlated landing ($1\leqslant\delta<\infty$). A random landing represents the case of particles in the external source without correlation (i.e., $\delta\to\infty$). Since the particle distribution and kinetics at $\delta=1$ are hardly different from the case of geminate landing, the simulation results for $\delta=1$ can be extended to the geminate landing case.

3. Characterization of segregation

In A+B→0 reactions, there are two kinds of interparticle distances distributions, those between particles of the same species (IPDDAA an IPDDBB) and that for opposite species (IPDDAB). The IPDDAB is closely related to the reaction kinetics. The interparticle distance between the opposite species indicates the size of depletion zone of "reactive" particles since reactions only occur when this distance is less than 1. The number of A–B gaps is also called the number of boundaries, N_b, between aggregates (i.e., A-rich and B-rich domains). N_b can be used to indicate both the number of possible reactions in the system and the degree of segregation [9]. The number of aggregates is equal to N_b for a one-dimensional lattice with cyclic boundary condition. To construct segregation parameters for various kinds of lattices, N_b is normalized by the total number of particles, N,

$$S=(N-N_b)/(N-2)\,, \tag{1}$$

where $N_b=2$, and $S=1$ for total segregation, $S\approx 0.5$ for a random distribution (obtained from simulations), and $N_b=N$, and $S=0$ for exact alternation of A and B particles.

An aggregate mass (m), defined as the number of continuous particles of the same kind, is also associated with segregation. For systems with a finite number of particles, the number of aggregates (N_{agg}) with mass smaller than or equal to a cut-off mass ($M_{agg}\equiv x$) is given in fig. 1 for three relevant distributions. Here, N_{agg} is normalized by the total number of particles (N), giving $N_{agg}/N\equiv f(x)$.

For total segregation,

$$\begin{aligned} f(x)&=0, \qquad 1\leqslant x<N/2,\\ &=2/N, \quad N/2\leqslant x\leqslant N. \end{aligned} \tag{2}$$

For a random distribution,

$$f(x)=\sum_{m=1}^{x}(\tfrac{1}{2})^{m+1}, \quad 1\leqslant x\leqslant N. \tag{3}$$

For exact alternation of A and B particles,

$$f(x)=1, \quad 1\leqslant x\leqslant N. \tag{4}$$

The extent of segregation is given by a "characteristic" M_{agg} defined to be an M_{agg} at which $f(x)$ has reached at least 99% of its highest value. The characteristic M_{agg} values are $N/2$ for the total segregated case, around 7 for the random distribution, and 1 for exact alternation of A and B particles, respectively. Eq. (3) was checked by simulations, and is independent of the particle density.

The particle landing process creates small aggregates at steady state, which make characterization of segregated particle distribution difficult to compare with theory. Thus, a "filter" is constructed to filter out small aggregates and then examine the aggrega-

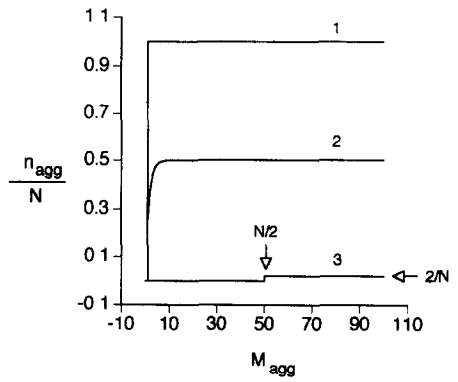

Fig. 1. The function $f(x)=M_{agg}/N$, given by the number of aggregates (n_{agg}) with the mass smaller than a cut-off aggregate mass ($x=M_{agg}$). n_{agg} is normalized by the total number of particles (N). Three distributions are given: (1) for exact alternation of A and B particles; (2) for a random distribution; and (3) for total segregation.

tion distribution. The width of a filter is an aggregation mass. Fig. 2 shows the normalized number of aggregates after filtering out the mass of aggregate smaller than or equal to the width of the filter for three defined cases. The data for random distribution were obtained from simulation. Again, the characteristic M_{agg} values may indicate the extent of segregation. For the random distribution, the characteristic M_{agg} does not depend on the particle density. The advantage of this method over the previous aggregation distribution is that a cut-off aggregation mass can be set for a segregated steady state particle distribution. Since the aggregation smaller than the cut-off value may be considered as "noise" created by the landing particles, the particle distribution may be examined only after the cut-off in aggregation mass.

4. Results

4.1. Kinetics

For steady state reactions on one-dimensional lattices, Clément et al. [7] have solved the master equations for a system with hard-core interactions, and obtained particle spatial correlation, and macroscopic rate laws at low density, under some limiting conditions.

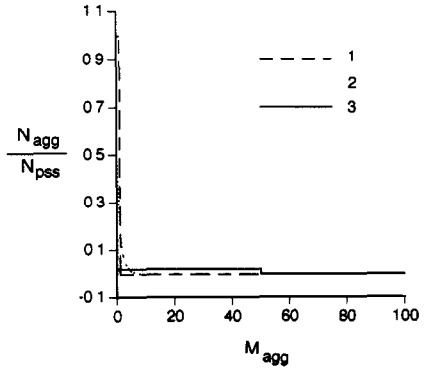

Fig. 2. The number of aggregates (N_{agg}) *after filtering out* the aggregates smaller than a cut-off aggregate mass (M_{agg}), normalized by the total number of particles (N). As in fig. 1, the three distributions are: (1) for exact alternation of A and B particles; (2) for a random distribution, obtained from simulation ($L=100\,000$, $N=100$); and (3) for total segregation.

For processes without vertical reaction in the random landing case

$$R=(12D/L)\rho_{ss}^2\,, \tag{5}$$

while in the correlated landing case

$$R=(2D/\delta)\rho_{ss}^2, \quad \delta/L\ll 1\,, \tag{6}$$

where R is the number of particles created per lattice site per each time step, D is the diffusion constant, L is the size of the lattice, and ρ_{ss} is the density at the steady state.

For random landing with vertical reactions,

$$Q=R(1-\tfrac{1}{2}\rho_{ss})^2=\frac{2D}{2+\xi_\gamma}\rho^2, \quad 1<\xi_\gamma\ll L\,, \tag{7}$$

where Q is an effective rate of particle creation on the lattice, and ξ_γ the correlation length, defined as

$$\xi_\gamma=\sqrt{D/R}\,. \tag{8}$$

Eq. (7) can be simplified to

$$R=4D\rho_{ss}^4, \quad 1\ll\xi_\gamma\ll L\,. \tag{9}$$

The correlation and segregation lengths for the specific conditions are listed in table 1.

Eqs. (5), (6) and (9) can be combined to a general form,

$$R=k\rho_{ss}^X\,, \tag{10}$$

where k is the rate constant and X is the effective order of reaction.

The comparison of simulation results with theory is shown in fig. 3. For the correlated landing at $\delta=1$ and the random landing, the rate constants from the simulations are very close to the theoretical predictions. For the other cases, the rate constants are higher than those predicted, and the deviation usually in-

Table 1
The theoretical prediction [7] of the correlation length and segregation size for A+B→0 reactions on one-dimensional systems

Landing conditions	correlation length	segregation size
random landing without vertical reaction	$\approx L$	$L/2$
correlated landing without vertical reaction	δ	δ
random landing with vertical reaction	$\approx\xi_\gamma$	$\xi_\gamma=\sqrt{D/R}$

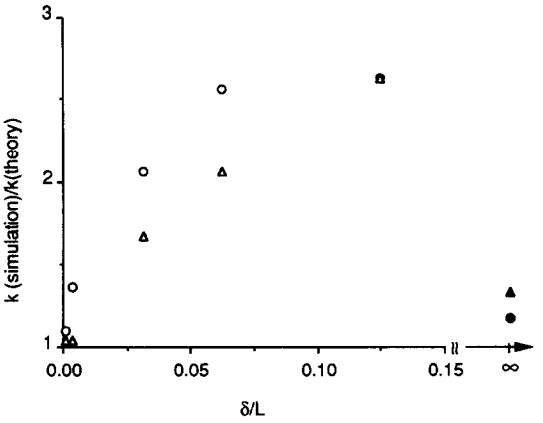

Fig. 3. Comparison of the predicted rate constants from theory with the simulation results on a one-dimensional lattice ($L=128$) without vertical reactions. The triangles are for $R=7.8\times10^{-4}$; the circles are for $R=3.9\times10^{-3}$. The solid symbols are for the random landing.

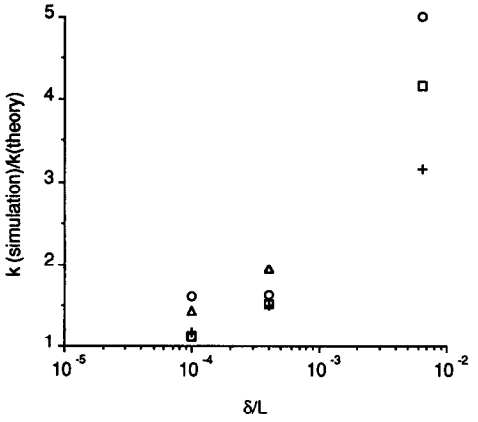

Fig. 4. Comparison of the predicted rate constants from theory with the simulation results on a one-dimensional lattice ($L=10\,000$) with vertical reactions. (+) $R=2\times10^{-4}$; (□) $R=4\times10^{-4}$; (○) $R=1.6\times10^{-3}$; (△) $R=3.2\times10^{-3}$.

creases with R. Without the vertical reaction, the ratio of k from simulation to k from theory increases with δ. The large ratio (>2) occurs at $\delta/L>0.03$ for $R=3.9\times10^{-3}$. For the vertical reaction (fig. 4), eq. (6) holds for $\delta=1$, but not for the large values of δ/L (>0.0005), where the ratio of k from simulations to k from theory is greater than 2 for $R=3.2\times10^{-3}$. These deviations are expected since eq. (6) is only derived when $\delta/L\ll1$ for the case of the vertical reactions. The magnitudes of the differences between the reactions with and without the vertical reaction indicate the effect of vertical reaction on the kinetics in the correlated landing process.

The vertical reaction effect can also be found from the random landing cases and can be seen in eqs. (5) and (9). The reaction order is 2 without the vertical reaction, and is 4 with the vertical reaction. Thus, the reaction rate, which is equal to the rate of particle landing at steady state, has higher dependence on the particle density for the random landing with vertical reaction. These predictions have already been confirmed by the simulations [10–12]. Here we extend the calculation of the reaction order to the correlated landing (fig. 5). Without the vertical annihilation, the reaction order X is about equal to 2 when the values of δ/L are very small or very large. With the vertical annihilation, X increases from 2 to 4 as the correlated landing changes from 1 to ∞ (the random landing).

4.2. Inter-particle distance distribution

The IPDDAA, IPDDBB and IPDDAB were examined (fig. 6). Since particles of the same species do not react, they may be "trapped" in an A-rich or a B-rich domain. The IPDDAA and the IPDDBB are determined by diffusion and collision. Thus, the normalized IPDDAA and IPDDBB are independent of the landing process, maintaining an approximately

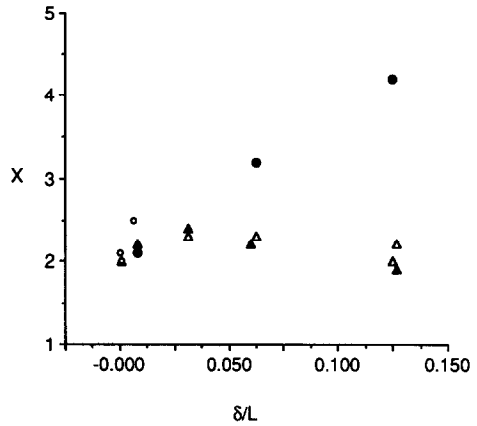

Fig. 5. The reaction order X as a function of the correlation length δ in the external source. (△) $L=1024$, $R=9.8\times10^{-5}$ and without vertical reaction; (▲) $L=128$, $R=1.6\times10^{-2}$, and without vertical reaction; (○) $L=10\,000$, $R=2\times10^{-4}$, and with vertical reaction; (●) $L=128$, $R=1.6\times10^{-2}$, and with vertical reaction.

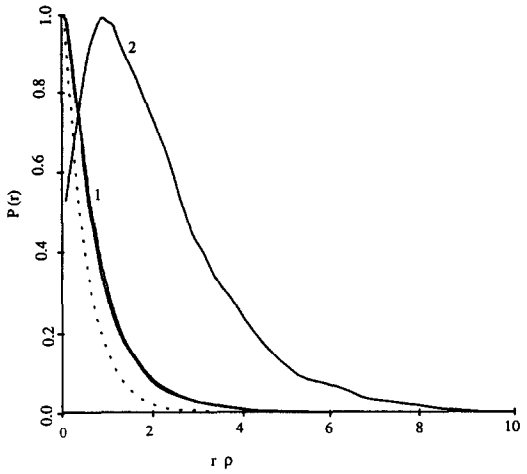

Fig. 6. The normalized inter-particle distance distributions for random landing ($R=9.8\times10^{-5}$) without vertical reaction at steady state on a one-dimensional lattice ($L=1024$ sites). (1) IPDDAA and IPDDBB are superimposed; (2) IPDDAB.

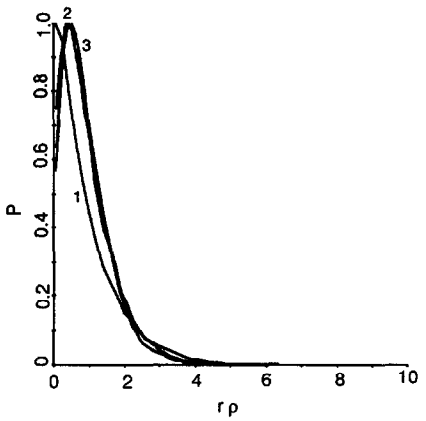

Fig. 7. The normalized inter-particle distance distribution of A–B pairs for steady state reactions with correlated landings and random landing on a one-dimensional lattice ($L=1024$ sites). (1) $\delta=1$; (2) $\delta=32$; (3) $\delta=64$ and random landing are superimposed.

exponential type of distribution with the most probable inter-particle distance being 1. However, the IPDDAB is affected significantly by the particle landing process (fig. 7). We observe a crossover from an exponential type distribution for $\delta=1$ to a skewed exponential type for the random landing. This crossover is similar to that in the A+A→0 reaction [1]. Furthermore, the normalized IPDDAB is comparable to the normalized IPDD in the A+A→0 reaction (fig. 8) for the random landing. It is interesting to notice that the behaviors of IPDDAA, IPDDAB, and IPDDAB for the random landing at steady state are similar to the asymptotic behavior of these distributions found in the A+B→0 batch reaction [13,14]. We also note that the nearest neighbor distributions of A+A→0 and A+B→0 batch reactions are also quite similar [13].

4.3. Segregation

The steady state of segregation can be reached (fig. 9) when the particle density reaches the steady state [9]. The effects of the particle landing process on the segregation at steady state can be divided into three aspects: (1) the vertical versus non-vertical reaction, (2) the source correlation length, and (3) the landing rate. Our simulation results show that having vertical reaction reduces the degree of segregation in the cases of the correlated landing with large δ and random landing (figs. 10 and 11).

The effect of the source correlation length on the segregation is demonstrated using the segregation parameter in figs. 9 and 10. For $\delta=1$, $S\approx0.5$, i.e., a random distribution is obtained. When $1<\delta\ll L/6$, S increases quickly with δ. When $L/6<\delta<L/2$, S increases slowly. The top three lines in fig. 9, corre-

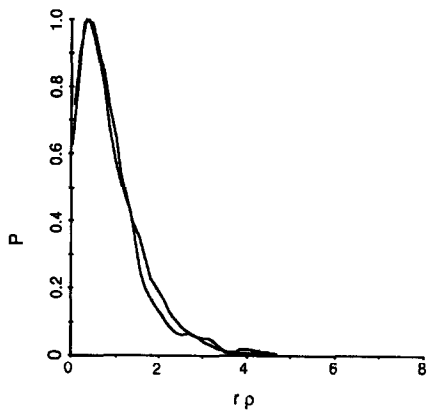

Fig. 8. The inter-particle distance distributions for steady state reactions with random landing on a one-dimensional lattice. The IPDD for A+A→0 and the IPDDAB for A+B→0 are superimposed at steady state. They are indistinguishable within the simulation uncertainties.

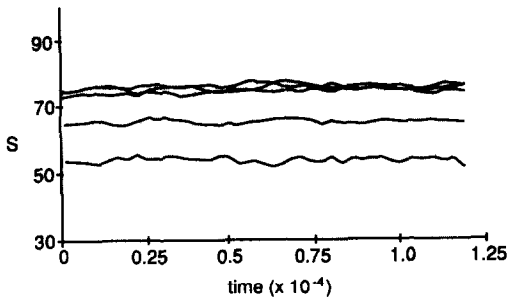

Fig. 9. The segregation parameter (eq. (1)) as a function of time on a one-dimensional lattice ($L=128$, $R=1.6\times10^{-2}$) with vertical reaction. From top to bottom, the lines are random landing, $\delta=64$, 32, 8, and 1.

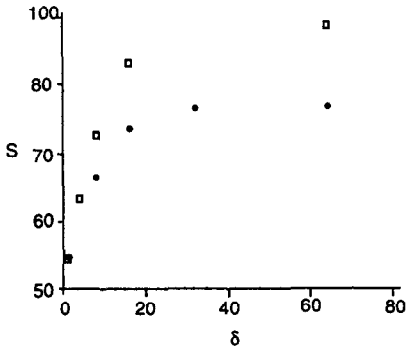

Fig. 10. The segregation parameter (eq. (1)) as a function of the correlated length δ in the external source on a one-dimensional lattice ($L=128$ sites, $R=1.6\times10^{-2}$) with (●) and without (□) vertical reaction.

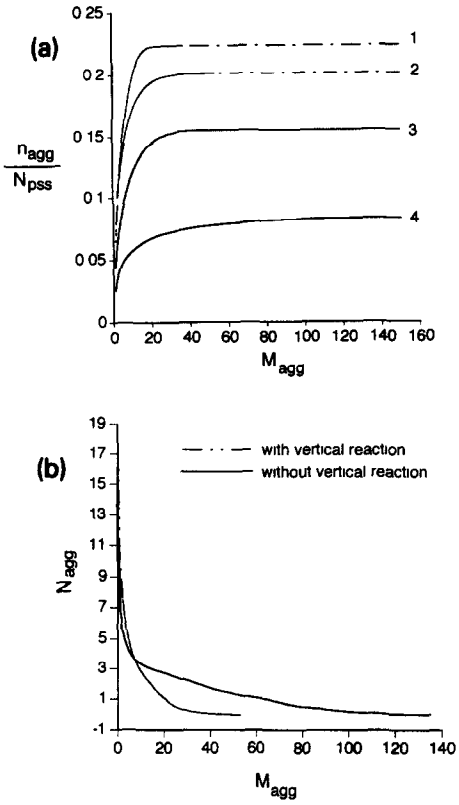

Fig. 11. The vertical reaction effect on the segregated particle distribution at steady state. (a) The normalized number of aggregates (n_{agg}) with the mass smaller than a cut-off value (M_{agg}). N_{pss} is the number of particles at steady state. From top to bottom, (1) $\delta=64$ *with* vertical reaction; (2) random landing *with* vertical reaction; (3) $\delta=64$ *without* vertical reaction; (4) random landing *without* vertical reaction. (b) The non-normalized number of aggregates (N_{agg}) *after filtering out* the aggregates smaller than a cut-off value (M_{agg}). The data were obtained from averaging over 506 realizations at steady state (from step 404 800 to 809 600) on a one-dimensional lattice *without vertical reaction* ($L=1024$, $R=4.9\times10^{-4}$).

sponding to $L/\delta=4$, $L/\delta=2$, and the random landing, are very close to each other. Particles are almost totally segregated at steady state for the random landing without the vertical reaction (fig. 10).

A similar conclusion can be reached from examining the aggregation distributions (figs. 12 and 13). The charateristic M_{agg} values increases with δ, and fall into the range of 7 to $N/2$. It is noticed that there are more small aggregations in the $\delta=1$ case (figs. 12 and 13) comparing to a true random distribution (figs. 1 and 2). For the random landing at steady state, a sharp transition was observed around $M_{agg}=5$ in fig. 13. Thus, the systems are examined for $M_{agg}>5$. Comparing to fig. 2, the particle distribution is close to a totally segregated distribution. As shown here, the aggregate distributions may provide more details on the particle distributions.

The effect of the particle landing rate on the segregation is described in figs. 14 and 15. Without vertical reaction (fig. 14), the degree of segregation remains almost constant as R changes both for the random landing and the correlated landing for $\delta=1$. However, when $1<\delta<\infty$, the segregation increases with R. With vertical reaction (fig. 15), in the case of random landing, the degree of segregation de-

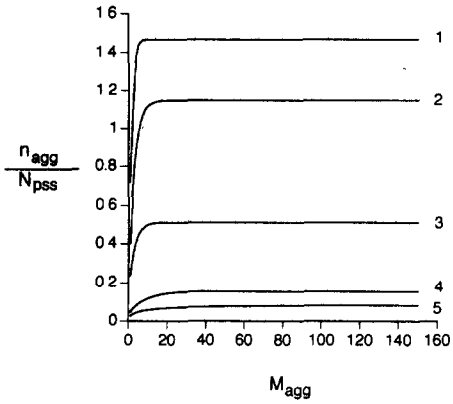

Fig. 12. The normalized number of aggregates (n_{agg}) with the mass smaller than a cut-off value (M_{agg}). N_{pss} is the number of particles at steady state. The data were obtained from averaging over 506 realizations at steady state (from step 404 800 to 809 600) on a one-dimensional lattice *without vertical reaction* ($L=1024$, $R=4.9\times10^{-4}$). The correlated landing length (δ) is: (1) 1; (2) 16; (3) 32; (4) 64. The curve (5) is for random landing ($\delta\to\infty$).

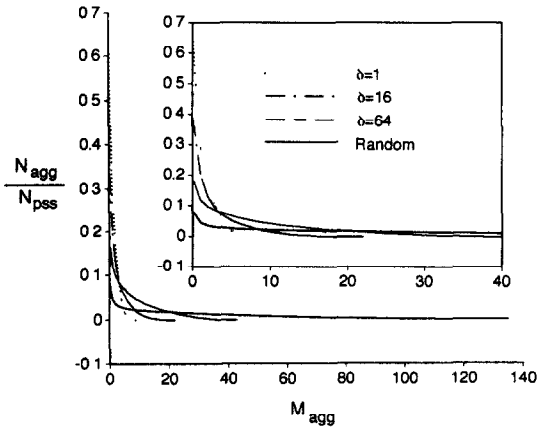

Fig. 13. The normalized number of aggregates (N_{agg}) *after filtering out* the aggregates smaller than a cut-off value (M_{agg}). N_{pss} is the number of particles at steady state. The data were obtained from averaging over 506 realizations at steady state (from step 404 800 to 809 600) on a one-dimensional lattice *without vertical reaction* ($L=1024$, $R=4.9\times10^{-4}$). The correlated landing length (δ) is: (1) 1; (2) 16; (3) 32; (4) 64. The curve (5) is for random landing ($\delta\to\infty$).

creases as R increases, which is consistent with eq. (7). In the cases of correlated landing, the segregation does not significantly change when R increases.

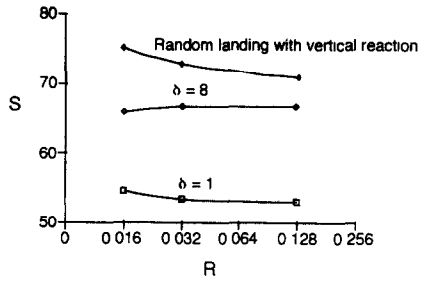

Fig. 14. The segregation parameter (eq. (1)) as a function of the landing rate R at steady state on a one-dimensional lattice ($L=128$ sites) with vertical reaction.

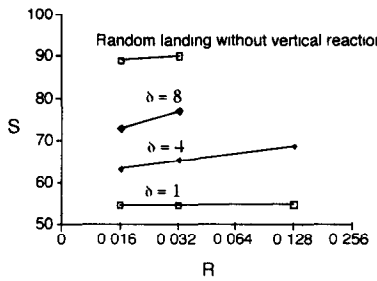

Fig. 15. The segregation parameter (eq. (1)) as a function of the landing rate R at steady state on a one-dimensional lattice ($L=128$ sites) without vertical reaction.

5. Discussion

In the presence of the steady state source, the normalized IPDD in A+A→0 reactions is similar to the normalized IPDDAB in A+B→0 reactions. Although the size of the normalized depletion zone is about the same in both types of reactions, the definition of reaction zone is very different. The number of reaction zones is equal to the number of the particles in the A+A→0 reaction, but is equal to the number of aggregates in the A+B→0 reaction. For complete segregation in the A+B→0 reaction, the number of reaction zones is about 2, which is independent of the density. In comparison with the A+A→0 reaction, the difference in reaction zones has brought a new set of kinetic equations for the A+B→0 reaction.

The number of aggregates, or the segregation, affects the macroscopic rate law. It is known that local fluctuations initiate segregation. The magnitude of a local fluctuation [5,6], $|N_A-N_B|/(N_A+N_B)$, is determined by the structures in the external source. The

fluctuation occurs when particles land randomly on a lattice. The correlation between the particles in the external source reduces the local fluctuation. At $\delta = 1$, there is not any local fluctuation, and the segregation does not occur. When δ is greater than $\langle r \rangle$ or r_{max} of IPDDAB (i.e., the average or the most probable distance between the domains) at steady state for the random landing, the correlation of the particles will not be able to reduce the fluctuation in a reaction domain. This may explain the observation that the segregation is greater for the higher landing rate since the value of $\delta/\langle r \rangle$ at steady state increases for a fixed δ without the vertical reaction (fig. 14).

In the landing process, the other important factor is the vertical reaction. An increase in the vertical reaction results in a decrease in the segregation. When a particle lands on an occupied site in an A-rich or a B-rich domain, if this site is occupied by a particle of the same species, the landing particle will be rejected and try to land again; if the particle on the occupied site is of the opposite species, this particle will be eliminated by reaction. In this way, the magnitude of fluctuations tends to decrease in that domain. As discussed in the case without vertical reaction, the segregation increases with R for a fixed δ. On the other hand, the segregation decreases due to the increase of the vertical reactions when R increases. Thus, the system is in a balance between the two processes, and no significant change is observed as R changes in the case with the vertical reaction (fig. 15).

Beside the vertical reaction, there are "horizontal" reactions in the system. In a vertical reaction, one of the reactive particles is a landing particle that has zero life-time on the lattice. In a horizontal reaction, both reactive particles have a life-time greater than zero on the lattice. In other words, reactive particles meet by diffusion. The fluctuation, $|N_A - N_B|/(N_A + N_B)$, in a domain is magnified by horizontal reactions since the $A + B \rightarrow 0$ reaction eliminates an equal amount of A and B particles in that domain. The domain then will expand. The horizontal reaction coupled with the particle diffusion promotes aggregates to grow to their maximum size. We have observed an almost total segregation for the random landing without vertical reaction, where there is no structure in the external source, such as the correlated landing and the vertical reaction, to disturb the expansion of the aggregates. The size of segregation is about the size of the lattice for random sources is also found theoretically for systems without hard-core repulsion on one-dimensional systems [5,15].

In $A + B \rightarrow 0$ reactions, the structures of the external source affect the kinetics not only for steady state reactions, but also for batch reactions. Theory [8] and simulations [16,17] have shown that the asymptotic kinetic behavior depends on the correlation length in the initial particle distribution. In contrast, the initial particle distribution does not influence the asymptotic kinetic behavior in $A + A \rightarrow 0$ batch reactions [16–18].

6. Summary

The steady external source influences the particle organization patters in the $A + B \rightarrow 0$ reaction. Although the behavior of the normalized IPDDAB in the $A + B \rightarrow 0$ reaction is similar to the normalized IPDD in the $A + A \rightarrow 0$ reaction, the segregation plays an important role in the determination of the kinetic behavior in the $A + B \rightarrow 0$ reaction.

The correlation of the particles in the external source reduces the local fluctuation in the particle landing process, and the degree of segregation. For the geminate reaction and the correlated landing at $\delta = 1$, particles are randomly distributed, and thus the classical kinetics holds at steady state. For the random landing without vertical reaction, almost total segregation is obtained. The reaction order is still 2, but the reaction rate is much slower than that in correlated landing process with small δ. The vertical reaction decreases the magnitude of the segregation, but increases the rate of reaction: for random landing with vertical reaction, the order of reaction is 4. We thus see that segregation and reaction orders are not simply related. Our simulation results are in good agreement with theory.

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