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# Exact solution for a steady-state aggregation model in one dimension 

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Received 9 August 1988


#### Abstract

The diffusion-limited steady-state particle coalescence model of aggregation is solved in one dimension. The method presented can also be used to solve the timedependent problem and is extensible to higher dimensions.


Meakin (1983) and independently Kolb et al (1983) introduced a computer simulation model, cluster-cluster aggregation (Clcl), to describe the flocculation of colloids. In this model $N$ particles are randomly placed on an $L^{d}$ lattice and started moving diffusively. When ever two particles occupy adjacent sites they stick irreversibly and the newly formed cluster of two particles continues diffusing. The same irreversible welding occurs whenever two particles in different clusters occupy adjacent sites and thus ever larger clusters are formed. The clusters formed by ClCl are stochastic fractals (Mandelbrot 1982) with a complicated structure.

Kang and Redner (1984) introduced a simplified version of ClCl , the particle coalescence model (PCM) in order to concentrate on the kinetic aspects of CICl. In the PCM clusters are defined to be single lattice sites which aggregate/coalesce whenever two or more clusters occupy the same lattice site. CICl and PCM are equivalent in one dimension provided the excluded volume effect in ClCl is properly taken into account.

Vicsek et al (1985) performed steady-state CICl simulations; single particles were injected into the aggregating system at a constant rate and large clusters were removed from it. This system was studied theoretically by Rácz (1985a) in a companion paper.

Computer simulations and scaling arguments were used by Family et al (1986) to study a steady-state PCM in which the bonds joining clusters together could break. This system has also been studied by Elderfield (1987) using a field theoretic approach. Recently Family and Meakin (1988) have studied a computer simulation model of droplet growth in one, two and three dimensions in which droplets are placed one-byone at random and overlapping droplets coalesce. In this model there is no droplet diffusion except as a consequence of coalescence.
$\mathrm{PCM} / \mathrm{ClCl}$ in one dimension has been solved by Spouge (1988) when all clusters diffuse at the same rate. This problem is tractable for two reasons. In one dimension any particle can only bond with its two neighbours; and since all clusters diffuse at the same rate any chosen particle undergoes a random walk independent of the other clusters in the system. Rácz (1985b) has solved the steady-state diffusion-limited annihilation problem (when two particles aggregate they disappear from the system) in one dimension and Torney and McConnel (1983) have solved the time-dependent version of the same problem. Spouge (1988) has shown how the solution of PCM in
one dimension solves a set of aggregation problems, PCM modulo $N$, in which an $i$ cluster and a $j$ cluster aggregate to form an $(i+j)$ modulo $N$ cluster. These models include time-dependent diffusion-limited annihilation in one dimension which is PCM modulo 2.

In this paper we solve the canonical steady-state $\operatorname{PCM}$ in one dimension for the cluster mass distribution. In this model single particles are fed into the system and no clusters are removed. The increase in the total cluster concentration is balanced by the continual coalescence of clusters. The concentration of clusters of any fixed mass reaches a steady state but the large-mass tail of the cluster mass distribution evolves forever.

We now precisely define our aggregation model and fix some notation. Consider a linear lattice on which point clusters hop randomly from bond to bond. When two clusters hop onto the same bond they aggregate; a cluster of $k$ particles (a $k$-mer) occupying one bond aggregates with a $p$-mer on the same bond to form a $(k+p)$-mer. The hopping rate of the $k$-mers is $2 \alpha$ hops per unit time independent of $k$. Thus in the time interval $\mathrm{d} t$ the probability that any given $k$-mer has hopped one bond to the left is $\alpha \mathrm{d} t$ and the probability that it has hopped one bond to the right is $\alpha \mathrm{d} t$. Monomers are added to the lattice at random at the rate $\beta$ per unit time per bond. This model generalises the diffusion-limited annihilation model solved by Rácz (1985b).

Consider a set of probabilities $P_{i j}^{N}$ where $P_{i j}^{N}$ is the probability that exactly $N$ particles occupy the bonds between the lattice points $i$ and $j(i<j)$. Also consider the sets $A_{i j}^{N}$ corresponding to the probabilities $P_{i j}^{N} . A_{i j}^{N}$ is the set of states of the aggregating system for which there are exactly $N$ particles in $(i, j)$. Thus $P_{i j}^{N}=P\left(A_{i j}^{N}\right)$ where $P$ is the probability measure on the states of the aggregating system.

There are several points to note.
(i) We write $(i, j)$ for the set of $j-i$ bonds between $i$ and $j$.
(ii) $N$ particles in ( $i, j$ ) may consist of any combination of $k$-mers with total mass $N$.
(iii) The boundary values of $P_{i j}^{N}$ are $P_{i i}^{0}=1$ and $P_{i i}^{N}=0, N>0$. This is because there are no bonds in (i,i) and hence there must always be exactly 0 particles in ( $i, i$ ). The other boundary condition on $P_{i j}^{N}$ is $P_{i j}^{N} \rightarrow 0$ as $j-i \rightarrow \infty, N \geqslant 0$.
(iv) $P_{i, i+1}^{k}$ is the probability of finding a $k$-mer in $(i, i+1)$. Coupled with an appeal to translational invariance this will give us the concentration of $k$-mers.

The system of equations satisfied by the $P_{i j}^{N}$ is easily derived. We shall first consider $P_{i j}^{0}$ and then the general case. Broadly the technique will be to find the set of states which can make the transition to a state in $A_{i j}^{N}$ as a result of only one hop of a $k$-mer and those states in $A_{i j}^{N}$ which can leave $A_{i j}^{N}$ with only one hop.

Consider the set of states $A_{i, j-1}^{0}-A_{i j}^{0}$. There are no particles in ( $i, j-1$ ) but not in $(i, j)$. Therefore there must be a $k$-mer on the bond $(j-1, j)$ and a transition to a state in $A_{i, j}^{0}$ will occur if that $k$-mer hops right. Since no particles in (i,j) implies that there are no particles in $(i, j-1)$ we have $A_{i, j-1}^{0} \supseteq A_{i j}^{0}$. Thus the probability of a $k$-mer occupying the bond ( $j-1, j$ ), $P\left(A_{i, j-1}^{0}-A_{i j}^{0}\right)$, is given by $P_{i, j-1}^{0}-P_{i j}^{0}$. Similarly the probability of a $k$-mer occupying bond $(i, i+1)$ is $P_{i+1, j}^{0}-P_{i j}^{0}$ and thus $T_{i j}^{0+}$, the transition rate into $A_{i j}^{0}$, is

$$
\begin{equation*}
T_{i j}^{0+}=\alpha\left(P_{i+1, j}^{0}+P_{i, j-1}^{0}-2 P_{i j}^{0}\right) . \tag{1}
\end{equation*}
$$

The transition rate from $A_{i j}^{0}, T_{i j}^{0-}$, can be derived in a similar way by considering $A_{i j}^{0}-A_{i, j+1}^{0}$ and $A_{i j}^{0}-A_{i-1, j}^{0}$ which correspond to states with no particles in (i,j) and a $k$-mer on the bond $(j, j+1)$ or $(i-1, i)$ respectively. Transitions from $A_{i j}^{0}$ occur when
the $k$-mer hops into $(i, j)$; they also occur when a monomer is added to one of the $j-i$ bonds in ( $i, j$ ). Upon putting the pieces together we find

$$
\begin{equation*}
T_{i j}^{0-}=\alpha\left(-P_{i-1, j}^{0}-P_{i, j+1}^{0}+2 P_{i j}^{0}\right)+\beta(j-i) P_{i j}^{0} . \tag{2}
\end{equation*}
$$

We now write down $(\mathrm{d} / \mathrm{d} t) P_{i j}^{0}=T_{i j}^{0+}-T_{i j}^{0-}$ :
$(\mathrm{d} / \mathrm{d} t) P_{i j}^{0}=\alpha\left(P_{i+1, j}^{0}+P_{i, j-1}^{0}-2 P_{i j}^{0}\right)+\alpha\left(P_{i-1, j}^{0}+P_{i, j+1}^{0}-2 P_{i j}^{0}\right)-\beta(j-i) P_{i j}^{0}$.
The system of equations satisfied by $P_{i j}^{N}$ where $N>0$ is slightly more complicated to derive. Consider the pairs of sets $A_{i, j-1}^{N}-A_{i j}^{N}, A_{i, j+1}^{N}-A_{i j}^{N}$, and $A_{i j}^{N}-A_{i, j-1}^{N}, A_{i j}^{N}-$ $A_{i, j+1}^{N}$. Transitions into and out of $A_{i j}^{N}$ occur from the first and second pair respectively. For example, $A_{i, j+1}^{N}-A_{i j}^{N}$ is the set of states with $N$ particles in ( $i, j+1$ ) but not in $(i, j)$. Thue a $k$-mer must occupy the bond $(j, j+1)$ and a transition into $A_{i j}^{N}$ will occur if that $k$-mer hops left onto the bond $(j-1, j)$ which is in $(i, j)$.

Ignoring for the moment the effects of the addition of monomers into the aggregating system $\quad T_{i j}^{N^{+}}=\alpha\left[P\left(A_{i, j-1}^{N}-A_{i j}^{N}\right)+P\left(A_{i, j+1}^{N}-A_{i j}^{N}\right)\right] \quad$ and $\quad T_{i j}^{N-}=\alpha\left[P\left(A_{i j}^{N}-A_{i, j-1}^{N}\right)+\right.$ $\left.P\left(A_{i j}^{N}-A_{i, j+1}^{N}\right)\right]$. Now by elementary set theory $A-B=A-A \cap B$ and thus $P(A-B)=$ $P(A-A \cap B)=P(A)-P(A \cap B)$. Using this result we find that $T_{i j}^{N^{+}}+T_{i j}^{N-}$ has a remarkably simple form

$$
\begin{equation*}
T_{i j}^{N^{+}}+T_{i j}^{N-}=\alpha\left(P_{i+1, j}^{N}+P_{i-1, j}^{N}+P_{i, j-1}^{N}+P_{i, j+1}^{N}-4 P_{i j}^{N}\right) . \tag{4}
\end{equation*}
$$

We now return to the effect of the addition of monomers. Their random injection into the aggregating system causes transitions into $A_{i j}^{N}$ at the rate $\beta(j-i) P_{i j}^{N-1}$ and transitions out of $A_{i j}^{N}$ at the rate $\beta(j-i) P_{i j}^{N}$.

Collecting these two sets of results we find that
$(\mathrm{d} / \mathrm{d} t) P_{i j}^{N}=\alpha\left(P_{i+1, j}^{N}+P_{i-1, j}^{N}+P_{i, j-1}^{N}+P_{i, j+1}^{N}-4 P_{i j}^{N}\right)$

$$
\begin{equation*}
+\beta\left[(j-i) P_{i j}^{N-1}-(j-i) P_{i j}^{N}\right] . \tag{5}
\end{equation*}
$$

Since the evolution rules which define this model are translationally invariant if we assume a translationally invariant initial distribution it follows that $P_{i j}^{N}$ is only a function of $j-i$. Hence we define $P_{k}^{N}$ to be the probability that there is a total of $N$ particles in any randomly chosen $k$ consecutive bonds. $P_{k}^{N}$ satisfies the system of equations
$(\mathrm{d} / \mathrm{d} t) P_{k}^{0}=2 \alpha\left(P_{k-1}^{0}-2 P_{k}^{0}+P_{k+1}^{0}\right)-\beta k P_{k}^{0}$
$(\mathrm{d} / \mathrm{d} t) P_{k}^{N}=2 \alpha\left(P_{k-1}^{N}-2 P_{k}^{N}+P_{k+1}^{N}\right)+\beta k P_{k}^{N-1}-\beta k P_{k}^{N} \quad N>0$.
Equations (6) and (7) describe the aggregation of point clusters. In their derivation it is necessary for the cluster to occupy only one bond because of the following possibility. Consider the situation of $N$ particles in $(i, j+1)$ and but not $N$ in $(i, j)$. The extended $k$-mer partially situated on ( $j, j+1$ ) could occupy all of $(i, j+1)$. If it hopped left the number of particles in $(i, j)$ would remain constant rather than increase to $N$ as assumed in the derivation of equations (6) and (7). It is also necessary for the derivation that $k$-mers aggregate when they occupy the same bond because it is assumed that all of the particles on a bond hop together. If this were not the case the derivation would again fail in the situation of $N$ particles in $(i, j+1)$ and but not $N$ in $(i, j)$ because only $p$, say, of the $k$ particles on $(j, j+1)$ might hop left into $(i, j)$.

Equations (6) and (7) can also be derived indirectly by extending the method of Rácz (1985b) who mapped steady-state diffusion-limited annihilation onto the zerotemperature kinetic Ising model with Glauber dynamics. The present aggregation
model can be similarly mapped onto an infinite-state Potts model at zero temperature; however, the present direct method seems a more natural approach.

If no new monomers are added to the aggregation system, i.e. $\beta=0$, equations (6) and (7) describe standard time-dependent CICl on a line. This problem has been solved by Spouge (1988) using a different approach; in effect, he studied the evolution of the gaps between the pairs of monomers $i, j$ and $i, j+1$.

We shall solve for the steady state described by equations (6) and (7) in the continuous limit. The continuous limit is taken by letting the lattice spacing $\Delta \rightarrow 0$ whilst the diffusion constant for individual $k$-mers, $D=\alpha \Delta^{2}$, and the feed rate per unit distance, $\beta^{*}=\beta / \Delta$, remain constant. The steady-state continuous system equations are

$$
\begin{align*}
& 0=2 D\left(\mathrm{~d}^{2} / \mathrm{d} x^{2}\right) P^{0}(x)-\beta^{*} x P^{0}(x)  \tag{8}\\
& 0=2 D\left(\mathrm{~d}^{2} / \mathrm{d} x^{2}\right) P^{N}(x)+\beta^{*} x P^{N-1}(x)-\beta^{*} x P^{N}(x) \quad N>0 . \tag{9}
\end{align*}
$$

The boundary conditions are $P^{0}(0)=1, P^{N}(0)=0, N>0$ and $P^{N}(x) \rightarrow 0$ as $x \rightarrow \infty$, $N \geqslant 0$. The concentration of $k$-mers is simply $c_{k}=(\mathrm{d} / \mathrm{d} x) P^{N}(0), N \geqslant 0$.

We have assumed that the aggregating system reaches a steady state in which $P^{N}(x)$ is constant; this is intuitively clear for any fixed $N$ but a rigorous proof remains a topic for further research. A hand-waving argument that the concentration $c_{N}$ reaches a steady-state value is based on induction. Assume $c_{k}$ reaches a steady-state value for $k<N$. Thus the concentration of the $k$-mers which can aggregate to form an $N$-mer is stable. It is assumed that the total number of clusters reaches a steady-state value with the influx of single clusters balanced by the coalescence of clusters. Thus the concentration of clusters with which an $N$-mer can aggregate and so reduce $c_{N}$ is constant. Since the sources and sinks for $N$-mers are stable $c_{N}$ must reach a constant value. Thus we have (in a hand-waving fashion) reduced the problem to showing that the total number of clusters reaches a steady-state value which can be shown by solving directly

$$
\begin{equation*}
\partial P^{0} / \partial t=2 D\left(\partial^{2} / \partial x^{2}\right) P^{0}(x)-\beta^{*} x P^{0}(x) . \tag{10}
\end{equation*}
$$

We now simplify equations (8) and (9) by changing variables. Define $R^{N}$ by $R^{N}(s x)=$ $P^{N}(x)$ where $s=\left(\beta^{*} / 2 D\right)^{1 / 3} . R^{N}$ satisfies

$$
\begin{align*}
& \left(\mathrm{d}^{2} / \mathrm{d} x^{2}\right) R^{0}(x)=x R^{0}(x)  \tag{11}\\
& \left(\mathrm{d}^{2} / \mathrm{d} x^{2}\right) R^{N}(x)=x R^{N}(x)-x R^{N-1}(x) \quad N>0 . \tag{12}
\end{align*}
$$

These equations can be solved in turn for $N=0, N=1, \ldots$ using Fourier transform techniques. We shall solve them for $N=0,1,2$ and implicitly establish some notation before formally introducing it. We shall then proceed to the general solution.

For $N=0$ the Fourier transform of equation (11) is

$$
\begin{equation*}
-k^{2} \tilde{R}^{0}=\mathrm{i}(\mathrm{~d} / \mathrm{d} k) \tilde{R}^{0} \tag{13}
\end{equation*}
$$

where we define the Fourier transform of $f(x)$ to be $\tilde{f}(k)=\int f(x) \exp (-\mathrm{i} k x) \mathrm{d} x$. Thus

$$
\begin{align*}
& \tilde{R}^{0}(k)=A_{0}^{0} \exp \left(\frac{1}{3} \mathrm{i} k^{3}\right)  \tag{14}\\
& R^{0}(x)=A_{0}^{0} \mathrm{Ai}(x) \tag{15}
\end{align*}
$$

where $\mathrm{Ai}(x)$ is the Airy function (Abramowitz and Stegun 1970) and $A_{0}^{0}=\mathrm{Ai}(0)^{-1}$.
For $N=1$ the Fourier transform of equation (12) is

$$
\begin{equation*}
-k^{2} \tilde{R}^{1}(k)=\mathrm{i}(\mathrm{~d} / \mathrm{d} k) \tilde{R}^{1}(k)-\mathrm{i}(\mathrm{~d} / \mathrm{d} k) \tilde{R}^{0}(k) \tag{16}
\end{equation*}
$$

which has the solution

$$
\begin{equation*}
\tilde{R}^{1}(k)=\left(A_{1}^{1}(\mathrm{i} k)^{3}+A_{0}^{1}\right) \exp \left(\frac{1}{3} 1 k^{3}\right) \tag{17}
\end{equation*}
$$

where $A_{1}^{1}=-\frac{1}{3} A_{0}^{0}$ and $A_{0}^{1}$ is determined by the normalisation constraint $R^{1}(0)=0$.
Similarly for $N=2$

$$
\begin{equation*}
\tilde{R}^{2}(k)=\left(\dot{A}_{2}^{2}(\mathrm{i} k)^{6}+A_{1}^{2}(\mathrm{i} k)^{3}+A_{0}^{2}\right) \exp \left(\frac{1}{3} \mathrm{i} k^{3}\right) \tag{18}
\end{equation*}
$$

where $A_{2}^{2}=-\frac{1}{6} A_{1}^{1}, A_{1}^{2}=A_{1}^{1}-\frac{1}{3} A_{0}^{1}$ and $A_{0}^{2}$ is determined by the normalisation constraint $R^{2}(0)=0$. The inverse transforms of $\tilde{R}^{1}(k)$ and $\tilde{R}^{2}(k)$ are

$$
\begin{align*}
& R^{1}(x)=\left[A_{1}^{1}(\mathrm{~d} / \mathrm{d} x)^{3}+A_{0}^{1}\right] \mathrm{Ai}(x)  \tag{19}\\
& R^{2}(x)=\left[A_{2}^{2}(\mathrm{~d} / \mathrm{d} x)^{6}+A_{1}^{2}(\mathrm{~d} / \mathrm{d} x)^{3}+A_{0}^{2}\right] \mathrm{Ai}(x) \tag{20}
\end{align*}
$$

The high-order derivatives of $\mathrm{Ai}(x)$ can be reduced using $\left(\mathrm{d}^{2} / \mathrm{d} x^{2}\right) \operatorname{Ai}(x)=x \operatorname{Ai}(x)$ (Abramowitz and Stegun 1970). From $R^{1}(x)$ and $R^{2}(x)$ we can calculate the concentrations of 1 -mers and 2 -mers which are proportional to $(\mathrm{d} / \mathrm{d} x) R^{1}(0)$ and $(\mathrm{d} / \mathrm{d} x) R^{2}(0)$ respectively. However $(\mathrm{d} / \mathrm{d} x) R^{N}(0)$ is more easily evaluated directly from $\hat{R}^{N}(k)$ so we shall not pursue this approach further.

We now show how to calculate $(\mathrm{d} / \mathrm{d} x) R^{N}(0)$ for general $N$. First we note that the formal definition of $A_{n}^{N}$ is the coefficient of $(\mathrm{i} k)^{3 n} \exp \left(\frac{1}{3} i k^{3}\right)$ in the expression for $\tilde{R}^{N}(k)$.

Let us assume that $\tilde{R}^{N}(k)$ can be expressed

$$
\begin{equation*}
\tilde{R}^{N}(k)=\sum_{n=0}^{N} A_{n}^{N}(\mathrm{i} k)^{3 n} \exp \left(\frac{1}{3} \mathrm{i} k^{3}\right) . \tag{21}
\end{equation*}
$$

From equation (12)

$$
\begin{equation*}
k^{2} \tilde{R}^{N+1}(k)=\mathrm{i}(\mathrm{~d} / \mathrm{d} k) \tilde{R}^{N+1}(k)-\mathrm{i}(\mathrm{~d} / \mathrm{d} k) \tilde{R}^{N}(k) \tag{22}
\end{equation*}
$$

it follows that

$$
\begin{equation*}
\tilde{R}^{N+1}(k)=\left(-\sum_{n=0}^{N} A_{n}^{N} \frac{(\mathrm{i} k)^{3 n+3}}{3 n+3}+\sum_{n=1}^{N} A_{n}^{N}(\mathrm{i} k)^{3 n}+A_{0}^{N+1}\right) \exp \left(\frac{1}{3} i k^{3}\right), \tag{23}
\end{equation*}
$$

and
$R^{N+1}(x)=\left(-\sum_{n=0}^{N} A_{n}^{N} \frac{1}{3 n+3}\left(\frac{\mathrm{~d}}{\mathrm{~d} x}\right)^{3 n+3}+\sum_{n=1}^{N} A_{n}^{N}\left(\frac{\mathrm{~d}}{\mathrm{~d} x}\right)^{3 n}+A_{0}^{N+1}\right) \operatorname{Ai}(x)$.
Since $\tilde{R}^{0}(k)=\mathrm{Ai}(0)^{-1} \exp \left(\frac{1}{3} \mathrm{i} k^{3}\right)$ (equation (14)) induction shows that we have chosen the correct form for $\tilde{R}^{N}(k)$.

From the general form of $R^{N+1}(x)$ we now show that the boundary condition $R^{N+1}(x) \rightarrow 0$ as $x \rightarrow \infty$ is satisfied. $R^{N+1}(x)$ must be of the form $f(x) \operatorname{Ai}(x)+g(x) \times$ $(\mathrm{d} / \mathrm{d} x) \operatorname{Ai}(x)$ where $f$ and $g$ are polynomials of degree $\leqslant \frac{3}{2} N$ since $\left(\mathrm{d}^{2} / \mathrm{d} x^{2}\right) \operatorname{Ai}(x)=$ $x \operatorname{Ai}(x) . \operatorname{Ai}(x)$ and $(\mathrm{d} / \mathrm{d} x) \operatorname{Ai}(x)$ decay exponentially for large positive $x$ (Abramowitz and Stegun 1970) so the boundary condition at infinity is satisfied.

From equation (23) the $A_{n}^{N}$ satisfy

$$
\begin{equation*}
A_{n}^{N+1}=A_{n}^{N}-A_{n-1}^{N} / 3 n \quad n \geqslant 1 \tag{25}
\end{equation*}
$$

where we take $A_{N+1}^{N}=0$ and $A_{0}^{N+1}$ is determined by the normalisation constraint $R^{N+1}(0)=0$.

In order to satisfy the normalisation constraint we first note that (Abramowitz and Stegun 1970)

$$
\begin{equation*}
\frac{1}{2 \pi} \int(\mathrm{i} k)^{3 n} \exp \left(\frac{1}{3} \mathrm{i} k^{3}\right) \mathrm{d} k=\operatorname{Ai}(0) 3^{n}\left(\frac{1}{3}\right)_{n} \tag{26}
\end{equation*}
$$

where

$$
\begin{equation*}
3^{n}\left(\frac{1}{3}\right)_{n}=1,1,1 \times 4,1 \times 4 \times 7, \ldots \quad \text { for } n=0,1,2,3, \ldots \tag{27}
\end{equation*}
$$

and we define

$$
\begin{equation*}
B_{n}=3^{n}\left(\frac{1}{3}\right)_{n} . \tag{29}
\end{equation*}
$$

Using this notation the normalisation constraint can be written

$$
\begin{equation*}
\sum_{n=0}^{N} A_{n}^{N} B_{n}=0 \tag{29}
\end{equation*}
$$

and thus

$$
\begin{equation*}
A_{0}^{N+1}=\frac{1}{3} A_{0}^{N}-\sum_{n=1}^{N}\left(B_{n}-\frac{B_{n+1}}{3(n+1)}\right) A_{n}^{N} \quad N \geqslant 0 \tag{30}
\end{equation*}
$$

Since we have already found that $A_{0}^{0}=\mathrm{Ai}(0)^{-1}$ we can now calculate any $A_{n}^{N}$ in a sequence of simple steps using equations (25) and (30).

We now combine equations (25) and (30) in a linear operator 2. We identify $\tilde{R}^{N}(k)$ with the vector

$$
\begin{equation*}
A^{N}=\left(A_{0}^{N}, A_{1}^{N}, A_{2}^{N}, \ldots\right) \tag{31}
\end{equation*}
$$

where $A_{n}^{N}=0$ for $n>N$. The $\boldsymbol{A}^{\boldsymbol{N}}$ satisfy

$$
\begin{equation*}
A^{N+1}=2 A^{N} \tag{32}
\end{equation*}
$$

where $\mathscr{2}$ is defined by equations (25) and (30):
$2=\left[\begin{array}{ccccc}-\left(0-\frac{1}{3}\right) & -\left(1-\frac{1 \times 4}{6}\right) & -\left(1 \times 4-\frac{1 \times 4 \times 7}{9}\right) & -\left(1 \times 4 \times 7-\frac{1 \times 4 \times 7 \times 10}{12}\right) & \cdots \\ -\frac{1}{3} & 1 & 0 & 0 & \cdots \\ 0 & -\frac{1}{6} & 1 & 0 & \cdots \\ 0 & 0 & -\frac{1}{9} & 1 & \cdots \\ \vdots & \vdots & \vdots & \ddots & \cdots\end{array}\right]$.
It can be shown by induction using equation (32) and the definition of 2 that
$A_{n}^{N} \operatorname{Ai}(0)^{-1}= \begin{cases}0 & n>N \\ {\left[1\left(-\frac{1}{3}\right)\left(-\frac{1}{6}\right) \ldots\left(-\frac{1}{3 n}\right)\right]\left[\left(\frac{3 n+1}{3}\right)\left(\frac{3 n+4}{6}\right) \ldots\left(\frac{3 N-2}{3(N-n)}\right)\right]} & n \leqslant N .\end{cases}$
This is trivial for $n>0$ and for $n=0$ recall that $A_{0}^{N+1}=-\Sigma_{n=1}^{N+1} A_{n}^{N+1} B_{n}$ and the result falls out quickly.

We can now derive the recurrence relation satisfied by the concentration of $N$-mers,

$$
\begin{equation*}
c_{N+1}=\frac{3 N-1}{3 N+3} c_{n} \quad N \geqslant 0 . \tag{35}
\end{equation*}
$$

We first recall that $c_{N}=(\mathrm{d} / \mathrm{d} x) P^{N}(0)=(\mathrm{d} / \mathrm{d} x) R^{N}(0)\left(\beta^{*} / 2 D\right)^{1 / 3}$. Now

$$
\begin{equation*}
\frac{\mathrm{d}}{\mathrm{~d} x} R^{N}(0)=\int(\mathrm{i} k) \tilde{R}^{N}(k) \mathrm{d} k \tag{36}
\end{equation*}
$$

and

$$
\begin{equation*}
\frac{1}{2 \pi} \int(\mathrm{i} k)^{3 n+1} \exp \left(\frac{1}{3} \mathrm{i}^{3}\right) \mathrm{d} k=\frac{\mathrm{d}}{\mathrm{~d} x} \mathrm{Ai}(0) 3^{n}\left(\frac{2}{3}\right)_{n} \tag{37}
\end{equation*}
$$

where (Abramowitz and Stegun 1970)

$$
\begin{equation*}
3^{n}\left(\frac{2}{3}\right)_{n}=1,2,2 \times 5,2 \times 5 \times 8, \ldots \quad \text { for } n=0,1,2, \ldots \tag{38}
\end{equation*}
$$

We define

$$
\begin{equation*}
B_{n}^{+}=3^{n}\left(\frac{2}{3}\right)_{n} . \tag{39}
\end{equation*}
$$

Thus

$$
\begin{equation*}
c_{N}=\frac{\mathrm{d}}{\mathrm{~d} x} \operatorname{Ai}(0)\left(\beta^{*} / 2 D\right)^{1 / 3} \sum_{n=0}^{N} A_{n}^{N} B_{n}^{+} \tag{40}
\end{equation*}
$$

and we have reduced the problem of deriving the recurrence relation satisfied by the $c_{N}$ to showing that

$$
\begin{equation*}
\sum_{n=0}^{N+1} A_{n}^{N+1} B_{n}^{+}=\frac{3 N-1}{3 N+3} \sum_{n=0}^{N} A_{n}^{N} B_{n}^{+} . \tag{41}
\end{equation*}
$$

This is established by multiplying the $n$th term in $\sum_{n=0}^{N} A_{n}^{N} B_{n}^{+}, A_{n}^{N} B_{n}^{+}$by

$$
\begin{equation*}
\left(\frac{3 N-1}{3 N+3}+\frac{3 N+2}{3 N+3}\right)-\frac{3 N+2}{3 N+3} . \tag{42}
\end{equation*}
$$

The $n$th term in $\sum_{n=0}^{N+1} A_{n}^{N+1} B_{n}^{+}$is given by

$$
\begin{equation*}
A_{n}^{N+1} B_{n}^{+}=\left(\frac{3 N-1}{3 N+3}+\frac{3 N+2}{3 N+3}\right) A_{n}^{N} B_{n}^{+}-\frac{3 N+2}{3 N+3} A_{n-1}^{N} B_{n-1}^{+} \tag{43}
\end{equation*}
$$

and the result falls out.
This relation is valid for $N \geqslant 0$; thus from $A_{0}^{0}=\mathrm{Ai}^{-1}(0)$

$$
\begin{equation*}
c_{N}=\mathrm{Ai}^{-1}(0) \frac{\mathrm{d}}{\mathrm{~d} x} \operatorname{Ai}(0)\left(\beta^{*} / 2 D\right)^{1 / 3}\left(\frac{1}{3}\right)\left(\frac{2}{6}\right)\left(\frac{5}{9}\right) \ldots\left(\frac{3 n-4}{3 n}\right) \tag{44}
\end{equation*}
$$

The total concentration of $k$-mers, $\Sigma_{N=1}^{\infty} c_{N}$, is the probability that there is a $k$-mer on any chosen bond, $1-P_{1}^{0}$. This normalisation relationship is easily verified by considering the Taylor series expansion of $(1+x)^{1 / 3}$ about $x=0$ for $x=-1$ which gives

$$
\begin{equation*}
1=\frac{1}{3}+\frac{1}{3} \frac{2}{6}+\frac{1}{3} \frac{2}{6} \frac{5}{9} \ldots . \tag{45}
\end{equation*}
$$

Spouge (1988) has pointed out that the solution of the PCM solves an entire class of aggregation models, those in which a $k$-mer and a $p$-mer aggregate to form a ( $k+p$ ) modulo $N$ mer. Here a 0 -mer is a ghost cluster which does not change the mass of the $k$-mer with which it aggregates. In particular this class of aggregation models includes diffusion-limited annihilation which is aggregation modulo 2. Thus the concentration of 1 -mers in the diffusion-limited annihilation counterpart of the steady-state model we have considered is

$$
\begin{equation*}
c_{-}=\sum_{k=1}^{\infty} c_{2 k-1} . \tag{46}
\end{equation*}
$$

This series can be summed by again considering Taylor series expansion of $(1+x)^{1 / 3}$ about $x=0$ for $x=-1$ and $x=1$;

$$
\begin{equation*}
2^{1 / 3}=(1+1)^{1 / 3}-(1-1)^{1 / 3}=2\left(\frac{1}{3}+\frac{1}{3} \frac{2}{6} \frac{5}{9} \ldots\right) \tag{47}
\end{equation*}
$$

Thus $c_{-}=\mathrm{Ai}^{-1}(0)(\mathrm{d} / \mathrm{d} x) \operatorname{Ai}(0)\left(\beta^{*} / 2 D\right)^{1 / 3} 2^{-2 / 3}$ as found by Rácz (1985b).
We have performed preliminary simulations which confirm equation (44) for $c_{N}$; the results of more extensive simulation work on this aggregation model will be reported elsewhere.

This paper has described the derivation and solution of equations describing steady-state diffusion-limited $\mathrm{PCM} / \mathrm{ClCl}$ in one dimension. The methods used are widely applicable. A similar set of equations to equations (8) and (9) for $d$-dimensional PCM can be derived; $\left(\mathrm{d}^{2} / \mathrm{d} x^{2}\right)$ is replaced by the Laplacian operator for $d$ dimensions and multiplication by $x$ becomes multiplication by the 'volume' operator $x y z \ldots$. In fact, it is probable that the obvious generalisation of equations (8) and (9) for PCM on a fractal is correct. (See Anacker and Kopelman (1987), Lindenberg et al (1988) and Murat and Aharony (1986) for examples of diffusion-limited reactions on fractals.) Finally, this method can be extended to derive equations describing fluctuations.

## Acknowledgment

It is a pleasure to acknowledge the support of Active Memory Technology for this research.

## References

Abramowitz M and Stegun I (ed) 1970 Handbook of Mathematical Functions (New York: Dover)
Anacker L and Kopelman R 1987 Phys. Rev. Lett. 58289
Elderfield D 1987 J. Phys. A: Math. Gen. 20 L135
Family F and Meakin P 1988 Phys. Rev. Lett. 61428
Family F, Meakin P and Deutch J 1986 Phys. Rev. Lett. 57727
Kang K and Redner S 1984 Phys. Rev. A 302833
Kolb M, Botet R and Jullien R 1983 Phys. Rev. Lett. 511123
Lindenberg K, West B and Kopelman R 1988 Phys. Rev. Lett. 601777
Mandelbrot B 1982 The Fractal Geometry of Nature (San Francisco: Freeman)
Meakin P 1983 Phys. Rev. Lett. 511119
Murat M and Aharony A 1986 Phys. Rev. Lett. 571875
Rácz Z 1985a Phys. Rev. A 321129
__ 1985b Phys. Rev. Lett. 551707
Spouge J 1988 Phys. Rev. Lett. 60871
Torney D and McConnell H 1983 J. Phys. Chem. 871941
Vicsek T, Meakin P and Family F 1985 Phys. Rev. A 321122

