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Exact solution of an infinite class of stochastic models describing random sequential adsorption and other irreversible processes

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An exact general solution is presented for a multiply infinite class of irreversible filling or exchange processes on a rectangular lattice. There are many known applications in physics, chemistry, and biology. The dynamics of this class of models follows from the generating function for the number of permutations of n elements with specified cycles. As examples, general solutions are given for random sequential adsorption on a $2\times\infty$ strip with nearest-neighbor exclusion and for a conserved-order dynamics model recently formulated by Privman [Phys. Rev. Lett. **69**, 3686 (1992)].

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The stochastic filling of lattices has been studied for many years and continues to generate considerable interest [1–4]. An early solution of a problem of this type was the derivation of the jamming density for an infinite one-dimensional ($D=1$) array, given a rule that forbids occupying the nearest neighbor of an already filled cell [5]. Many other special cases have since been treated, assuming particular initial conditions on the underlying lattice. Here, I give an exact general solution for all initial states, for a multiply infinite class of irreversible filling or exchange processes. This solution encapsulates the dynamics of many systems known to be solvable by exact truncation, either fully or as an infinite subhierarchy. Its compact form enables easy computation of jamming densities and derivation of dynamical behavior. In more complex multidimensional lattices, one finds isomorphisms with direct sums of systems in this class. This may lead to an insight into unsolved problems in $D>1$.

Consider a rectangular lattice in a space of D dimensions, with $N\gg 1$ total sites. Random filling (or an equivalent exchange process) is assumed to occur under a rule that a site may be occupied only if certain conditions on its local neighborhood are met. In general, as is well known, such a process can be described by a homogeneous linear stochastic system

$$\frac{d\mathbf{P}}{dt}=\mathbf{M}\cdot\mathbf{P}\ . \tag{1}$$

In (1), $\mathbf{P}$ is a vector of (unnormalized) probabilities whose precise representation and interpretation are partly matters of choice and partly depend on the nature of the rule restricting the filling process. Explicit examples of representations of $\mathbf{P}$ are given below.

The defining feature of the processes discussed here is an $N\times N$ evolution matrix of the form

$$\mathbf{M}=-\begin{pmatrix}
\epsilon & c_1 & c_2 & c_3 & \cdots & \cdots & \cdots & c_{N-1}\\
0 & \epsilon+\Delta & c_1 & c_2 & c_3 & \cdots & \cdots & c_{N-2}\\
0 & 0 & \epsilon+2\Delta & c_1 & c_2 & c_3 & \cdots & \\
0 & 0 & 0 & \epsilon+3\Delta & c_1 & c_2 & \cdots & \cdots\\
\cdots & \cdots & \cdots & \cdots & \cdots & \cdots & \cdots & \cdots\\
0 & 0 & 0 & 0 & \cdots & & \cdots & c_1\\
0 & 0 & 0 & 0 & 0 & \cdots & \cdots & \epsilon+(N-1)\Delta
\end{pmatrix}. \tag{2}$$

In (2), $\{c_i\}$ is a set of (real) free parameters. The spectral scale Δ sets the unit of time. The zero point of the spectrum, ϵ, is a positive parameter that results in an overall multiplicative factor $\exp(-\epsilon t)$ in $\mathbf{P}$ and plays no further dynamical role. For notational convenience, the indices of the superdiagonals with $c_i\neq 0$ will be referred to as the sequences $S=\{\ldots,s_i,\ldots,s_j,\ldots\}$.

The class of generators defined by (2) has four important characteristics. (i) These matrices are upper triangular. The system of rate equations (1) can therefore be interpreted as describing a hierarchical process in which larger connected patterns (lattice animals) of "live" cells are destroyed as cells are occupied, and smaller lattice animals are thereby created [6]. (ii) The eigenvalues of (2) are equally spaced. This permits a representation of the probability vector $\mathbf{P}$ in which P_k is the probability of a configuration in which there are $k+n$ live cells in the interior of a lattice animal. Here, n is a fixed integer, typically small, set by the filling rule and the dimensionality of the underlying lattice. Characteristics (i) and (ii) typify

random sequential adsorption (RSA) and other irreversible processes [7]. (iii) The spectrum is nondegenerate. (iv) All elements on a superdiagonal are equal. Characteristics (iii) and (iv) are specific to the class of processes discussed here. They imply that there is only one type of lattice animal with a fixed number of interior points, and that a system of size $N+1$ described by (1) and (2) incorporates the dynamics of a system of size N or smaller. This permits exact solution by truncation, providing an opportunity that has been used to advantage in previous analyses [4] employing other methods. The theorem that follows makes the nesting property of the general solution of (1) and (2) explicit. It specifies the fundamental matrix $\exp(\mathbf{M}t)$ for the class of evolution operators defined by (2), generalizing the ansatz in [8] and the general solution for $D=1$ random dimer filling [9].

Theorem. In units of the spectral scale Δ, a fundamental matrix associated with (2) is

$$\exp(\mathbf{M}t)=x^{\epsilon}\begin{bmatrix} 1 & Q_1(x) & Q_2(x) & \cdots & \cdots & Q_{N-1}(x) \\ 0 & x & xQ_1(x) & xQ_2(x) & \cdots & xQ_{N-2}(x) \\ 0 & 0 & x^2 & x^2Q_1(x) & \cdots & x^2Q_{N-3}(x) \\ 0 & 0 & 0 & x^3 & \cdots & \cdots \\ 0 & 0 & 0 & 0 & \cdots & x^{N-2}Q_1(x) \\ 0 & 0 & 0 & 0 & \cdots & x^{N-1} \end{bmatrix} . \tag{3}$$

In (3), $Q_m(x)$ is a polynomial in $x=e^{-t}$, of order at most m, with $Q_0=1$. The generating function of this family is

$$G(s,x)=\sum_{m=0}^{\infty} Q_m(x)s^m=\exp\left[\sum_{m\in S} c_m\frac{(sx)^m-s^m}{m}\right] . \tag{4}$$

It is assumed that (3) is well defined for N arbitrarily large, and in particular that the $\{c_m\}$ in (2) are suitably bounded for $m\to\infty$ [10] so that the sum in the exponential in (4) converges in a finite neighborhood of $s=0$.

As illustrated in examples below, in a typical application with an infinite sequence of nonzero superdiagonals, $c_m=c$ for sufficiently large m. The resulting geometric subsequence in (4) leads to an overall factor in $G(s,x)$ that is polynomial in x. Note from (3) that increasing the size of the system from N to $N+1$ adds new information but does not change the preexisting structure. Note also that $G(s,x)=F(sx)/F(s)$ in (4), where $F(z)=\exp(\sum c_k z^k/k)$ has the form of the generating function of the cycle indicators, $C_n(c_1,c_2,\ldots,c_n)$, of the symmetric group. In principle, this allows the description of a given model in permutation terms.

These results can be verified as follows. By substituting (3) into (1), one confirms that (3) is a fundamental matrix solution of (1) and (2), provided the $Q_m(x)$ satisfy the differential equations [11]

$$x\frac{dQ_m}{dx}=\sum_{n=0}^{m-1} c_{m-n}x^{m-n}Q_n(x) , \tag{5a}$$

and the algebraic relations

$$mQ_m(x)=\sum_{n=0}^{m-1}(m-n)c_{m-n}(x^{m-n}-1)Q_n(x) . \tag{5b}$$

From (5b), $Q_m(1)=0$ for $m>0$, and therefore (3) equals the identity matrix $\mathbf{E}$ at $t=0$, as is required to enforce the initial conditions. By a theorem of linear systems theory, if a fundamental matrix solution $\mathbf{X}(t)$ of (1) has $\mathbf{X}(0)=\mathbf{E}$, $\mathbf{X}=\exp(\mathbf{M}t)$. This confirms (3). Linear systems theory also tells us that a system (1) and (2) of finite size has a general solution $\mathbf{P}(t)=\exp(\mathbf{M}t)\cdot\mathbf{P}(0)$. Using (3), this solution is

$$P_k(t)=x^{k-1+\epsilon}\sum_{m=0}^{N-1} Q_m(x)P_{k+m}(0) . \tag{6}$$

One observes from (4) and (6) that the initial conditions $P_k(0)=s^{k-1+\epsilon}$ decouple the system and result in a solution $P_k=(xs)^{k-1+\epsilon}G(s,x)$. Substitution of this result in (1) confirms (4).

Random dimer filling in $D=1$. It is instructive to apply these considerations to the standard $D=1$ example in which an arbitrarily long array of cells is randomly filled by dimers (o-o). Two equivalent representations of $\mathbf{P}$ are available. In the first, $P_k(t)$ is defined as the probability that a random k-long segment is empty, without regard to cells outside this segment [12]. Then $S=\{1\}$, $c_1=2$, and one has the evolution operator

$$\mathbf{M}_1=-\begin{bmatrix} 0 & 2 & 0 & 0 & \cdots \\ 0 & 1 & 2 & 0 & 0 \\ 0 & 0 & 2 & 2 & 0 \\ 0 & 0 & 0 & 3 & 2 \\ \cdots & \cdots & \cdots & \cdots & \cdots \end{bmatrix} , \tag{7}$$

in which the main diagonal entries correspond to filling interior pairs of a k segment and the superdiagonal 2's to filling the end cells and their external nearest neighbors. Using (4), (7) gives $G_1(s,x)=\exp[2s(x-1)]$, and we read off the polynomials $Q_m=2^m(x-1)^m/m!$ known to determine the general solution (6) for random dimer filling in $D=1$ [9] and to characterize the combinatorics of that process [6]. For an initial empty lattice, the jamming density is $\theta^*=1-P_1(\infty)=1-G_1(1,0)=1-e^{-2}$.

The second representation defines $P_k(t)$ as the probability of finding an empty segment precisely k cells long, as in the early treatment of monomer $D=1$ RSA with nearest-neighbor (NN) blocking [13]. Given this representation, there is a parent-child tower of rate equations in which the population P_k is fed by an infinite sequence $\{P_{k+2},P_{k+3},\dots\}$, and the evolution operator takes the form

$$\mathbf{M}_2=-\begin{bmatrix} 0 & 0 & -2 & -2 & \cdots & \cdots \\ 0 & 1 & 0 & -2 & -2 & \cdots \\ 0 & 0 & 2 & 0 & -2 & -2 \\ 0 & 0 & 0 & 3 & 0 & -2 \\ \cdots & \cdots & \cdots & \cdots & \cdots & \cdots \end{bmatrix}. \quad (8)$$

From (4), (8) is associated with the generating function

$$G_2(s,x)=e^{2s(x-1)}\left[\frac{1-sx}{1-s}\right]^2. \quad (9)$$

For an initial empty lattice, $P_1(t)=\lim_{s\to 1}[(1-s)^2G_2(s,x)]=\exp[2(x-1)](1-x)^2$, and we again have $\theta^*=1-e^{-2}$.

The two representations $\mathbf{M}_i$ are related by the similarity transformation $S^{-1}M_1S=M_2$, which is realized by the k-weighted sums and (shifted) second differences of the respective representations P_k:

$$S=\begin{bmatrix} 1 & 2 & 3 & 4 & \cdots & \cdots \\ 0 & 1 & 2 & 3 & 4 & \cdots \\ 0 & 0 & 1 & 2 & 3 & 4 \\ 0 & 0 & 0 & 1 & 2 & 3 \\ \cdots & \cdots & \cdots & \cdots & \cdots & \cdots \end{bmatrix}, \quad (10)$$

$$S^{-1}=\begin{bmatrix} 1 & -2 & 1 & 0 & \cdots & \cdots \\ 0 & 1 & -2 & 1 & 0 & \cdots \\ 0 & 0 & 1 & -2 & 1 & 0 \\ 0 & 0 & 0 & 1 & -2 & 1 \\ \cdots & \cdots & \cdots & \cdots & \cdots & \cdots \end{bmatrix}.$$

More general families of $D=1$ irreversible processes, such as the infinite set of cooperative RSA models considered in [8], also fall into the class defined by (2).

$D=2$ square ladder RSA with NN blocking. Recently, several groups have analyzed the dynamics of random filling of a $2\times\infty$ strip with nearest-neighbor exclusion [1–3]. This process has an infinite subhierarchy which is the direct sum $A\oplus B$ of two $D=1$ processes of class (2), equivalent under (10). In particular, dropping the lowest-order terms, omitting an overall factor x, and scaling time by $t\to 2t$, the rate equations in [1] can be written as independent subsystems of the form (1), with generators

$$\mathbf{M}_A=-\begin{bmatrix} 0 & -1 & -2 & -2 & \cdots & \cdots \\ 0 & 1 & -1 & -2 & -2 & \cdots \\ 0 & 0 & 2 & -1 & -2 & -2 \\ 0 & 0 & 0 & 3 & -1 & -2 \\ \cdots & \cdots & \cdots & \cdots & \cdots & \cdots \end{bmatrix},$$

$$\mathbf{M}_B=-\begin{bmatrix} 0 & 1 & 0 & 0 & \cdots & \cdots \\ 0 & 1 & 1 & 0 & 0 & \cdots \\ 0 & 0 & 2 & 1 & 0 & 0 \\ 0 & 0 & 0 & 3 & 1 & 0 \\ \cdots & \cdots & \cdots & \cdots & \cdots & \cdots \end{bmatrix}. \quad (11)$$

Using (4) and (11) we derive generating functions $g_B=\exp[s(x-1)]$ and $g_A=g_B(1-sx)^2/(1-s)^2$. The $s=1$ limit gives solutions for an initial empty lattice. Taking account of the asymptotic behavior of the lowest-order terms and incorporating the proper normalization, each g yields the known jamming limit [1–3] $\frac{1}{2}(1-1/2e)$. By inspection of g_B, the general solution (6) for B can be expressed in terms of the polynomials $(x-1)^m/m!$. The general solution for A follows from the observation that the systems defined by (11) are equivalent under (10). One may interpret subsystem B as a cooperative sequential adsorption process in $D=1$ in which there is unit probability per time that an empty cell will be occupied if (say) its left nearest neighbor is already occupied, and also if its next-nearest neighbor or any other cell further away is occupied (cf. [8]). As pointed out in [3], any sequential adsorption process on an $n\times\infty$ strip is equivalent to a $D=1$ process, in general involving competitive adsorption and long-range cooperativity. The results obtained here raise the question of which such processes, particularly for $D>1$, incorporate infinite subhierarchies that are equivalent to direct sums of exactly solvable processes in the class (2).

Conserved-order-parameter dynamics. Privman recently presented an interesting model in this class based on pairwise exchanges among cells, rather than random filling [14]. He considers conserved-order dynamics on a $D=1$ infinite lattice equally populated by particle species A and B. The only allowed dynamical moves are energy-lowering exchanges of nearest neighbors that decrease the number of adjacent A-B pairs. Given the locally conserved difference of the A and B densities, the dynamics of the system depends on the initial configuration, even for $t\to\infty$.

Privman solved the model exactly for two special choices of initial conditions in which the hierarchical rate equations decouple: the alternating pattern $\dots ABABAB\dots$, and random filling. In the notation used here, P_k represents the probability that a random $(k+1)$-long segment is ordered as $\dots ABABAB\dots$. This model then has $S=\{1,2\}$, with $c_1=c_2=2$, corresponding to two types of AB pair exchanges at the ends of a sequence of fully $\dots ABAB\dots$ ordered cells: (1) pairwise exchange of the pairs of cells at the ends of the sequence; (2) exchange of the two end cells with their nearest neighbors outside the sequence. The evolution matrix then has the form of the $D=1$ dimer filling generator (7), but there is an extra superdiagonal of 2's:

$$\mathbf{M}_{\rm ex}=-\begin{bmatrix} 0 & 2 & 2 & 0 & \cdots & \cdots \\ 0 & 1 & 2 & 2 & 0 & \cdots \\ 0 & 0 & 2 & 2 & 2 & 0 \\ 0 & 0 & 0 & 3 & 2 & 2 \\ \cdots & \cdots & \cdots & \cdots & \cdots & \cdots \end{bmatrix} . \quad (12)$$

From (4) and (12), the associated generating function is

$$G_{\rm ex}(s,x)=e^{2[s^2x^2/2+sx-s^2/2-s]} . \quad (13)$$

The factors of the form $\exp[(z^2/2+z)]$ in (13) are exponential generating functions for the number of involutions of n letters. For $s=(1,\frac{1}{2})$, (13) reproduces the jammed asymptotic states and dynamics of the interface densities derived in [14] for initial alternating and randomly filled states. From (5b), we have the recursion relations

$$(m+1)Q_{m+1}=-2(1-x)[Q_m+(1+x)Q_{m-1}] , \quad m=1,2,3,\ldots , \quad (14)$$

for the polynomials in (6). Explicitly, $Q_1=2(x-1)$, $Q_2=(3x-1)(x-1)$, $Q_3=2/3(1+5x)(x-1)^2,\ldots$. These results, the freedom to parametrize $\mathbf{M}$ in (2), and the nesting property of $\exp(\mathbf{M}t)$ suggest that the class defined by (2), augmented by similarity transforms, may contain all models exactly solvable by truncation. If so, this would give an alternative way to test for exact solvability in processes with $D>1$.

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[1] Y. Fan and J. K. Percus, J. Stat. Phys. **66**, 263 (1992).
[2] A. Baram and D. Kutasov, J. Phys. A **25**, L493 (1992).
[3] J. W. Evans and R. S. Nord, J. Stat. Phys. **69**, 151 (1992).
[4] J. W. Evans, Rev. Mod. Phys. (to be published).
[5] P. Flory, J. Am. Chem. Soc. **61**, 1518 (1939).
[6] A. Baram and D. Kutasov, J. Phys. A **22**, L251 (1989), link the Glauber dynamics of the hierarchy to a count of lattice animals. Equivalent combinatoric derivations are presented by R. Dickman *et al.*, J. Chem. Phys. **94**, 8252 (1991); and by Y. Fan and J. K. Percus, Phys. Rev. A **44**, 5099 (1991).
[7] See Ref. [4], Sec. IV D. For a discussion of the structure of infinite evolution matrices for the most general adsorption processes, see J. W. Evans and R. S. Nord, J. Stat. Phys. **38**, 681 (1985). There is a representation of $\mathbf{P}$ outside the class (2) that permits a formal general solution of multidimensional sequential adsorption. In particular, for a general adsorption process, let P_k be the probability of a random lattice animal with precisely k internal points, $k=1,2\ldots$. Then the associated evolution matrix $\mathbf{M}$ has the spectrum $\{0,-1,-2,\ldots\}$. Only the first superdiagonal is nonzero, with entries $\{-d_1,-d_2,-d_3,\ldots\}$, where d_i is the mean ratio of boundary to interior points of all lattice animals with i interior points. By a Laplace transform of the resulting system (1), the nonzero elements of $\mathbf{X}=\exp(\mathbf{M}t)$ are $X_{ii}=x^{i-1}$, $X_{i,i+m}=d_id_{i+1}\ldots d_{i+m}x^{i-1}(x-1)^m/m!$. For an initial empty lattice, the density of occupied cells can then be expressed as $\theta=-\sum_{j=1}^{\infty}a_j(x-1)^j/j!$, $a_j=d_1d_2\cdots d_j$. For NN blocked random filling, series of this form have previously been derived by Baram and Kutasov [6], and by Fan and Percus [6], using combinatorial arguments.
[8] J. J. Gonzalez *et al.*, Chem. Phys. **3**, 288 (1974).
[9] M. Bartelt and V. Privman, Int. J. Mod. Phys. B **5**, 2883 (1991).
[10] If one considers (1) as *a priori* an infinite system, rather than associating the behavior for $N\to\infty$ with arbitrarily large N in (3), the solution involves an arbitrary function $f(t)$. E. Hille, Ann. Mat. Pura Appl. 55 (4), 133 (1961). The nesting property of $\exp(\mathbf{M}t)$ noted below in the text allows an unambiguous solution for cases in which S is an infinite sequence, without the necessity to introduce a (possibly unphysical) boundary condition.
[11] Because x appears only in the combination sx in $G(s,x)$, Eqs. (5) define polynomials of the Brenke type. R. P. Boas and R. C. Buck, *Polynomial Expansions of Analytic Functions* (Springer, New York, 1964).
[12] E.g., E. R. Cohen and H. Reiss, J. Chem. Phys. **38**, 680 (1963).
[13] B. Widom, J. Chem. Phys. **58**, 4043 (1973).
[14] V. Privman, Phys. Rev. Lett. **69**, 3686 (1992).