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Random and Cooperative Sequential Adsorption on Infinite Ladders and Strips

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We analyze various processes where particles are added irreversibly and sequentially at the sites of infinite ladders or broader strips (i.e., on terraces) of adsorption sites. For "sufficiently narrow" strips or ladders, exact solution in closed form is possible for a variety of processes. Often this is most naturally achieved by mapping the process onto an equivalent one-dimensional process typically involving *competitive* adsorption. We demonstrate this procedure for sequential adsorption with nearest-neighbor exclusion on a $2 \times \infty$ square ladder. For other select processes on strips "slightly too broad" for exact solution, "almost exact" analysis is possible exploiting an empty-site shielding property. In this way, we determine a jamming coverage of 0.91556671 for random sequential adsorption of dimers on a $2 \times \infty$ square ladder. For "broader" strips, we note that the complexity of these problems quickly approaches that for $\infty \times \infty$ lattices.

KEY WORDS: Random and cooperative sequential adsorption; jamming coverage; ladders; strips; shielding.

1. INTRODUCTION

Currently, there is considerable interest in random sequential adsorption (RSA) processes on lattices.[1-4] Here particles are added irreversibly and sequentially at randomly chosen empty sites (or ensembles of empty sites for RSA of animals). More generally, one might consider a richer class of cooperative sequential adsorption (CSA) processes, where adsorption rates depend on the local environment.[2-4] Exact solution in closed form is possible for a broad class of RSA and CSA processes on one-dimensional (1D) lattices,[5] and on branching media,[6] by virtue of a shielding

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property of suitable walls of empty sites.[4] Interest in such 1D processes was motivated by application to the analysis of kinetics of reactions on polymer chains.[2,3]

Exact solution is *not* possible for sequential adsorption processes on 2D lattices, although an empty-site shielding property persists.[4] This is unfortunate, since these 2D processes also have important application to the study of kinetics and adlayer structure in chemisorption systems (in the simplest case, at low temperatures where adatom mobility is negligible). Roberts[7] was first to consider random dimer filling of adjacent pairs of empty sites as a model for adsorption of diatomics on single-crystal surfaces. Other examples of interest here are: (i) H_2O adsorption on Fe(001) modeled as random monomer filling on a square lattice with nearest neighbor (NN) exclusion,[8] i.e., single sites are filled randomly with the constraint that no NN pairs of filled sites can be formed; (ii) O_2 adsorption on Pt(100) modeled as random dimer filling on diagonal or next-NN sites of a square lattice with NN exclusion.[9]

Interest in 2D chemisorption systems provides some motivation to consider sequential adsorption processes on infinite strips of finite width. The hope is that one might obtain some insight into the behavior of "full" 2D systems, while exploiting some of the features which allow exact solution in 1D. Indeed this program has been initiated by Fan and Percus[10] and Baram and Kutasov,[11] who independently considered random monomer filling with NN exclusion on a $2 \times \infty$ square ladder. Exact solution was obtained here by combinatorial analysis,[10] by development of a closed set of rate equations,[10] and by formal expansion.[11]

Our objective here is to provide a concise overview of RSA (and CSA) on $n \times \infty$ strips with emphasis on the issue of solvability. In Section 2, we note that solution in closed form is possible for a variety of processes on "sufficiently narrow" strips. We show that often the most direct and natural method of analysis is to map the process onto an exactly solvable 1D process, typically involving *competitive* adsorption. This is the case, e.g., for the process considered by Fan and Percus.[10] Next, in Section 3, we note that for some select processes on strips "slightly too broad" for closed-form solution, analysis is readily achieved to arbitrary degree of accuracy. The basic strategy here is exploitation of the empty-site shielding property to greatly simplify the associated hierarchical rate equations. We thus obtain for the first time "almost exact solutions" to nontrivial truly 2D RSA processes. This procedure is demonstrated in detail for random dimer filling on a $2 \times \infty$ square ladder. Finally, we describe the situation for broader strips, and draw some general conclusions, in Section 4.

2. EXACTLY SOLVABLE PROCESSES

2.1. Cooperative Sequential Adsorption with NN Exclusion on a $2\times\infty$ Square Ladder

As noted above, Fan and Percus[10] considered RSA with NN exclusion on a $2\times\infty$ square ladder, choosing free boundary conditions on both infinite edges. In the context of chemisorption, the assumption of random adsorption is an idealization.[12] For this reason, and for general theoretical interest, we are thus motivated to consider a natural cooperative generalization of this problem. Specifically, we assume that adsorption with NN exclusion occurs with rates 1, α, and β for sites on a square ladder having zero, one, and two occupied next-NN sites, respectively (see Fig. 1). It is interesting to note here that the saturation or jamming coverage θ_J is independent of $\beta>0$. This is because sites with two occupied next-NNs do not influence adsorption elsewhere, and must eventually fill if $\beta>0$. The analogous feature has been exploited for a linear lattice.[13] We analyze this problem exactly in two ways.

In the *first approach*, we map this problem onto one of competitive cooperative adsorption on a linear lattice with sites corresponding to rungs on the ladder (see Fig. 2). The lattice sites are initially empty (0). Two species (1 and 2) compete for adsorption on the linear lattice corresponding to adsorption on different sides of the ladder. No 11 or 22 NN pairs can be formed. The adsorption rate for each species is 1, α, or β for sites with zero, one, or two NN sites occupied by the other species.

This problem is clearly a special case of the general process of competitive adsorption with NN cooperativity.[14] Here one denotes empty sites by 0, and considers competitive adsorption, $0\to i$, of species $i=1, 2,..., N$. Adsorption rates $\tau_{j,k}(i)$ depend on the states j and k of the NN sites relative to that being filled (where $0\leqslant j, k\leqslant N$). For convenience, we assume reflection invariance of rates, and set $\tau_{j,k}=\sum_{i=1,N}\tau_{j,k}(i)$. One can determine the coverage, pair correlations, cluster size distributions, etc., exactly as functions of time t. Here we focus on the determination of the

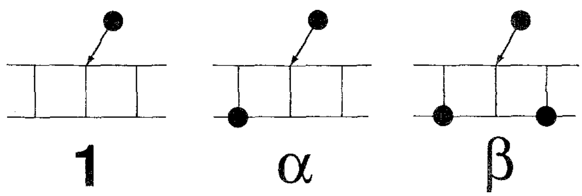

Fig. 1. Adsorption with NN exclusion at the sites of a $2\times\infty$ ladder with rates 1, α, and β for empty sites with zero, one, and two occupied next-NN sites, respectively.

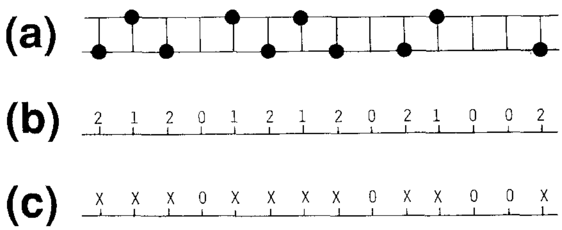

Fig. 2. (a) Sequential adsorption with NN exclusion on a $2\times\infty$ ladder, (b) mapped onto competitive adsorption on a linear lattice, and (c) related to single-species adsorption on a linear lattice.

coverage. Let P_0, P_{00}, P_{i0j},... denote the probabilities of finding 0, 00, $i0j$,... configurations, respectively. These quantities clearly are governed by the rate equations[14]

$$\begin{aligned} dP_0/dt &= -\sum_{j,k} \tau_{j,k} P_{j0k} \\ dP_{00}/dt &= -2\sum_{j} \tau_{j,0} P_{j00} \\ dP_{i0m}/dt &= \sum_{j} \tau_{j,0}(i)\, P_{j00m} - \tau_{i,m} P_{i0m} + \sum_{k} \tau_{0,k}(m)\, P_{i00k} \end{aligned} \tag{1}$$

etc., where we have used reflection invariance, and all sums run from 0 through N. Solution of these equations relies on a shielding property of empty pairs of sites.[4] One thus considers the conditional probabilities Q_j of finding a site in state j, given an adjacent empty pair. These satisfy[14] $dQ_0/dt = -\tau_{0,0}Q_0$ [so $Q_0 = \exp(-\tau_{0,0}t)$ for an initially empty lattice] if $j=0$, and a more complicated set of equations if $j\neq 0$. Knowledge of these Q_j allows factorization and truncation of the above equations.

For our problem where $N=2$, dramatic simplification of (1) is possible using the symmetry $1\leftrightarrow 2$, e.g., $P_{101}=P_{202}$, $P_{100}=P_{200}$, and conservation of probability, e.g., $P_{100}=(P_{100}+P_{200})/2=(P_{00}-P_{000})/2$. Incorporating our specific choice of rates, one thus obtains from (1) the closed set of equations

$$\begin{aligned} dP_0/dt &= -2[\alpha+(1-\alpha)\,Q_0]\,P_{00} - 2\beta P_{101} \\ dP_{00}/dt &= -2[\alpha+(2-\alpha)\,Q_0]\,P_{00} \\ dP_{101}/dt &= -\beta P_{101} + (1-Q_0)[\alpha+(2-\alpha)\,Q_0]\,P_{00}/2 \end{aligned} \tag{2}$$

where $Q_0 = e^{-2t}$. The coverage in our original problem is related to P_0 by $\theta = (1-P_0)/2$. Thus, the jamming coverage, $\theta_J = \theta(t=\infty)$, can be obtained via integration of the above equations from $t=0$ to ∞. For $\alpha=1$, we find that $\theta_J=(1-e^{-1})/2$ when $\beta=0$, and $\theta_J=(2-e^{-1})/4$ when $\beta>0$. The second result recovers that of Refs. 10 and 11. From the above equations, it is also clear that $\alpha=2$ has special significance. Here we find that $\theta_J=1/3$ when $\beta=0$, and $\theta_J=5/12$ when $\beta>0$. Further elucidation of these results comes from our second approach, where we also describe the asymptotic approach of θ_J to 1/2 as $\alpha\to\infty$.

In the *Second approach*, we retain the mapping of the original problem onto a linear lattice. However now imagine that we cannot distinguish between the species 1 and 2 (see Fig. 2). For simplicity, suppose first that $\beta=0$. Then what we see is adsorption of a single species X on a linear lattice with rates 2, α, and 0 for sites with zero, one, and two occupied NN, respectively. Of course, this problem can be solved exactly,[15] and again the desired θ is obtained from $(1-P_0)/2$. In fact, θ_J can be expressed in terms of complete and incomplete gamma functions[4] with arguments involving α.

It is interesting to note that for our original RSA problem with $\alpha=1$, these rates form an arithmetic progression. This constitutes a *very special case* of sequential adsorption with NN cooperativity on a linear lattice where single empty sites, rather than the usual empty pairs, suffice to shield[3,16]; here $P_0(t=\infty)=e^{-1}$, so $\theta_J=(1-e^{-1})/2$ as above for $\beta=0$. It is also clear now why the case $\alpha=2$ has special significance. This corresponds to "almost random filling" (ARF), where sites on a lattice are filled randomly, with the exception that completely surrounded sites cannot fill[17]; here $P_0(t=\infty)=1/3$, so $\theta_J=1/3$ as above for $\beta=0$. ARF problems are known to be solvable in all dimensions, and they have the special property that the spatial correlations are strictly finite range.[17] It is also instructive to consider the large-α regime. Using asymptotic expansions for the gamma functions mentioned above (see ref. 4), one finds that $P_0(t=\infty)=(2\pi/\alpha)^{1/2}/2+O(\alpha^{-1})$, so $\theta_J=[1-(2\pi/\alpha)^{1/2}/2+O(\alpha^{-1})]/2$ for $\beta=0$. We comment further on this regime below.

Next we turn to consideration of the general case where $\beta>0$. Now, two growing strings of contiguous filled sites $0XXX...XXX0$ *sometimes* merge. Whether this happens depends on the "phase" details of the impinging strings lost in this second approach, where we are blind to the state (1 or 2) of filled sites. Clearly, strings impinging to form 101 and 202 configurations will merge, and those forming 102 and 201 will not. However, it is clear that these two possibilities occur with equal frequency. Thus, $P_0(t=\infty)$ values for $\beta>0$ are simply obtained by halving those above for $\beta=0$. Associated θ_J values for $\alpha=1$ and 2 recover those reported from the first method for $\beta>0$, and we obtain $\theta_J=[1-(2\pi/\alpha)^{1/2}/4+O(\alpha^{-1})]/2$ for $\beta>0$ and large α.

2.2. Other Solvable Processes

One naturally asks what class of RSA (or CSA) problems are exactly solvable on a square ladder or broader strips. As a natural extension of the above example, and motivated by O_2/Pd(100) chemisorption,[9] we first consider RSA of dimers on next-NN sites with NN exclusion on $n \times \infty$ strips. For a $2 \times \infty$ ladder, the process is equivalent to competitive RSA on a linear lattice of either (i) two dimer species where different species cannot occupy adjacent sites, or (ii) two monomer species with longer-range exclusion effects (see Fig. 3). These problems can be solved exactly using the techniques of ref. 14. For a $3 \times \infty$ ladder, again exact solution is possible. This is most easily seen by mapping onto a process involving competitive filling of four distinct monomer species on a linear lattice (Fig. 3). Note that here blocking effects depend on whether one species is to the left or the right of another!

Another broad class of solvable processes involve RSA of molecules or "animals" of fixed shape, the only constraint being no double occupancy of sites. We assert that if the animals span the strip, for *all* adsorption orientations, then exact analysis is possible. To see this, write down the rate equations for (the probability of finding) a single empty site, then for the empty configurations to which it couples, and so on. In this way one generates an infinite set of complicated empty configurations of increasing length. The key is to note that implicitly all sites in the "middle" of these configurations must be empty. Then (probabilities for) these configurations can be exactly

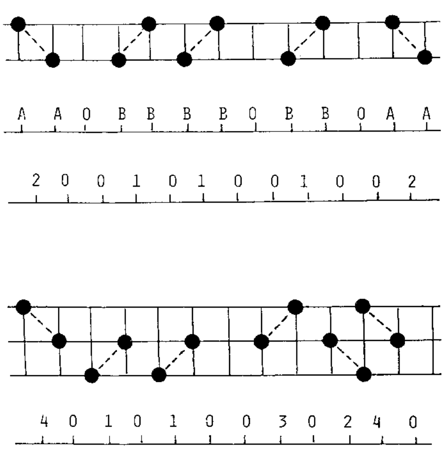

Fig. 3. Sequential adsorption of dimers with NN exclusion at next-NN sites on $n \times \infty$ ladders mapped onto competitive adsorption on linear lattices. Cases $n = 2$ and $n = 3$ are shown.

factorized into a finite set using the shielding property of walls of empty sites of suitable thickness.[4,19] One example of this type is RSA of "bent trimers" on a $2 \times \infty$ square ladder, where one finds the exact result, $\theta_J = 0.81545570$.

3. "ALMOST" EXACTLY SOLVABLE PROCESSES

3.1. Random Dimer Filling on a $2 \times \infty$ Square Ladder

Here we consider conventional RSA of dimers on a $2 \times \infty$ square ladder with free boundary conditions on both infinite edges: pairs of sites are selected at random and filled only if both are empty. For convenience, we set the deposition rate to unity. We believe that, as for regular 2D lattices,[18,19] exact closed-form solution of this random dimer-filling problem is not possible. However, this case has *special significance* since "almost exact" solution can be achieved by suitably exploiting the following empty-wall shielding property. For RSA of dimers on a square lattice, a continuous wall of empty sites of thickness one, which separates the lattice into two topologically separated parts, shields sites on one side from the influence of those on the other.[4,19] (Here "continuous" means nearest- or next-nearest-neighbor connectivity.) Thus, for the square ladder, a vertical pair of empty sites spanning the ladder constitutes a shielding wall.

Our approach here is to write down the exact infinite coupled hierarchy of rate equations for the probabilities of various configurations of empty sites. These include the probabilities L_n, B_n, and U_n of linear, bent, and U-shaped configurations of n, $n+1$, and $n+2$ empty sites, respectively (see Fig. 4). The procedure is straightforward: one simply accounts for all possible ways that an empty configuration can be destroyed by a dimer landing completely within or partly overlapping the configuration. Each such possibility produces a corresponding loss term in the rate equation for that configuration.[19] Clearly a dimer can partly overlap a single empty site in two ways when landing aligned with the ladder, and in one way when landing across the ladder. Thus one has $(d/dt)\, L_1 = -2L_2 - B_1$. Similarly, for an empty pair aligned with the ladder, a dimer can land completely overlapping the pair in one way, partly overlapping the pair in two ways when aligned with the ladder, and in two different ways when across the ladder. Thus one has $(d/dt)\, L_2 = -L_2 - 2L_3 - 2B_2$. Other examples are generated similarly.

It is clear that when writing the rate equations for L_n with $n \geq 2$, or B_n and U_n with $n \geq 3$, one generates configurations not in the set shown in Fig. 4. However, the *key simplification* for this process is that application of the shielding property allows us to factor probabilities of these new

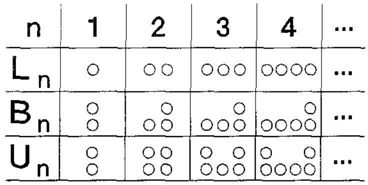

Fig. 4. Classification of empty-site configurations whose probabilities satisfy a minimal closed set of rate equations for RSA of dimers on a $2\times\infty$ ladder.

configurations in terms of the B_n and U_n. See Fig. 5 for examples. In this way, we obtain an infinite closed subset of equations for L_n, B_n, and U_n. These are explicitly

$$(d/dt)\,L_1 = -2L_2 - B_1$$

$$(d/dt)\,L_n = -(n-1)\,L_n - 2L_{n+1} - 2B_n - \sum_{k=2}^{n-1} B_k B_{n-k+1}/B_1 \qquad \text{for} \quad n \geqslant 2$$

$$(d/dt)\,B_1 = -B_1 - 4B_2$$

$$(d/dt)\,B_n = -(n + 2B_2/B_1)\,B_n - B_{n+1} - U_n - U_2 B_{n-1}/B_1 - \sum_{k=2}^{n-1} U_k B_{n-k+1}/B_1 \qquad \text{for} \quad n \geqslant 2$$

$$(d/dt)\,U_2 = -4(1 + B_2/B_1)\,U_2$$

$$(d/dt)\,U_n = -(n + 1 + 4B_2/B_1)\,U_n - 3U_2 U_{n-1}/B_1 - \sum_{k=2}^{n-2} U_k U_{n-k+1}/B_1 \qquad \text{for} \quad n \geqslant 3 \tag{3}$$

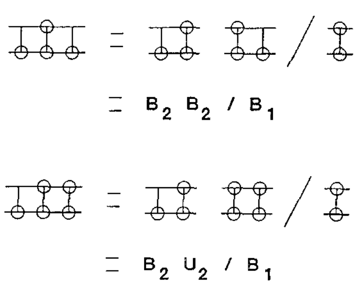

Fig. 5. Application of the empty-site shielding property for RSA of dimers to exactly factorize probabilities for various configurations of empty sites. Here these probabilities are represented by the configurations themselves. (See the text and Fig. 4 for terminology.) Note that these factorizations can be naturally recast in terms of conditional probabilities.[19]

It is not clear how to obtain an exact solution to these equations. Nonetheless, one can readily generate a sequence of truncation approximations of arbitrarily high Mth order. Here we retain L_n, B_n, and U_n for $n \leqslant M$, and close the above equations using the factorization approximations

$$L_{M+1} \approx L_M L_M / L_{M-1} \qquad \text{and} \qquad B_{M+1} \approx B_M L_M / L_{M-1} \tag{4}$$

These factorizations have simple physical interpretations. The first equates the conditional probabilities L_{M+1}/L_M and L_M/L_{M-1} for finding an empty site at the end of strings of M and $M-1$ empty sites, respectively. The second has a similar interpretation. These factorizations, and thus our approximation, become exact in the limit $M \to \infty$. In Table I, we present values for θ_J obtained by integrating Mth-order closures of (3) for a range of large M. These clearly show convergence to the exact $M = \infty$ value of $\theta_J = 0.91556671$. Estimates of the kinetics from integration of the truncated rate equations will be correspondingly precise. These can be compared against the known exact asymptotic behavior[20] $d\theta/dt \sim \theta_J - \theta$, so $\theta_J - \theta \sim ce^{-t}$ as $t \to \infty$. Finally, for comparison, we have run 1000 Monte Carlo simulation trials for random dimer filling on a $2 \times 10{,}000$ lattice (with periodic boundary conditions in the long dimension) to obtain $\theta_J = 0.91556 \pm 0.00013$, consistent with our analytic result.

3.2. Other Examples

For another example of the "almost solvable" type, we consider RSA of square tetramers (i.e., 2×2-mers) on $n \times \infty$ strips. For $2 \times \infty$ and $3 \times \infty$ strips, the problem trivially maps onto the 1D random dimer-filling problem solved long ago by Flory.[21] For a $4 \times \infty$ strip, exact analysis is

Table I. Estimates, Using *M*th-Order Truncation Approximations, for the Jamming Coverage θ_J for RSA of Dimers on a Square Ladder

M	θ_J
3	0.91447434
4	0.91547673
5	0.91556114
6	0.91556642
7	0.91556670
$\geqslant 8$	0.91556671

not possible. However, a continuous wall of empty sites of thickness one still shields.[19] (Here, however, continuous means NN connectivity.) For this problem, one proceeds as above, developing the rate equations for empty configurations. Note that from simple geometric considerations, if in any column across the ladder three sites are empty, then the fourth must also be empty. This observation, together with the above shielding property, allows one to obtain a reduced set of equations for probabilities of just a few types of empty configurations, analogous to (3). The Mth-order Markov-type approximations are then readily implemented. Another example of this type is RSA of 2×3-mers, which is exactly solvable on $2\times\infty$ and $3\times\infty$ strips and "almost solvable" on $4\times\infty$ strips.

4. GENERAL REMARKS CONCERNING PROBLEMS ON STRIPS

The above examples of sequential adsorption processes reflect general features of exact solvability on sufficiently narrow strips, possible "almost solvability" on slightly broader strips, and lack of solvability on even broader strips. RSA of monomers with NN exclusion is solvable on a $1\times\infty$ (1D) strip with $\theta_J=(1-e^{-2})/2\approx 0.4323$, on a $2\times\infty$ strip with $\theta_J=(2-e^{-1})/4\approx 0.4080$, but is not solvable or even "almost solvable" on $n\times\infty$ strips with $n\geqslant 3$ (where $\theta_J\to 0.3642$ as $n\to\infty$[4,8,22]). Random dimer filling is solvable on a $1\times\infty$ (1D) strip with $\theta_J=1-e^{-2}\approx 0.8646$, "almost solvable" on a $2\times\infty$ strip with $\theta_J\approx 0.9156$, but not on $n\times\infty$ strips with $n\geqslant 3$ (where $\theta_J\to 0.9068$ as $n\to\infty$[18,19]).

Some general remarks on "almost solvability" and its disappearance or absence for broader strips are appropriate here. "Almost solvability" relies on the following feature: application of the shielding condition results in simplification of the hierarchical rate equations producing a closed subset of equations for probabilities of just a few *types* of empty configurations. This, in turn, allows ready application of arbitrarily high-order truncation approximations. This feature is lost for broader strips. Consider RSA of dimers on a $3\times\infty$ strip. Here shielding is achieved by a vertical triple of empty sites spanning the strip, and this allows some simplification of the rate equations. However, one must still consider an infinite number of different *types* of empty configurations which do not span the strip. The situation is analogous to the treatment of RSA on 2D or 3D lattices, where configurations with all shapes must be considered, and arbitrarily high-order truncation approximations cannot be simply implementated.[4,19] Note that for RSA of monomers with NN exclusion on a $3\times\infty$ strip, one finds the same level of complication as in the $3\times\infty$ random dimer-filling problem. This precludes "almost exact" analysis.

Finally, we observe that any sequential adsorption process on an $n \times \infty$ strip can be mapped onto a sufficiently complicated 1D process. However, such 1D processes generally involve competitive adsorption and some complicated form of longer-range cooperativity. Because of the latter feature, exact solution is not generally possible, and the mapping is not particularly useful. However, for many processes on sufficiently narrow strips, this mapping procedure is a natural and efficient way to determine exact solvability and to obtain closed-form solutions.

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