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# Nonstandard convergence to jamming in random sequential adsorption: The case of patterned one-dimensional substrates

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## HIGHLIGHTS

- We study approach to the large-time jammed state in RSA.
- MC studies suggest new convergence laws for RSA on patterned 1D substrates.
- The standard assumption of constant small-gap size distribution is not always valid.
- Distribution can vanish linearly at zero gap sizes or have a non-zero size threshold.
- New power-law and exponential times power-law convergences to jamming are identified.

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## ABSTRACT

We study approach to the large-time jammed state of the deposited particles in the model of random sequential adsorption. The convergence laws are usually derived from the argument of Pomeau which includes the assumption of the dominance, at large enough times, of small landing regions into each of which only a single particle can be deposited without overlapping earlier deposited particles and which, after a certain time are no longer created by depositions in larger gaps. The second assumption has been that the size distribution of gaps open for particle-center landing in this large-time small-gaps regime is finite in the limit of zero gap size. We report numerical Monte Carlo studies of a recently introduced model of random sequential adsorption on patterned one-dimensional substrates that suggest that the second assumption must be generalized. We argue that a region exists in the parameter space of the studied model in which the gap-size distribution in the Pomeau large-time regime actually linearly vanishes at zero gap sizes. In another region, the distribution develops a threshold property, i.e., there are no small gaps below a certain gap size. We discuss the implications of these findings for new asymptotic power-law and exponential-modified-by-a-power-law convergences to jamming in irreversible one-dimensional deposition.



## 1. Introduction

Random sequential adsorption (RSA) is an important dynamical model [1–6] that describes irreversible deposition of particles or other objects on one-dimensional (1D) linear substrates, on two-dimensional (2D) surfaces, on scaffolds, etc. The objects are randomly transported to the substrate, but are attached only provided they do not overlap earlier-deposited objects. Once attached, the objects cannot move on the substrate or detach from it. Recently, there has been a renewed

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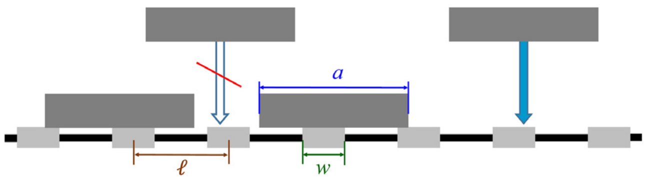


**Fig. 1.** Model showing random sequential adsorption with particle centers only depositing in landing intervals of width $w$ centered at the sites of a 1D lattice of spacing $l$. Deposition of an incoming object aimed at a $w$-interval can be rejected (shown by the crossed arrow) because of overlap, here with two earlier-deposited objects. An allowed-deposition configuration is shown on the right.

interest in the applications of RSA models [5,7–17] to pre-patterned 1D and 2D substrates. This has been in response to new experimental capabilities [18–43] to prepare micro- and nano-patterned substrates, including surfaces with well-defined preferential sites for specific particle attachment. In the studies of RSA one frequently focuses on the density and structure of the infinite-time jammed-state configuration. In applications, rapidly achieving dense coverage is usually preferred. Therefore, the asymptotic large-time laws describing approach to the jammed state coverage are of interest. Two standard prototype convergence laws have been found in extensively studied RSA models [1–6,9,44,45]. These include fast exponential vs. slow power-law approach to jamming as a function of time, $t$. The latter, power-law convergence can be modified by a power-of-a-logarithm factor [45] for some geometries of the depositing objects.

Recent technological advances have allowed tailoring the landing-site geometry for particle attachment, in addition to the earlier-studied effects of the particle shape and orientation, in order to control the RSA process. Presently, various growth and deposition processes have been experimentally realized on 1D lines [18–23] or nanotubes [24–26,43], etc., or 2D patterned surfaces [27–42]. Patterned surfaces find applications in electronics [24–28]; photovoltaics, optics, optoelectronics [29–32]; sensors, microarrays [33–38]; crystal growth and particle assembly [18–23,39].

The 1D RSA model provides a convenient test bench for studying the two convergence laws: exponential (fast) or power-law (slow). Specifically, lattice-aligned deposition gives exponential approach to jamming, whereas continuum, so-called "car parking" 1D deposition yields a power law, $\sim 1/t$, and these behaviors can actually be obtained by exact solution, e.g., [6]. More generally, convergence to jamming in 1D RSA can be understood by either directly studying the time-dependence of the coverage increase or be considering the behavior of the distribution of gaps [44] available for landing of the centers of particles the deposition attempts of which are not rejected (due to overlap with previously deposited particles). The "gaps" are generally regions of various possible shapes and orientations in more than 1D [45]. For discrete deposition, the "gap-size distribution" consists of delta function(s) representing available landing-site points, yielding exponential convergence for large times. For continuum deposition, the argument of Pomeau [44] has suggested that the gap size — measured by the length, $x$, into which a particle's center can land — distribution, $g(x)$, in 1D at large times is such that only the gaps that can fit a single particle dominate the dynamics. A further assumption of Pomeau [44] that $g(x)$ — defined such that $g(x)\,dx$ is the number of gaps of length between $x$ and $x+dx$ per unit length of the 1D line — is finite, non-singular at $x=0$, yields the $1/t$ convergence.

Swendsen [45] extended this argument to more than 1D; see also [4,6] for arguments on how the continuum limit is obtained from discrete deposition of decreasing "mesh." In higher dimensions, particle shapes, rotational freedom vs. fixed orientation, and other "degrees of freedom" in particle positioning, as well as substrate patterning can all affect the approach to jamming. Specifically, this argument [45] for fixed-orientation hypercubes depositing on a continuous $d$-dimensional "substrate" suggests convergence to jamming according to $\sim(\ln t)^{d-1}/t$. This offers an interesting example of a deviation from a purely power-law behavior. When substrate is patterned — which has been a topic of recent interest, there is numerical evidence for both exponential and power-law convergence in 2D [10,11] for various geometries of the particles or surface pattern. However, in some cases the form of the convergence in 2D could not be numerically classified as power-law or exponential [10]. A recent study [9] of 1D models with "imprecise particle positioning" — represented by a pattern of lattice landing sites broadened into intervals — has offered analytical arguments for both fast and slow convergence depending on the model parameters. We note that, theoretical studies have also included extensions of the RSA model to allow particle motion, detachment, and other dynamical effects, as well as surface heterogeneity, disorder, and non-uniformity, e.g., [7,46–57].

In this work, we consider the model [9] illustrated in Fig. 1. Here a uniform flux of particles of length $a$ reaches landing regions on a linear 1D substrate, but particles can only attach if they do not overlap one or two earlier-deposited particles. Substrate patterning is represented as follows: Particles can only be deposited if their centers fall within landing intervals that are broadened sites of a lattice of spacing $l$. These intervals have length $0 \le w \le l$, which extrapolates between lattice ($w=0$) and continuum ($w=l$) deposition. This model was introduced in [9], were analytical arguments were presented for that, the convergence to jamming can be fast, exponential-type in some regions of the parameter space of varying $a/l$ vs. $w/l$, and it can be slow, power-law-type in other regions. Here we revisit a Pomeau-type argument and adapt it for the present model. We then report a numerical study of convergence properties as the model parameters are varied. There are regions in the parameter space of this model with the standard exponential, $\sim e^{-\mathrm{const}\cdot t}$, or power-law, $\sim 1/t$, 1D convergences to jamming. However, our main finding in this work has been that, we also identified a segment with the $\sim 1/t^2$