Posted as e-print 0712.3567 at www.arxiv.org

Journal of Computational and Theoretical Nanoscience
Vol. 5, 2119–2123, 2008

Random Sequential Adsorption Model of Damage and Crack Accumulation: Exact One-Dimensional Results

Oleksandr Gromenko, Vladimir Privman*, and M. L. Glasser

Department of Physics, Clarkson University, Potsdam, New York 13699–5721, USA

The random sequential adsorption (RSA) model is modified to describe damage and crack accumulation. The exclusion for object deposition (for damaged region formation) is not for the whole object, as in the standard RSA, but only for the initial point (or higher-dimensional defect) from which the damaged region or crack initiates. The one-dimensional variant of the model is solved exactly.

Keywords: Damage Spreading, Crack Formation, Random Sequential Adsorption.

1. INTRODUCTION

Random sequential adsorption (RSA) has found many applications,[1,2] notably in irreversible surface deposition.[3,4] In this article, we consider a variant of the RSA model motivated by study of damage accumulation, e.g., multicracking. The first adaptation of the RSA model in mechanical-engineering literature to analyze fiber fragmentation in a unidirectional composite,[5] was followed by defect modeling in thin coatings.[6–9] In parallel, RSA-type models were also utilized in the statistical mechanics literature to explore approaches to damage development in several situations.[10–12]

Formation of a crack relieves stress in the nearby material, thus creating a region along the crack, possibly exclusive of the crack tip/edge area, in which new cracks cannot be initiated. Cracks formed in a thin coating can be approximated as a collection of one-dimensional (1D) "rods" (segments), subject to the no-overlap restriction as they form sequentially, similar to the traditional RSA. Experiments on multiple cracking in metal[13–15] and nonmetal[16–18] coatings have been modeled in this way. Furthermore, experiments[19–22] on load-induced damage in uniaxial fiber materials have also been interpreted in terms of RSA-type models that involve mathematics[5,9] similar to that encountered in the present article.

Generally, in materials subject to stress, cracks are initiated from static microscopic defects.[23,24] In the standard formulation of the RSA model, newly formed (or deposited on surfaces) objects cannot overlap objects formed earlier. Here we consider a variant of the RSA model that is arguably a better representation for damage accumulation in materials. Indeed, since the formed cracks relieve stress, except perhaps at their edges (as long as they grow), then new cracks cannot be initiated at or near the existing cracks. Therefore, as will be described later, the RSA model has to be modified to allow for "exclusion" of only the initial region of the crack. We do allow new cracks to be initiated close to the tips of the earlier formed cracks.

Despite the seeming simplicity of the RSA model, exact results are usually available only in one dimension[25,26] and in other special lattice geometries.[1] In this article, we consider a 1D model of deposition of segments of length ℓ on a line, and we allow exclusion to apply only to the head points of the segments. We argue that this model describes the accumulation of cracks which can reach lengths up to ℓ in 1D geometries.

In Section 2, we describe the model and then in Section 3, carry out an exact calculation of the open-interval probability, which allows us to evaluate the density of cracks as a function of time, t. In Section 4, we analyze the probability distribution of gaps, exactly and asymptotically for large times.

2. RSA OF SEGMENTS WITH SINGLE-END EXCLUSION

Our model consists of the deposition of segments of length ℓ on a linear substrate, as illustrated in Figure 1(a). The segments are transported to the substrate at the rate R (number of deposition attempts per unit length per unit time). Each segment has a "head" point, and the segment orientation is otherwise random: right-to-left or left-to-right segments arrive at the rates $R/2$ per unit length.

*Author to whom correspondence should be addressed.

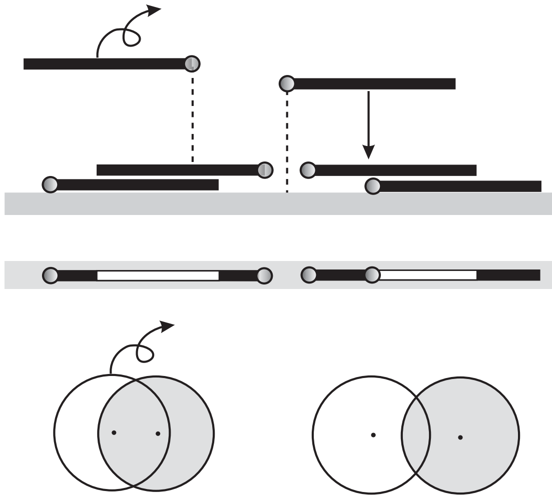

Fig. 1. Definition of the model: (a) Examples of possible configurations of sequential deposition of segments with head-point exclusion on a line. The curly arrow marks an arriving segment the deposition attempt of which will be rejected. (b) Formation of cracks that maps onto the deposition problem corresponding to the deposited pairs of segments in panel (a) above. The white area denotes the parts of the later-formed cracks that overlap already formed cracks. (c) A higher-dimensional example: Here circular damage areas are assumed to spread from center points. New damage area cannot be initiated from a center-point that overlaps earlier damaged areas. A configuration that will not be realized is marked by a curly arrow. As further explained in the text, this model illustrates that the point-exclusion RSA can be nearly identical to the ordinary RSA (here, of deposition of half-radius circles) especially when the objects and the exclusion point location are symmetrical.

Unlike the ordinary RSA, exclusion applies only to the adsorption of head points, which can be deposited only in areas that are not covered by the previously deposited segments. The lower pairs of segments in Figure 1(a) show two such possible deposition sequences. However, the segments with head points arriving above the covered areas are rejected (their deposition attempts fail and they are transported away from the substrate). One such example is illustrated in Figure 1(a).

Figure 1(b) illustrates the equivalence of the deposition model to crack formation. The head points of the segments correspond to the initiation points of the cracks in a 1D structure, e.g., fiber.[5–9] For tractability (otherwise our model would not be exactly solvable by the utilized technique), we assume that the cracks grow up to a fixed length ℓ, except when they run into the cracks formed earlier, as shown in the figure for two configurations, each corresponding to the first two segments deposited in the corresponding locations in Figure 1(a). Furthermore, we consider the situation where the cracks grow instantaneously once initiated, as compared to the time scales of new crack initiation.

Since there is a zone near a crack with reduced stress (except perhaps near the tip/edge), new cracks cannot initiate nearby. In fact, in real materials higher-dimensional damage configurations should be considered. They can originate not only from point-like defects and not all be identical in size and shape.[23,24] A simple illustration is given in Figure 1(c). Here circular damage regions originate from random centers in a plane. Additional damage can be initiated only from a center-point located in the previously undamaged areas. Figure 1(c) illustrates that the present models can be quite close to the ordinary RSA models. Indeed, unless the exclusion point is taken off-center or the objects are made less symmetrical, the model in the figure is nearly identical to the ordinary RSA of half-diameter circles (with no-overlap exclusion): the only difference is in the way the "covered" area is counted, whereas the center deposition kinetics is identical for both models.

We note that in the crack-formation nomenclature the parameter R corresponds to the rate (per unit time) of crack initiation per unit length of the 1D structure. Further discussion (and literature citation) is available[27] on how such statistical-mechanical parameters relate to the actual system properties. In this work we consider the simplest model, the main virtue of which is the possibility of deriving analytically exact or asymptotic results. More complicated models, which can be realistically compared with experimental data, would require large-scale numerical simulations and will be the subject of future work.

The quantities of interest for the model just defined are as follows. It is interesting to calculate the fraction of the total length that is undamaged, $F(t)$, which is expected to decrease monotonically from $F(0) = 1$ to $F(\infty) = 0$. A second quantity of interest is the density of deposited segment heads (the density of cracks initiated), $N(t)$. Obviously, $N(0) = 0$ because, for simplicity, we assume that initially the system is undamaged. However, we will establish that this quantity actually diverges logarithmically as $t \to \infty$. The calculation of $F(t)$ and $N(t)$ will be carried out exactly in the next section.

Another quantity of interest is the density (per unit length), $Q(x,t)dx$, of gaps (between the damaged areas) of sizes from x to $x+dx$. The quantity, $Q(x,t)$, gives us the size-distribution of undamaged regions in the 1D material sample. The total density (number per unit length) of gaps between the damaged regions is then

$$G(t) = \int_0^\infty Q(x,t)\,dx \tag{1}$$

Note that $Q(x,0) = 0$ and, obviously,

$$G(t \ll (R\ell)^{-1}) \simeq Rt \tag{2}$$

In addition to exact results, it is of interest to explore the small-gap statistics leading to the derivation of the asymptotic results for $Q(x,t)$ in the large-time regime, when the total number of gaps has reached the saturation value $G(\infty)$. This matter will be further elucidated in Section 4, where the results for the gap density are presented.

3. EMPTY-INTERVAL PROBABILITY

Exact results for the present model can be derived for the empty-interval probability, $P(x, t)$. This quantity represents the probability that a randomly chosen interval of length x is fully undamaged, i.e., open for deposition of segment-heads. Since such an interval could be a part of a longer empty region, $P(x, t)$ is *not* proportional to the gap density $Q(x, t)dx$ introduced earlier. These quantities will be related in Section 4. However, one can easily convince oneself that the fraction of the total length that is undamaged is given by

$$F(t) = P(0, t) \tag{3}$$

The probability $P(x, t)$ satisfies the equation

$$\frac{\partial P(x,t)}{\partial t} = -RxP(x,t) - R\int_x^{x+\ell} dy\, P(y,t) \tag{4}$$

where the first term on the right-hand side accounts for depositions where a head-point lands in the interval x. The second term on the right-hand side corresponds to deposition of segments with the head-point landing outside the interval x, which requires that a larger interval, of length y (up to $x+\ell$), be open for deposition.

For the initial condition $P(x, 0) = 1$, the x dependence in (4) can be eliminated by the Anzatz

$$P(x,t) = \exp(-Rxt)F(t) \tag{5}$$

Substituting (5) in (4), one obtains a differential equation for $F(t)$ with the initial condition $F(0) = 1$. The result is

$$P(x,t) = \exp\left[-Rxt + \int_0^t \frac{\exp(-R\ell u) - 1}{u}\, du\right] \tag{6}$$

For the fraction of the undamaged area, we have

$$F(t) = \frac{e^{-\gamma}}{R\ell t} e^{-E_1(R\ell t)} \tag{7}$$

where $E_1(\tau) = \int_\tau^\infty (e^{-\omega}/\omega)\, d\omega$ is a standard exponential integral function, and $\gamma = 0.57721566490\ldots$ is the Euler gamma constant.[28] For large times, the fraction of the undamaged (available for deposition) length vanishes according to

$$F(t \gg (R\ell)^{-1}) = \frac{e^{-\gamma}}{R\ell t}\left(1 - \frac{e^{-R\ell t}}{R\ell t} + \cdots\right) \approx \frac{e^{-\gamma}}{R\ell t} \tag{8}$$

where from now on the sign $\approx$ will denote the large-time asymptotic behavior.

While the total undamaged length shrinks to zero at large times, the total density of the deposited segments (the total density of cracks initiated), $N(t)$, actually diverges logarithmically,

$$N(t \gg (R\ell)^{-1}) \approx \frac{e^{-\gamma}}{\ell} \ln(R\ell t) \tag{9}$$

This behavior, characteristic of the present model, is opposed to ordinary 1D RSA. Indeed,

$$N(t) = R\int_0^t F(v)\, dv = R\int_0^t \exp\left[\int_0^v \frac{e^{-R\ell u} - 1}{u}\, du\right] dv \tag{10}$$

This can be also written

$$N(t) = \frac{e^{-\gamma}}{\ell} \times \left[e^{-E_1(R\ell t)} \ln(R\ell t) - \int_0^{R\ell t} du \frac{\ln u}{u} e^{-u} e^{-E_1(u)}\right] \tag{11}$$

where the first term dominates the divergence (9).

4. THE GAP DENSITY DISTRIBUTION

In order to relate the gap density distribution $Q(x, t)$ to the empty interval probability $P(x, t)$, let us discretize the distances in the problem in steps Δx, where ultimately, $\Delta x \to 0$. Specifically, the linear substrate will become a lattice of spacing Δx, and all the other lengths, including x and ℓ, will be assumed multiples of Δx.

Let us denote by $\Omega(x, t)$ the probability that a randomly chosen interval of length $x = n\Delta x$ has its rightmost lattice site blocked, while all the other, $n-1$, lattice sites are empty. We can write a relation between probabilities,

$$P(x - \Delta x, t) = P(x, t) + \Omega(x, t) \tag{12}$$

which, in the limit of small Δx, gives

$$\Omega(x,t) = -\frac{\partial P(x,t)}{\partial x}\Delta x \tag{13}$$

Let us now define the probability $\Phi(x, t)$ that a randomly chosen interval of length $x = n\Delta x$ has both its rightmost lattice site and its leftmost lattice site blocked, while all the other, $n-2$, lattice sites are empty. The probability relation is then

$$\Omega(x - \Delta x, t) = \Omega(x, t) + \Phi(x, t) \tag{14}$$

from which we get, for small Δx,

$$\Phi(x,t) = \frac{\partial^2 P(x,t)}{\partial x^2}(\Delta x)^2 \tag{15}$$

Since the density (per unit length) of fixed-length intervals on the lattice is $1/\Delta x$, for the density of gaps of size x we get

$$Q(x,t)\Delta x = \Phi(x,t)/\Delta x = \frac{\partial^2 P(x,t)}{\partial x^2}\Delta x \tag{16}$$

Finally, we get

$$Q(x,t) = \frac{\partial^2 P(x,t)}{\partial x^2} = (Rt/\ell) e^{-Rxt - E_1(R\ell t) - \gamma} \tag{17}$$

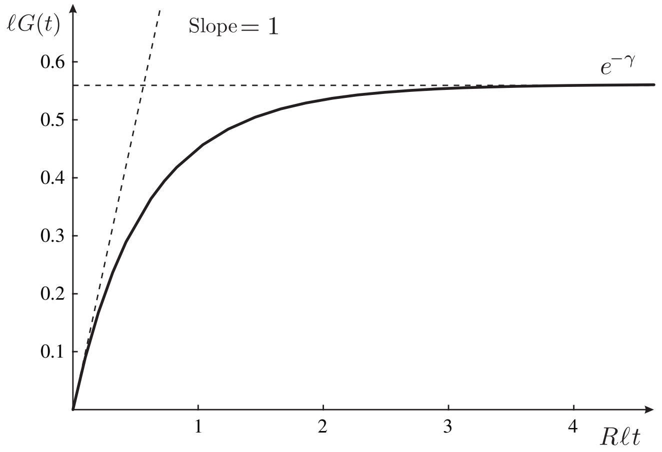

Fig. 2. The behavior of the density of gaps as a function of time. The asymptotic large-time regime is realized to a good approximation for $t > t_0$, where $t_0 \simeq 3(R\ell)^{-1}$.

The total density of gaps per unit length, cf. (1), is thus

$$G(t) = e^{-E_1(R\ell t)-\gamma}/\ell \tag{18}$$

This function is shown in Figure 2.

Since exact solutions are not possible in most geometries, considerations that can yield asymptotic large-time behavior play an important role in the study of RSA models. We point out that in the opposite, short-time limit, when deposition events are initially not correlated, a low-density approximation leads to relations of the type (2). The large-time behavior, however, is less straightforward. It can be analyzed to various degrees, depending on the model details, by considering the dynamics of gaps small enough such that their evolution can be described approximately in term of their size and shape. Specifically, for ordinary RSA one considers[29–31] the regime of late times when most of the remaining gaps are small enough to accommodate at most one depositing object. One further assumes that the distribution of these gaps at some large t_0, from which time on the small-gap-dominated behavior sets in, is smooth with respect to size, and their shape distribution is also known (since we know the geometry of the deposited objects surrounding the gaps) and is similarly smooth. The kinetics of these gaps, blocked by the arriving depositing object, is then evaluated[29,31] in order to obtain the evolution of the gap size and shape distributions for $t > t_0$, from which other deposit properties can be found.

In our case, it is natural to consider the large-time behavior of the gap distribution under the assumption that it is dominated by gaps not larger than the segment size ℓ. Indeed, such gaps cannot be fragmented by segment deposition: they can only be shortened. Therefore, the asymptotic regime is for times $t > t_0$, see Figure 2, late enough for the function $G(t)$ to have reached its constant value. However, other quantities, including the gap distribution $Q(x, t)$, with $x \leq \ell$, still vary with time. The equation that describes this variation is

$$\frac{\partial Q(x,t)}{\partial t} \approx -RxQ(x,t) + R\int_x^{\ell(\to\infty)} Q(y,t)\,dy \tag{19}$$

Here, not only is the equation already approximate, but we seek the asymptotic large-time solution in the regime dominated by small gaps, for which we can further replace $\ell \to \infty$ in the upper limit of the integration, as shown, because ℓ actually plays no role in the evolution of small gaps (most of the length of the deposited segments covers already blocked area and is not active in further blocking).

The solution of the resulting equation is

$$Q(x,t) \approx qte^{-Rxt} \tag{20}$$

where q is a constant that cannot be determined from the large-time analysis. It can be found by assuming properties for the distribution at a reference time t_0, as mentioned for the standard RSA problem. Alternatively, we can calculate (or numerically estimate) some less detailed property related to the gap density distribution, for instance the fraction of the total length that remains open for deposition

$$F(t) = \int_0^\infty xQ(x,t)\,dx \approx \frac{q}{R^2 t} \tag{21}$$

Here we just use the exact large-time expression (8) to get

$$q = Re^{-\gamma}/\ell \tag{22}$$

which, finally, gives the asymptotic form of the gap density distribution,

$$Q(x,t) \approx Rte^{-Rxt-\gamma}/\ell \tag{23}$$

consistent with the exact result (17).

In summary, we have studied an exactly solvable RSA-type model motivated by the problem of accumulation of cracks in a 1D system. The exclusion condition applies only to the head-points of the segments, deposition of which models crack formation. Study of more realistic systems in higher dimensions will require numerical simulations. We point out that even in the simplest 1D model considered here, some interesting quantities are not amenable to exact solution. For example, we can define the probability, $B(x,t)dx$, of the blocked intervals of length from $x(\geq\ell)$ to $x + dx$, similarly to the distribution of the unblocked gaps, $Q(x,t)dx$. The quantity $B(x,t)$ is of particular interest because it is expected to have a delta-function singularity at $x = \ell$, corresponding to a final (initially increasing, but eventually decaying as $t \to \infty$) density of isolated deposited segments forming blocked-intervals of length exactly ℓ. We were not able to calculate $B(x,t)$ exactly. Finally, we explored the large-time small-gap-dominated asymptotic behavior along the lines of the derivation appropriate to standard RSA models.

Acknowledgments: The authors thank Professor A. Cadilhe for instructive discussions, and acknowledge support of their research by the ARO under grant W911NF-05-1-0339 and by the NSF under grants DMR-0509104 and CCF-0726698.

References

1. J. W. Evans, *Rev. Mod. Phys.* 65, 1281 (**1993**).
2. M. C. Bartelt and V. Privman, *Int. J. Mod. Phys. B* 5, 2883 (**1991**).
3. V. Privman, *J. Adhesion* 74, 421 (**2000**).
4. V. Privman (ed.), Adhesion of Submicron Particles on Solid Surfaces, Special Volume of Colloids and Surfaces A (**2000**), Vol. 165, pp. 1–428.
5. W. A. Curtin, *Mater. Sci.* 26, 5239 (**1991**).
6. P. Calka, A. Mézin, and P. Vallois, *Stochast. Proc. Appl.* 115, 983 (**2005**).
7. F. M. Zhao, T. Okabe, and N. Takeda, *Compos. Sci. Technol.* 60, 1965 (**2000**).
8. A. Mézin and B. Sajid, *Thin Solid Films* 358, 46 (**2000**).
9. C.-Y. Hui, S. L. Phoenix, M. Ibnabdeljalil, and R. L. Smith, *J. Mech. Phys. Solids* 43, 1551 (**1995**).
10. P. L. Krapivskyy and E. Ben-Naim, *Phys. Rev. E* 50, 3502 (**1994**).
11. P. L. Krapivskyy and E. Ben-Naim, *J. Phys. A* 29, 2959 (**1996**).
12. K. M. Crosby and R. M. Bradley, *Phil. Mag. Lett.* 75, 131 (**1997**).
13. P. M. Ramsey, H. W. Chandler, and T. F. Page, *Thin Solid Films* 201, 81 (**1991**).
14. F. S. Shieu, R. Jay, and S. L. Sass, *Acta Metall. Mater.* 38, 2215 (**1990**).
15. M. S. Hu and A. G. Evans, *Acta Metall. Mater.* 37, 917 (**1989**).
16. D. C. Agrawal and R. Raj, *Acta Metall. Mater.* 37, 1265 (**1989**).
17. T. R. Watkins and D. J. Green, *J. Am. Ceram. Soc.* 77, 717 (**1994**).
18. F. Delannay and P. Warren, *Acta Metall. Mater.* 39, 1061 (**1991**).
19. W. A. Fraser, F. H. Ancker, A. T. DiBenedetto, and B. Elbirli, *Polym. Compos.* 4, 238 (**1983**).
20. A. N. Netravali, R. B. Henstenburg, S. L. Phoenix, and P. Schwartz, *Polym. Compos.* 10, 226 (**1989**).
21. H. D. Wagner and A. Eitan, *Appl. Phys. Lett.* 56, 1965 (**1990**).
22. V. Rao and L. T. Drzal, *Polym. Compos.* 12, 48 (**1991**).
23. B. Lawn, Fracture in Brittle Solids, Cambridge University Press, Cambridge (**2004**).
24. K. B. Broberg, Cracks and Fracture, Academic Press, San Diego (**1999**).
25. V. Privman (ed.), Nonequilibrium Statistical Mechanics in One Dimension, Cambridge University Press, Cambridge (**1997**).
26. J. J. Gonzáles, P. C. Hemmer, and J. S. Høye, *Chem. Phys.* 3, 228 (**1974**).
27. A. Dementsov and V. Privman, *Physica A* 385, 543 (**2007**).
28. I. S. Gradshteyn and I. M. Ryzhik, Table of Integrals, Series and Products, Academic Press, Burlington (**2007**).
29. Y. Pomeau, *J. Phys. A* 13, L193 (**1980**).
30. R. H. Swendsen, *Phys. Rev. A* 24, 504 (**1981**).
31. V. Privman, J.-S. Wang, and P. Nielaba, *Phys. Rev. B* 43, 3366 (**1991**).

Received: 8 March 2008. Accepted: 25 April 2008.