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Random Sequential Adsorption: Relationship to Dead Leaves and Characterization of Variability

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A new construction for the planar unbounded random sequential adsorption (RSA) model is presented, which allows for a direct comparison with Matheron's "dead leaves" model. Furthermore, for the case of disks with random radii the problem of statistical determination of the proposal radius distribution is discussed. Finally, second order characteristics related to the pair correlation function are suggested for describing the variability of the RSA disk systems.

KEY WORDS: Coverage; dead leaves model; Matérn's hardcore process; random disk packing; random sequential adsorption; two-point distribution; variability of spatial disk distribution.

1. INTRODUCTION

The random sequential adsorption (RSA) model has attracted considerable attention as random packing model in Euclidean spaces of various dimensions and bounded subsets thereof.[1,2] Both the RSA model on a lattice and the continuous RSA model have many applications.[1,3]

This paper considers the unbounded, and continuous case; the packed objects are disks with either constant or random radii. We restrict ourselves to the planar case; nevertheless all the presented concepts extend directly to higher dimensions. A mathematical definition of the RSA model is recalled and a new construction is given in Section 3, which is used to show that the RSA model is closely connected to Matheron's dead leaves model,[4,5] another model for randomly distributed disks. Both models

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contain the same subsystem of hard disks, the system of intact disks of the dead leaves model. If the disks have constant size, the subsystem can also be identified as a limit case of Matérn's second hard-core process.[6–8] The dead leaves model is used in microscopy and other fields to describe piled layers of grains.[5, 9] Figure 1 below shows a simulation restricted to a rectangle.

Sections 4 and 5 focus on the two-point (or binary) radius distribution which has already been examined by Meakin and Jullien,[3] whose main goal was the characterization of the time dependence of the incomplete RSA model and the determination of the final coverage. Here a statistical problem for an RSA model with disks of variable radii is discussed. Assume that an empirical structure is given which can be described by an RSA model of unknown radius distribution. Then one is confronted with two radius distributions: that from which the disks are selected for attempted addition, the *proposal distribution*, and the radius distribution of adsorbed disks, the *resulting distribution*. The radii corresponding to the latter tend to be smaller than those belonging to the proposal distribution; only in the case of constant radii both (trivial) distributions coincide. In practical

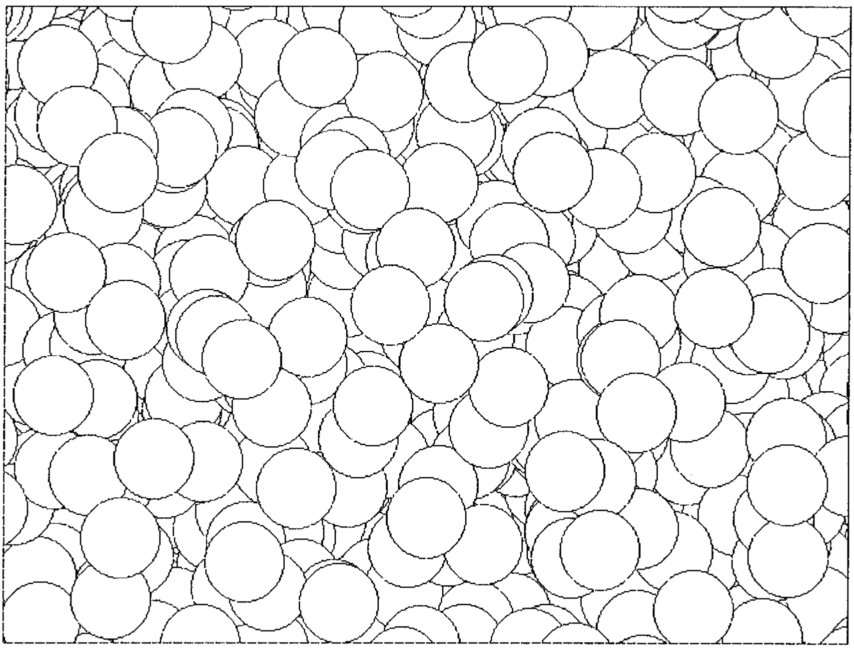

Fig. 1. A simulation of Matheron's dead leaves model. The "intact" disks form a system of non-intersecting disks, which is a subset of the RSA model.

problems (given only the pattern of disks) usually the resulting distribution is known, while the proposal distribution is unknown. The latter has to be determined for simulating and modelling.

Furthermore the variability of the spatial pattern of the complete RSA model, which is usually described by means of the pair correlation function, is considered. Namely, in the case of variable radii the characterization of the relationship between mutual disk positions and radii is also of interest. To this end suitable tools are presented, namely the "mark connection functions" $p_{ij}(r)$.

2. THE INFINITE RSA MODEL

In this section a mathematical definition is recalled and a new construction is given for the homogeneous and isotropic planar RSA model, i.e., for the thermodynamic limit. (In the following the term RSA model is used also for the "marked point process" that consists of the disk centres marked by the disk radii.)

The homogenous and isotropic, unbounded RSA model is defined as the limit of a spatial birth process[8, 10] as suggested by J. Møller;[11] this definition can be seen as an extension of the definition of the well-known car parking problem to higher dimensions.[1] A spatial birth process is a family of homogeneous point processes in the plane indexed by the time t, or a time continuous Markov process whose states are point patterns. At isolated instants of time, a further point is "born," i.e., added to the existing point pattern where the probability of a change depends only on the current configuration of the process. Births are controlled by a birthrate b, a positive function $b(x, \varphi)$ with

$$\int_W b(x, \varphi)\, dx < \infty$$

for any bounded set W and any configuration φ of disks. The probability that a birth occurs within the set W in the infinitesimal time interval $[t, t+ds]$, given that the process has configuration φ at time t, equals

$$ds \int_W b(x, \varphi)\, dx + o(ds)$$

In the case of varying radii the configurations φ consist of marked points $(x; r)$, where x is the disk centre and r the corresponding radius. The set-theoretic union of all the disks $B(x, r)$ of a configuration is denoted by s_φ.

Let f_{pr} be the probability density function of the proposal radius distribution. Then the birth rate of the RSA birth process is

$$b((x;r),\varphi)=(1-\mathbf{1}_{s_\varphi\oplus B(o,r)}(x))\,f_{pr}(r) \tag{2.1}$$

where $X\oplus B(o,r)=\bigcup_{y\in X}B(y,r)$ and $\mathbf{1}_A$ is the indicator function of the set A, i.e., $\mathbf{1}_A(x)=1$ for $x\in A$ and 0 otherwise. The birth-rate simply vanishes in all the points x where a disk of radius r cannot be placed.

The RSA model at the jamming limit is obtained for $t\to\infty$, while the configuration at time $t=0$ is the empty set.

The term "time" has a different meaning than in Meakin and Jullien:[3] There, *time* is proportional to the number of points attempted to add. Here the *points* depend on the time through a Poisson process. The latter is of unit rate if the sampling window W is a set of unit area.

An alternative construction of the RSA model is based on the system of marked points $(x;t,r)$ where x gives the location, t the instant of time, and r the random radius. The points $(x;t)$ follow a homogeneous Poisson point process of unit intensity on $R^2\times[0,\infty)$.

In order to obtain the RSA model in the jamming limit, the system of marked points is thinned in infinitely many steps according to the following rule; in each step the thinning is performed on the whole plane.

First Step. A point $(x;t,r)$ is retained if there is no other point $(x';t',r')$ with

$$t'<t \qquad \text{and} \qquad |x-x'|<r+r' \tag{2.2}$$

where $|\cdot|$ denotes the Euclidean distance. Each retained point $(x;t,r)$ eliminates all the points $(x^*;t^*,m^*)$ with

$$t^*>t \qquad \text{and} \qquad |x^*-x|<r^*+r \tag{2.3}$$

Clearly, as the collection procedure by rule (2.2) and the elimination procedure by rule (2.3) are applied sequentially and as condition (2.2) and (2.3) are exclusive, the order in which the points are collected respectively eliminated is unimportant.

Second Step. Rule (2.2) and subsequently rule (2.3) are applied to the reduced system of marked points; and so on.

By suppressing the time mark t of the retained marked points, one obtains the RSA model in the jamming limit.

Remarks. 1. The thinning steps do not correspond to a "natural time" like the birth time in the birth process construction.

2. Instead of a Poisson process on $R^2 \times (0, \infty)$, any other point process that is homogeneous and isotropic could be used at the very beginning. Such an initial point process could then consider interactions between particles in the surroundings of an absorbing medium, in contrast to the CSA model[1] which takes into account nearest neighbour relations *on* an adsorbing medium.

3. THE DEAD LEAVES MODEL

A famous, homogeneous and isotropic, model in spatial statistics is the dead leaves model.[4, 5, 9] In two dimensions, it can be obtained by placing disks (with constant or random radii) randomly and uniformly on the plane during the time interval $(-\infty, 0]$ so that (i) subsequent disks may (partially) cover already existing disks and (ii) infinitely many disks are placed in any subset W of the plane with positive area. At time $t=0$ the system is in its time-stationary state and yields a tessellation of the plane as shown in Fig. 1. (The disks can be interpreted as dead leaves and the union of all the disks as a layer of dead leaves.)

In the following we concentrate on an alternative construction of the dead leaves model which is useful for the comparison with the RSA model and Matérn's second hard-core process and which is attractive also for simulation purposes.[9, 12] At time $t=0$ the space is empty. Then, step by step, disks are randomly dropped onto the plane, so that parts of a new disk that intersect already existing disks become invisible. (Such a simulation resembles watching falling leaves from below.) The incidence times, the locations, and the radii of the disks are given by the system of marked points $(x; t, r)$ already defined in Section 2. For practical simulation it is important to note that whenever a (finite) sampling window is completely covered by disks, the simulation result will not change when further disks are added in the background, and the simulation can be stopped.[9, 12]

A disk is called "intact" if it is not (partially) covered by any other disk. In the case of the alternative construction, a disk $B(x, r)$ placed at time t is intact if and only if there is no disk $B(x', r')$ that has appeared at time $t' < t$ such that $|x' - x| \leqslant r + r'$, i.e., if it is a disk retained in the very first step (2.2) of the construction in Section 2. Thus, the system of intact disks of the dead leaves model is a homogeneous and isotropic subsystem of the RSA model. Consequently, its coverage is smaller than the jamming limit coverage of the RSA model. Nevertheless, this system may be of interest since it allows for analytical calculations; at least in some particular cases formulae can be given for the pair correlation function and the coverage. The latter is 1/4 if the disks have fixed radius, or, more generally, if the "leaves" are convex and their shape, orientation, and finite size are fixed.[13] (The coverage is smaller for variable objects; the case of isotropic line segments is discussed in Penttinen and Stoyan.[14])

Assume that Φ is the homogeneous planar marked Poisson process of intensity λ whose marks m are uniformly distributed on $[0, 1]$. Let h be a positive hard core distance. Then Matérn's second hard-core process[6] consists of all the unmarked points x in Φ where the disk $B(x, h)$ does not contain any point of Φ with a mark smaller than m. As a $\lambda \to \infty$, Matérn's second hard-core process coincides with the system of intact disks of the dead leaves model if the disks have constant diameter h.

If ϱ is the mean number of disk centres per unit area, the pair correlation function $g(r)$ is such that the probability of having a disk centre in each of the two infinitesimal areas dA_1 and dA_2 a distance r apart equals $g(r)\,\varrho^2\,dA_1\,dA_2$. For the particular case of disks with constant diameter, the pair correlation function for the intact disks of the dead leaves model is obtained from the formula for the product density of Matérn's second hard-core process in Stoyan *et al.*,[8] p. 164, as $\lambda \to \infty$. For disks of diameter $h = 1$, one gets

$$g(r) = \begin{cases} 2\pi/\Gamma(r) & \text{for} \quad r \geqslant 1 \\ 0 & \text{otherwise} \end{cases}$$

where $\Gamma(r)$ equals the area of the union of two disks that have unit radius and whose centres are a distance r apart.

4. RELATIONSHIP BETWEEN PROPOSAL AND RESULTING DISTRIBUTION

Whenever the RSA model is a candidate for modelling a given system of non-intersecting disks with variable radii, the proposal distribution of the radii has to be determined. By no means the resulting distribution, i.e., the distribution of the radii of the visible disks should be used, since this would lead to disks that are too small. Unfortunately, the relationship between the observable resulting distribution and the proposal distribution is very complicated, as the simulations in Meakin and Jullien[3, 15] indicate. However, the special case of the two-point distribution, which has already been studied by several authors for different purposes,[3, 16] allows for direct inference.

Exemplarily, the case where the radii equal 1 and 2 is considered in the following. Denote by f the probability of taking a 1-disk in the proposal distribution. Let $f_1(f)$ be the frequency of observing a 1-disk in the jamming limit. Clearly, $f_1(0) = 0$, $f_1(1) = 1$, and $f_1(f) \geqslant f$. Figure 2 shows the function $f_1(f)$ for $0 < f \leqslant 1$. The values were obtained by simulation which was

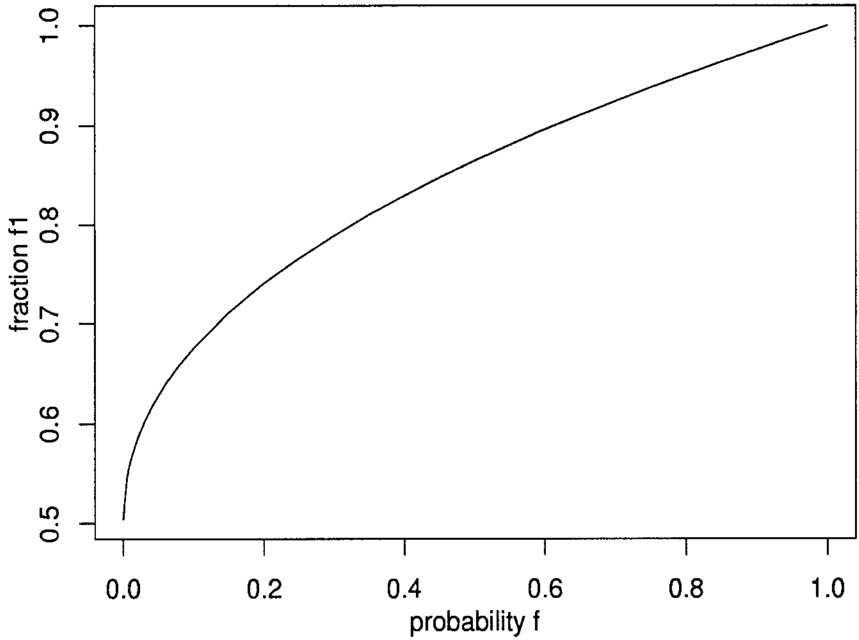

Fig. 2. The fraction f_1 of 1-disks as a function of the probability f of attempting a 1-disk in the two-radii RSA model with radius ratio 1:2.

carried out similarly as in Meakin and Jullien[3] and Wang,[17] based on 25 simulations of the RSA model on a 1000×1000 square. For $f_1(0+)$, the simulations yielded a value a bit greater than 0.5, namely $f_1(0+) = 0.5041 \pm 0.0002$. (If the ratio of the radii equals 1:3 and 1:4 then $f_1(0+)$ equals 0.7434 ± 0.0002 and 0.8499 ± 0.0002, respectively.)

Figure 2 can be used for solving the statistical problem mentioned in the introduction. Assume that an RSA model is observed where the disks have radii 1 and 2. While the 1-disk probability f belongs to the proposal distribution and is unknown, the frequency of 1-disks in the pattern can be observed. The latter can be considered as an empirical value of $f_1(f)$ and can be used to estimate f by means of Fig. 2.

5. VARIABILITY OF DISK POSITIONS

Usually, the variability of point systems (like the variability of the disk centres of the RSA model) is characterised by means of the pair correlation function $g(r)$.[18, 19] Figure 3 shows the pair correlation function for the RSA model as discussed in Section 4, where $f = 0.5$. This curve (as well as

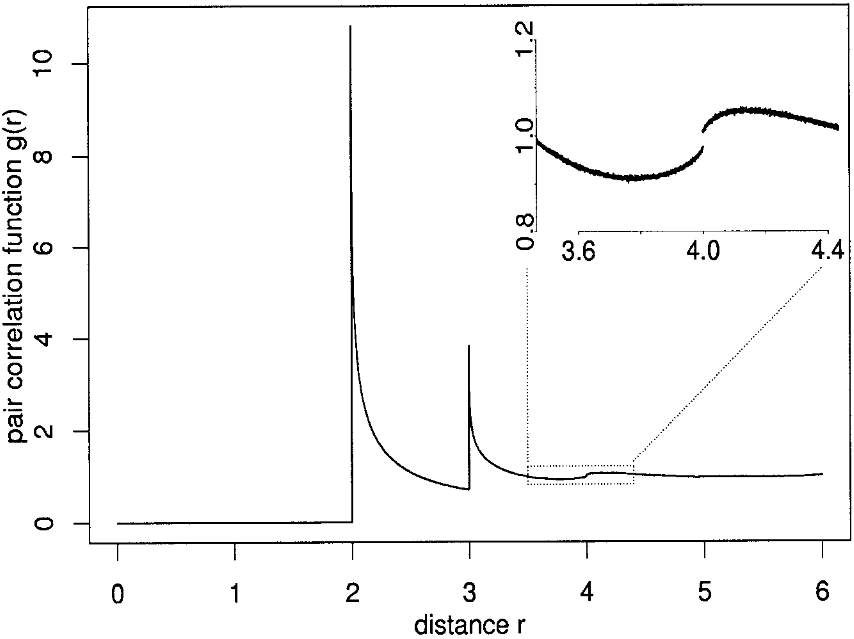

Fig. 3. The pair correlation function $g(r)$ of the point process of disk centres for the two-radii RSA model with $f = 0.5$ and radii 1 and 2. Each dot of the curve in the top right corner is a point estimate of the curve; the abscissa difference between consecutive points equals 0.0003.

the curves for $p_{ij}(r)$ below) is based on 1200 samples on a 1500×1500 square. It has cusps at $r = 2$ and $r = 3$ which correspond to pairs of disks with radius 1 and to pairs of 1- and 2-disks, respectively. Similar to the pair correlation function of the RSA model with monodisperse disks of radius 1,[18] the pair correlation function exhibits a minimum to the left of $r = 4$ and a local maximum to its right. However, in the binary case there is a jump discontinuity at $r = 4$, cf. Figs. 3 and 4.

The mutual positions of 1- and 2-disks can be characterised by functions $g_{ij}(r)$ that describe the probability of finding a pair of points at distance r where one of the points is the centre of an i-disk and the other one that of a j-disk, for i, $j = 1, 2$. The functions g_{ij} give an integrated information about the spatial structure of the unmarked point process of disk centres (which is also described by $g(r)$) and the interactions of points of class i with points of class j. The effect of such a mixing up is illustrated by Fig. 4 which shows the function $g_{11}(r)$ for $f = 0.5$ and radius ratio 1:2, and the function $g(r)$ in the monodisperse case with radius 1. Both curves show qualitatively a similar behaviour. Especially, they do not show that the

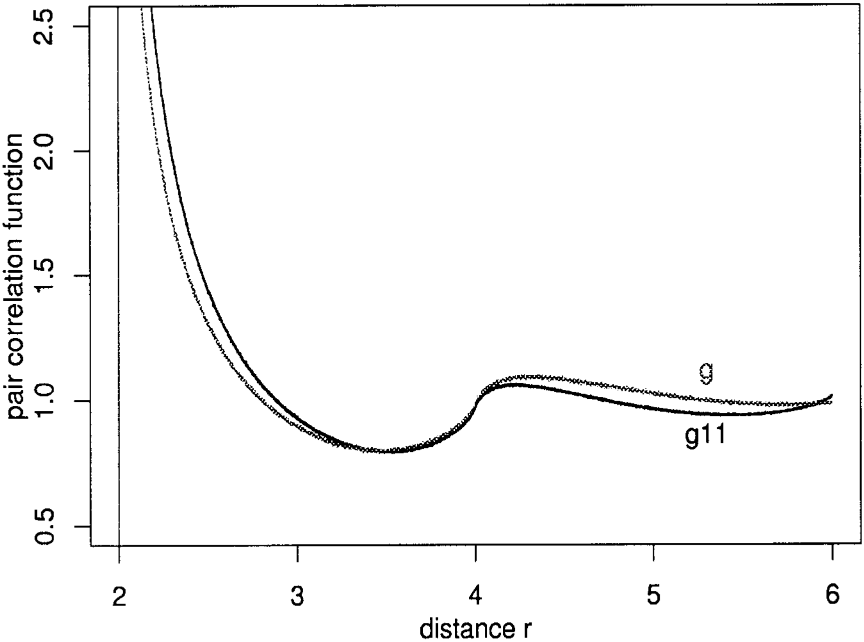

Fig. 4. The pair correlation function $g(r)$ of a monodisperse distribution with radius 1 and the function $g_{11}(r)$ of a binary distribution with $f = 0.5$ and radii 1 and 2.

interaction properties of the 1-disks change dramatically at $r = 3$ in the binary case.

Unlike the function $g_{ij}(r)$ the "mark connection function" $p_{ij}(r)$ is able to unfold information about the interaction of points of the classes i and j. It equals the *conditional* probability of having an i-disk and a j-disk centred at two given points at distance r, under the condition that there are indeed disk centres at both points.[20] If p_k is the fraction of k-disks in the resulting distribution, then

$$p_{ij}(r) = p_i p_j g_{ij}(r)/g(r) \qquad \text{for} \quad r > 0 \qquad \text{with} \quad g(r) \geqslant 0$$

Figure 5 shows the functions $p_{11}(r)$, $p_{12}(r)$ and $p_{22}(r)$, which are defined only for $r > 2$ and satisfy $p_{11}(r) + p_{12}(r) + p_{22}(r) = 1$ for any $r > 2$. As $r \to \infty$, the three functions tend to $p_i^2 = (f_1(0.5))^2$, $2p_1 p_2 = 2f_1(0.5)$ $(1 - f_1(0.5))$, and $p_2^2 = (1 - f_1(0.5))^2$, respectively. These limits result from the asymptotic independence of the radii 1 and 2 for large distances r. For $r < 3$ only 1-1 pairs are possible, and therefore $p_{11}(r) = 1$ for those r. At $r = 3$ and $r = 4$ there are jumps since 1-2 pairs respectively 2-2 pairs cannot occur for smaller distances.

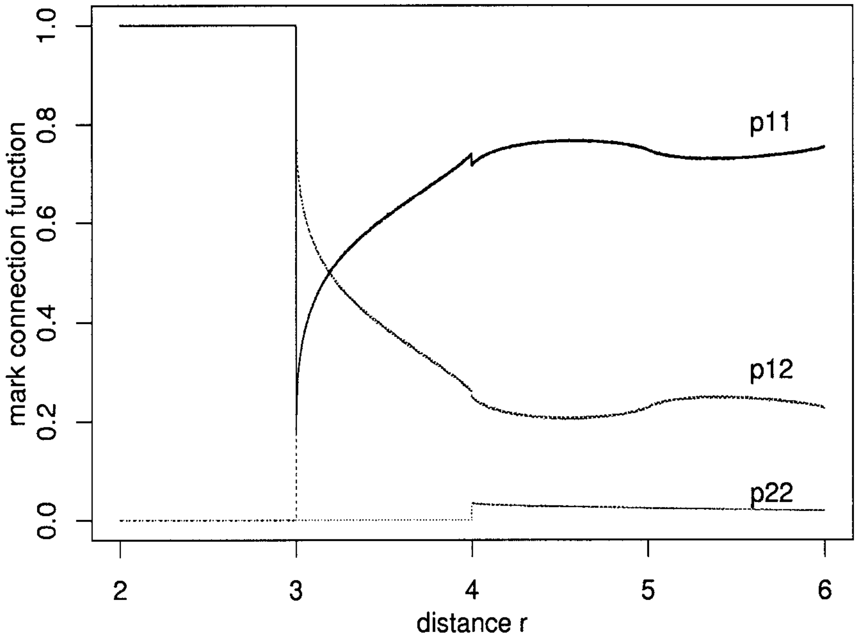

Fig. 5. The functions $p_{ij}(r)$ for the two-radii RSA model with $f = 0.5$ and radii 1 and 2.

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