A novel FastRSA algorithm: Statistical properties and evolution of microstructure
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Abstract

While the Random Sequential Adsorption (RSA) process for the generation of 2D geometries containing discrete entities has been extensively studied, both in terms of numerical simulations and in terms of its statistical properties, all the mechanisms involved are not fully understood, especially in dense configurations of elongated particles. This is mainly due to the very slow asymptotic approach to high packing configurations, especially when highly elongated particles are involved, which makes the creation of such configurations a time consuming task. For the estimation of the statistical properties of such configurations we therefore have to resort to extrapolations that do not always give accurate results. In this work we reveal the interaction of the mechanisms that come into play in the RSA process. We specifically show that the overall result of an RSA process is the summary outcome of these interwoven mechanisms, namely those involving the formation and destruction of Particle Area, Overlap Area and Influence Area – terms which we introduce and define in this work – resulting in a behavior that often appears counter-intuitive. We also show the shift of their importance as the particle aspect ratio \(\alpha\) varies and explain how nematic structures are created when high aspect ratio particles are involved as well as the mechanisms behind their appearance. Following this, we propose a new algorithm for the process of random sequential adsorption (named FastRSA) which is capable of creating very high count configurations through all the range of particle aspect ratios and which follows Feder’s law with a \(\theta \sim \tau^{-1/2}\) behavior instead of the \(\theta \sim \tau^{-1/3}\) of the classic approach, where \(\tau\) is the number of attempts to place a particle and \(\theta\) is the degree of packing. We show how the new algorithm can be coupled with the classic RSA approach and point out the benefits of such a coupling. Use of the FastRSA algorithm has enabled us to study the evolution of the extent of packing using actual geometries, without the need to resolve to extrapolations and assumptions. For the case of highly elongated particles, this is the first time in our knowledge that estimations of maximum packing from actual configurations near the jamming limit have been obtained.

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1. Introduction

Random Sequential Adsorption (RSA) is a process by which geometrical entities (particles) are randomly and sequentially added on a substrate surface \(A\) with the requirements that (i) no repositioning of an entity occurs after it has been placed and (ii) no overlap between entities occurs. The RSA process has been adequately described in the

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literature [1–4] for various cases and in 1D, 2D and 3D geometries; it has been extensively used for the modeling of chemical reactions [5], in biology [6,7] and in a variety of problems involving packing and packing estimations. In addition, much work has been done in studying various geometries [8] or in shedding light to the statistical background [9] and spatial properties [10] of RSA configurations. Recently, new methods for more precise estimations of maximum packing have been introduced [11,12]. In brief, the process of creating an RSA particle configuration involves the selection of one random point $C_p$ with coordinates $(C_x$ and $C_y$) and a random angle $\omega$ and the subsequent placement of a particle at that point with the selected orientation angle. If the thus placed particle overlaps with any of the previously deposited particles, this attempt $(C_x, C_y, \omega)$ is rejected, the newly added particle is removed and the process starts again with a different set of random coordinates and angle. The process stops when no more particles can be added, at which point the configuration is considered to have reached the ‘jamming limit $\theta_j$’ which is defined as the area occupied by particles divided by the total available area at the end of the RSA process. In practice, the RSA process stops after a predefined number of placement attempts (or elapsed CPU time) is reached or if no new particle can be added after a predefined number of attempts. A result of the statistical nature of the RSA process and, especially, of the fact that it follows Feder’s law [4], namely that the achieved packing scales with a (negative) power of the number of placement attempts, is that the difficulty of placing additional particles increases dramatically as the RSA process evolves. As a result, it is practically impossible to generate high density and high count configurations, especially when elongated particles are used such as, for example, in fibrous composites [13]. One consequence of this is that multi-particle, direct simulations (such as transport phenomena or mechanics) in heterogeneous materials at very high packing densities have to-date been impossible to carry out, even though there are numerical schemes which could make such simulations possible [14]. An additional consequence of this state of affairs is that existing estimates of the ultimate packing densities achieved by an RSA process are not the result of actual simulations that have actually reached the jamming limit, but are instead extrapolations and therefore subject to uncertainty. This is especially true for elongated particles, such as rod-like macromolecules, viruses, fibers etc. Earlier computations have shown [15–17] that, using the classic RSA approach the evolution of packing scales with a power of ($\tau$), typically $\theta_j \sim \tau^{-1/2}$ where $\tau$ is the number of (successful or not) placement attempts. Swendsen [2] has proposed that the exponent in the above relationship between packing and placement attempts is $(-1/d)$, where $(d)$ is the dimensionality of the problem. In this work we will try to clarify these results and to provide some insight into observed discrepancies.

We deal with square and elongated rectangular particles of length $l$ and width $t$ and various aspect ratios $\alpha = l/t$ ranging from 1 (randomly oriented squares) to more than 1000 (fiber-like elongated rectangles) and develop a new algorithm for the sequential adsorption process (named FastRSA in the rest of the manuscript). This algorithm is shown to offer a significant improvement in speed and effectiveness over the traditional RSA, by operating on the full scale of probabilities at any given candidate point. It belongs to the family of algorithms that are used to find the visibility polygon from a point, a significant problem in computer graphics [18], where such algorithms are used for faster rendering of 3D scenes. We show that the proposed FastRSA algorithm achieves significantly higher packing densities at realistic execution times and also that it consistent with Swendsen’s rule [2] since it follows Feder’s law with $\theta_j \sim \tau^{-1/2}$.

2. Geometrical definitions and formulation

Consider one particle (as part of a multi-particle assembly) placed at point $C$ with its long axis at an angle $\omega_e$ and a candidate particle at point $C_p$ with its long axis forming an angle $\omega_n$ with the horizontal, as shown in Fig. 1. Any attempt to place a new particle in such an assembly will be unsuccessful if the new particle overlaps with pre-existing ones. For a given set of $C$ and $C_p$ and angles $\omega_e$ and $\omega_n$, existence of such overlaps depends on the relative position and the relative angle $\omega = \omega_e - \omega_n$ between the new particle (subscript n) and the pre-existing one (subscript e). If we plot the space comprised of all points where a new particle cannot be placed with respect to a pre-existing particle, at all levels of the relative angle $\omega = \omega_e - \omega_n$, we form the highlighted polygonal areas shown in Fig. 1. These polygons are the result of the Minkowski Sum [19] between the pre-existing particle $P$ and the new particle $B_{\omega_n}$. The Minkowski Sum is defined as the locus of points satisfying

$$P + B := \{p + b: p \in P, b \in B\} \quad (1)$$

and can be described briefly as the polygon that is created if we define as $P$ an already placed particle and $B$ a newly arrived particle with its center at a random point $C_p$. For every pre-existing particle $P$ we can define the corresponding set of polygons $M$ for $\omega \in [0, \pi]$ (due to symmetry, the polygons at angle $\omega$ are equal to $-\omega$), as:

$$M = \sum_{\omega=0}^{\omega=\pi} \{P + B_{\omega} := \{p + b: p \in P, b \in B_{\omega}\}\} \quad (2)$$

where $P$ is an already placed particle and $B_{\omega}$ is a new particle at point $C_p$ and relative angle $\omega$.

By using the above definitions we can now define the probability of successfully placing a new particle at point $C_p$ with a relative angle $\omega$ as

$$p = 1 - \frac{|M'|}{|M|} \quad (3)$$
Fig. 1. Placement of new particles of relative angle $\omega$ and their relation to the corresponding Minkowski polygon. Particles with center point inside the Minkowski polygon intersect with the pre-existing particle while new particles with their center point outside do not intersect. (a) $\omega = 0^\circ$, (b) $\omega = 45^\circ$, (c) $\omega = 90^\circ$.

where $M$ is the set calculated from Eq. (2) and $M'$ is the subset of Minkowski polygons that $C_p$ resides in. Using the above result, we can define the following three areas that are formed around a particle as

(i) The Overlap Area ($A_O$) as the area of points where $p = 0$,
(ii) The Influence Area ($A_I$) as the area where $0 < p < 1$ and
(iii) The Particle Area ($A_P$) as the area inside the particle for which also $p = 0$.

We can define geometrically the above areas by observing that the Overlap Area ($A_O$) is the area that extends from the boundaries of the particle to a distance $\frac{t}{2}$ and the Influence Area ($A_I$) extends from the boundary of the overlap area up to a distance $R = \frac{1}{2}\sqrt{l^2 + t^2}$ from the particle’s boundaries (Fig. 2a). We see that every point in the Overlap Area belongs to every polygon that is created as the result of the Minkowski sum, that is $\forall B_\omega, C_p \in B_\omega$ as seen in Fig. 2b. This is also easily derived geometrically since every point $C_p$ that has a distance smaller than $\frac{t}{2}$ from a pre-existing particle will inevitably collide with it. Similarly, the points in the Influence Area belong to some but not all the polygons of set $M$.

From the above definitions it becomes easy to draw a conclusion for the acceptance or rejection of a candidate point for the placement of a new particle; if the point $C_p$ lies within the Minkowski polygon that is constructed from a pre-existing particle ($P$) and a candidate particle at point $C_p$ and angle $\omega$ ($B_\omega$), then this candidate point is rejected for particle placement using this angle. In the case however that the point resides in the Influence Area, successful particle placement may be possible using a different angle $\omega'$. Such a calculation, even though intuitively easy, is of little practical use due to the sheer amount of geometrical calculations that need to be performed for every $\omega \in [0, \pi]$. We can however generate a much faster implementation if we notice that we do not have to test against all the combinations of angles and polygons but instead we can check only for specific key angles that are formed between the new center point and existing particles. We describe this procedure in the following section and we see that by calculating a visibility polygon from point $C_p$ we can calculate the available arcs that allow for the placement of a newly arrived particle.

3. Computational

Visibility polygon algorithms consist of two phases - a broad phase and a narrow phase [18]. The broad phase is used to massively filter out elements – in our case particles – which are away from the neighborhood of the point of interest $C_p$; this is a first elementary step in speeding up calculations. During the narrow phase, we only deal with the elements that are near the area of interest. In the context of the FastRSA algorithm, we use a variation of such a procedure for the precise estimation of a visibility circle of radius $R$ and the arc segments over which an attempt to place a particle at $C_p$ will be successful with a probability $p = 1$. In its implementation, the algorithm computes and returns a pair of elements; the first element is the probability of placement ($p'$) which is 1 if a particle can be placed and 0 if a particle cannot be placed. The second element is a list containing the boundaries of the available arc segments over which the placement of this particle is possible; each element of the list consists of two angle values $(\omega_1, \omega_2)$ defining the edges of these arc segments. In the case where a particle cannot be placed this list is empty. This is the main difference between FastRSA and the classic approach (named ClassicRSA in the text) – the latter will use brute force to find an acceptable angle for the placement of a particle at each randomly chosen $C_p$. 
3.1. FastRSA calculation steps

I Pre-processing steps
   I.1 A uniform grid is constructed which partitions the available area in grid cells. This way the majority of particles is excluded from consideration since they belong in cells that are away from the point of interest.
   I.2 Pre-calculate numerical values that are to be used extensively during the course of the algorithm.

Following pre-processing, the main loop of the algorithm is executed, in which we aim to place a particle in a randomly selected point $C_p$. The loop runs as long as the desired packing is achieved or a specified number of tries has been reached. The following steps are followed for every placement trial.

II Broad phase
   Executed for every particle that is in the grid cells surrounding the point $C_p$.
   II.1 Check against the AABB (Axis Aligned Bounding Boxes) and a square AABB of size $\sqrt{l^2 + t^2}$ where $(l)$ is the length and $(t)$ the thickness of the particle, centered at $C_p$. If they overlap then we continue with this particle to the next step. Otherwise the next particle in the grid cells is selected and we return to step II.1.
   II.2 Check against the distance of the nearest point of the previously selected rectangle and point $C_p$. If the distance is smaller than $\frac{1}{2}\sqrt{l^2 + t^2}$ then we add this particle to the list of particles that have to be considered in the narrow phase, otherwise we continue with the next particle from step II.1.

III Narrow Phase
   If the list of particles for consideration is empty then the combination of point $C_p$ and angle $\omega$ can be used without further calculations. If the list is not empty then for every particle in the list we execute the following:
   III.1 Find the intersection points of a circle located at $C_p$ and having diameter $\sqrt{l^2 + t^2}$ and the segments that make the particle from the previous step (Fig. 3a).
   III.2 Create the circles that are centered in the intersection points $(C_{p1}, C_{p2})$ with radius $\frac{l}{2}$ (Fig. 3b). This is to avoid overlap and the reasoning for this is obvious from Fig. 3c.
   III.3 Find the tangent points $T_1, T_2$ of the segments originating from point $C_p$ to the previously formed circles (Fig. 3c).
   III.4 Exclude the arc that is formed from the points $T_1, C_p, T_2$ from the available arc spaces that we can use (Fig. 3d).

IV End Phase
   IV.1 If after the above subtraction the available arc space is not empty then (a) an angle $\omega$ that falls within the available arc space is selected randomly, (b) a new particle is placed after a final collision check is made and (c) we return to step I.
Fig. 3. FastRSA algorithm steps. (a) Calculation of points $P_1, P_2$, (b) Calculation of circles of radius $t/2$, (c) Calculation of points $T_1, T_2$, (d) Calculation of the resulting arc. In this particular case, the arc corresponding to the gray part of the circle corresponds to admissible angles for the placement of a particle at point $C_p$. Duet to symmetry the mirror arc is also excluded.

IV.2 If the resulting available arc space is empty we reject the point $P$, select a different point and we return to step II.1.

The geometrical calculations involved in the above algorithm may appear tedious, however they offer two significant advantages. The first is that they work for every $\alpha$ and they are not limited to square or near square particles. The second is that they do not require the creation and handling of large arrays that would limit their usage to small numbers of particles. In fact the algorithm works extremely well and produces configurations within 2 percentage points of the jamming limit for tenths of millions particles in reasonable CPU times with today's standards (less than an hour for $\sim$20 million particles of $\alpha = 1000$ on an i7 Desktop PC with 8 Gb of RAM).

From the above we can see that the probability $p$ of placing a particle defined earlier in Eq. (3) can be also expressed as:

$$p_p = \frac{\text{Available Arc Space}}{2\pi} = \frac{[\omega_1, \omega_2]_1 \cup \cdots \cup [\omega_1, \omega_2]_v}{2\pi}$$

(4)
4. Results and discussion

4.1. Statistical and geometrical properties

We see from the previous definitions that each particle creates a probability field around it which forbids other particles to be placed, at certain angles, within the surrounding space. The value of this probability field at every point \( C_p \) around a particle can be calculated using Eqs. (4) and (5). Examples of this probability field, as computed by FastRSA, around...
particles of various aspect ratios ($\alpha$), is shown in Fig. 5. At points inside the particle and inside the Overlap Area the probability value is 0 and at points outside the Influence Area is always 1.

In Fig. 6 we can see the computed probability field in multi-particle configurations of various aspect ratios ($\alpha$). It is interesting to note the unpredictable nature of the evolution of the Interaction Area $A_I$. As $\alpha$ increases the areas that are rendered un-useful for particle placement due to interactions of each particle’s Influence Area become progressively more important and constitute the limiting factor in achieving a high packing.

Over the entire area $A$, the total probabilities can be defined as:

\[
\text{Total } p_A = \int_A p \, dA
\]
\[
\text{Total } p^*_A = \int_A p^* \, dA
\]  

(6)

Obviously, each of these quantities changes as the RSA process evolves and each can be calculated for any ($\alpha$) of interest. This is illustrated in Fig. 7, which shows the evolution of the total probability, as function of the (evolving) packing density at various particle aspect ratios ($\alpha$). Evidently, this probability drops sharply as the RSA process evolves and maximum packing is approached. We also observe that $p^*$ is orders of magnitude larger than $p$, especially when particles of high
Fig. 6. Contour plot of probability $p$ and $p^*$ for placing a particle in a pre-existing configuration at two values of the particle aspect ratio $\alpha$. (a) $p$ plot for $\alpha = 2$. (b) $p^*$ plot for $\alpha = 2$. (c) $p$ plot for $\alpha = 10$. (d) $p^*$ plot for $\alpha = 10$. It is easily seen that $p^*$ creates a binary probability field where values are either 1 (gray areas in Fig. 6b and d) or 0 (white areas) while $p$ creates a field that varies from 0 to 1 as the distance from a particle increases.

aspect ratio are involved. From Fig. 7 it becomes clearer why FastRSA is much faster than ClassicRSA, especially in the case of elongated particles; since FastRSA utilizes $p^*$, which is several orders of magnitude larger than $p$, for a given area $A$, it can successfully place particles at a much faster rate.

In summary, the efficacy of a particle placement process depends on the available area. We show that, in turn, the evolution of this area depends on three mechanisms. These mechanisms remove available space from the total area $A$, and as a result, as new particles are added the total available space decreases. The interaction of these mechanisms gives valuable insights about the manner in which Random Sequential Adsorption works and is described in the following.

4.1.1. Particle area ($A_p$)

The first and obvious mechanism is the placement of the particles themselves. As each particle is added, an area equal to the area of a particle $(l \cdot t)$ is removed from $A$. This area obviously evolves linearly with the addition of particles as $A_p = N_r \cdot l \cdot t$ where $N_r$ is the number of added particles at attempt $\tau$. 
Fig. 7. Plot of $p$ and $p^*$ onto an area $A$ as particles are added. We can see that $p^*$ is orders of magnitude larger at the same packing $\theta$ especially as $\alpha$ increases.

Fig. 8. Detail view of the evolution of Overlapping areas between two steps. Overlap Area is shown in white. With the addition of a new particle (solid gray in image (b)), area that was characterized as Overlap Area (white) switches to Particle Area (black). Also the particle removes area from $A$ and transforms it into Overlap Area (dotted area) and Particle Area (colored gray).

4.1.2. Overlap area ($A_0$)

The second mechanism depends on the way the Overlap Area that surrounds the particles evolves as additional particles are placed. As discussed earlier in Section 2 as new particles are added the area surrounding each particle at a distance up to $\frac{d}{2}$ from the particle sides is rendered unusable because any particle with a center point inside this area will overlap with the previously placed particle, irrespective of its orientation. However this area can overlap with similar areas of surrounding particles as it can be seen in Fig. 8. As the process evolves, the extent of these overlaps increases and with the addition of new particles the Overlap Area surrounding an existing set of particles is reduced by being occupied by newly-added particles while also new Overlap Area is generated with the addition of each new particle. The process is illustrated in Figs. 8 and 9.

4.1.3. Interaction area and influence area

The third mechanism that comes into play is the evolution of Interaction Area and Influence Area. Interaction Area consists of the points that cannot be used for placement of particle centers because no particle can be added due to geometrical constraints. As particles are placed onto a surface, new particles cannot be added in various points around them because they cannot fit at any orientation. These areas are formed outside the Overlap Area of pre-existing particles but from the interaction of their Influence Areas ($A_I$). For example, a particle might allow the placement of a new particle
at a point \( C_p \) that falls inside its Influence Area, with an angle \( \omega \), but the same point might belong to the Influence Area of other particles which might not allow the placement of a new particle at this specific angle. As a result no particles can be placed in this point at any angle \( 0^\circ < \omega < 180^\circ \). From the above we define the Interaction Area as the area in which no particles can be added due to constraints that are imposed from the Influence Areas of surrounding particles. The evolution of Interaction Areas can be seen in Fig. 10. For elongated particles these areas tend to become extremely large and occupy a big part of the total area \( A \) even if the area of individual particles is small.

4.1.4. Total available surface area

From the previous discussion, it is evident that placement of a particle at a point \( C_p \) depends on where this point resides in respect to the other, pre-existing particles. If the point \( C_p \) falls within a particle or in its Overlap Area it cannot be selected for placement of a new particle. If a point falls outside the Areas of Influence of the existing particles then a new particle can be placed in this point at any angle (\( \omega \)). However if it falls within the Areas of Influence of nearby particles, the algorithm described above will screen potential candidate points. From the above we can define the Available Space (\( A_s \)) for the addition of a new particle as

\[
A_s = A - A_P - A_O - A_I
\]

The total Available Space (\( A_s \)) is the result of the interaction of the above three mechanisms, which are not evolving monotonically as the RSA process unfolds, as it can be seen in Fig. 11. We see that for small \( \alpha \) the mechanism that subtracts large areas from \( A \) is the evolution of Overlap Areas while for large \( \alpha \) the limiting mechanism becomes the evolution of Interaction Area. This observation is at the heart of the well-known differences in performance in terms of achieved packing of RSA algorithms when particles of small or large (\( \alpha \)) are used.

4.2. Particles of high aspect ratio and the formation of nematic structures

From the definitions of Overlap Area and Influence Area we see that as \( \alpha \) increases the Overlap Area around each particles decreases since it is proportional to \( \frac{1}{\alpha^2} \). On the contrary, the Influence Area is proportional to \( l \) and therefore increases. In the case of elongated particles \( (l \gg t, \alpha \gg 1) \) we see (Fig. 11) that the main mechanism that influences \( A_s \) is the evolution of the Interaction Area. Since Interaction Areas occupy a big part of the total area \( A \), new particles can be added only in close alignment with pre-existing ones. This can be easily seen in Fig. 10 where it becomes obvious that the only possible way for new particles to be placed is in close correlation with their angles and thus, the only mechanism that could allow the addition of new particles in dense configurations is the formation of nematic structures. It appears therefore that for elongated particles the creation of nematic structures is a fundamental consequence of the RSA process and it emerges as packing increases. It also shows us the importance of the initial steps of the adsorption process, since the particles that arrive first create the scaffold which guides the addition of new ones as it can be easily seen in Fig. 12. We can also note that, since the areas between existing particles become smaller, the available angles that can be used are limited to a very narrow range. Obviously, the ClassicRSA, by trying to randomly select, from the entire space \( 0 < \omega < \pi \), an angle \( \omega \) that has to fall within a very limited range, requires many attempts for the same point \( C_p \) before successfully picking one that would allow the placement of a particle.
Fig. 10. (a), (b). Evolution of Interaction Areas (shown as white) between time steps. The probability of placing a particle \( p^* \) is zero in these areas. Areas where \( p^* = 1 \) are shown as red. The potential for the formation of nematic structures, with locally aligned particles is clearly visible at latter stages of the process. (c), (d) detail of areas between particles where \( p^* = 0 \) (in white color) at \( \alpha = 512 \).

4.3. Hybrid mode

We have shown that as particles are deposited within an area \( A \) they change the properties of the surrounding space by creating a probability field around them. We define as “polarized space” in terms of an angle \( \omega \) the set of points at which the probability \( p \) is \( 0 < p < 1 \). If we plot this field not in terms of \( p \) and \( p^* \) as in Fig. 6 but in terms of relative angles \( \omega \) we can see how the total area gets polarized in these specific angles during the RSA process (Fig. 13). As new particles are deposited to a configuration their corresponding Minkowski Polygons at various angles \( \omega (B_\omega) \) occupy a progressively larger area of \( A \). As we explained earlier in Section 2, if a point lies within a Minkowski polygon \( B_\omega \) of a particle, it cannot be used for the placement of a new particle with this specific angle \( \omega \). Therefore as particles are added, the area \( A \) has less space left for specific angles \( \omega \) and eventually there is no more area left within \( A \) for these specific angles.

Plotting the evolution of the area that remains un-polarized for various angles \( \omega \) during the adsorption process we get the images shown in Fig. 13, in which the affected area around each particle is shown as shaded.
Fig. 11. Evolution of area coverage mechanisms for various $\alpha$. In the Y axis we can see the area fraction that is characterized as Interaction Area (left) and Overlap Area (right).

Fig. 12. (a), (b). Evolution of alignment of particles between time steps as packing increases. By comparing the two Figures it is easy to see that the newly arrived particles in Fig. b (thin lines) are forced to align parallel to the pre-existing particles (bold lines).

Using this approach we can define the ‘Freezing Point’ at a relative angle $\omega$ ($FP_\omega$) as the packing fraction ($\theta_\omega$) at which a specific relative angle $\omega$ cannot be used anymore for placement of particles. This means that no new particles can be entered at any point $C_\omega$ within the Area $A$ at angle $\omega$; the already existing particles will not allow this, since all area $A$ is occupied by the Minkowski polygons $B_\omega$ of these particles. For example, in regards to Fig. 14a, no more particles can be placed at $\omega = \pi/2$ after the packing has exceeded the value of $\theta \equiv 0.3$ - the corresponding unpolarized area fraction has dropped to zero, while at the same packing ($\sim 30\%$) there is a 20\% of the total area available for the placement of particles parallel to pre-existing ones (zero relative angle $\omega$). In Fig. 14b, which corresponds to more slender particles ($\alpha = 100$), the $FP_{45}$ is around $\theta \equiv 0.08$, at which packing there is an over 80\% probability of placing particles aligned with pre-existing ones ($\omega \sim 0$). Obviously, the formation of nematic structures is statistically favored at a much lower packing in the case of slender particles.

We can take advantage of the above observation and introduce a Hybrid Mode in the operation of the FastRSA algorithm; this begins the adsorption process with the ClassicRSA and after a point it switches to the FastRSA algorithm. This way we can have a configuration that has the statistical properties that ClassicRSA up to a user-defined point and the speed of FastRSA afterwards, along with the nematic structures that follow. For example, in the configurations of Fig. 15, switching to the FastRSA after $\theta = 0.5$ and $\theta = 0.2$ (for $\alpha = 10$ and $\alpha = 100$ respectively) will produce configurations that
Fig. 13. Polarization of space around deposited particles. \(a_1 \ldots a_3\): \(\theta = 0^\circ\), \(b_1 \ldots b_3\): \(\theta = 45^\circ\), \(c_1 \ldots c_3\): \(\theta = 90^\circ\). In images \(a_1 \ldots a_3\) we see the space occupied by the Minkowski Polygons of \(\omega = 0^\circ\). In images \(b_1 \ldots b_3\) the Minkowski polygons at \(\omega = 45^\circ\) are shown and in \(c_1 \ldots c_3\) the corresponding polygons at \(\omega = 90^\circ\). The space occupied by the Minkowski polygons is shown shaded.

are equivalent to Classic RSA up to a certain point. With the selection of such a switching point near the jamming limit we could produce configurations with very small statistical differences between the classic RSA approach and FastRSA since the configuration near jamming limit is essentially frozen in all but a small range of relative angles near \(\omega = 0^\circ\) which would lead to the formation of nematic structures using both algorithms. We can see in Fig. 15 the behavior of the hybrid mode in terms of speed and packing achieved.

4.4. Estimation of maximum packing

As a geometrical configuration evolves with the addition of new particles, the FastRSA algorithm, through its ability to calculate \(p\) and \(p^*\) at every point, allows the measurement of various spatial properties such as the amount of Overlap
Area and Interaction Area and thus leads to the accurate calculation of the total Available Space for particle placement $(A_S)$. Using a Monte Carlo sampling procedure to calculate $A_0$ and $A_I$, Available Space $(A_S)$ can be estimated using Eq. (7). Obviously, the jamming limit ($\theta_J$) of a configuration can thus be found as the point at which $A_S$ becomes zero. Extensive simulations (Fig. 16) have shown that the Available Space $(A_S)$ can be described by a function of the following form

$$A_S = M \cdot (\theta - \theta_J)^k \tag{8}$$

where $\theta_J$ is the maximum packing (jamming limit) at the point where the Available Space becomes zero and where $\theta$ is the packing after ($\tau$) attempts, at which point $N_f$ particles have been placed. The fit of Eq. (8) to computational results for various values of the particle aspect ratio $\alpha$ can also be seen in Fig. 16.

We also derive extrapolation predictions for the maximum packing using the assumption that it follows Feder's Law with $\theta_J \sim \tau^{-1/2}$. Results of more than 50 calculations in various $\alpha$ with an accuracy at the 4th decimal digit are shown in Table 1 where we list and compare:

(i) The estimation of maximum packing using FastRSA results based on Feder’s Law using an expression of the form $\theta_J \sim \tau^{-1/2}$

(ii) The estimation of maximum packing using FastRSA results based on Feder’s Law using an expression of the form $\theta_J \sim \tau^{-1/3}$

(iii) The coefficients $M$ and $k$ determined from fitting the computational packing results to Eq. (8).

(iv) The maximum packing as estimated from the extinction of Available Space (Eq. (7))

(v) The actual achieved maximum packing from our simulations with FastRSA algorithm

(vi) Literature estimation of maximum packing based on ClassicRSA [12]; such estimations are only available for $\alpha < 2$. 

Fig. 14. (a), (b). Evolution of polarization areas. (a) $\alpha = 10$, (b) $\alpha = 100$. We can see that as packing increases space ($A$) becomes completely polarized at various relative angles meaning that these angles cannot be used anymore.

Fig. 15. (a), (b). Packing evolution during hybrid mode. (a) $\alpha = 10$, (b) $\alpha = 100$. We see the difference in speed, especially as $\alpha$ increases. In this case simulation ended when a specific CPU time was reached.
Fig. 16. Evolution of the Available Space ($A_s$ as calculated from Eq. (7)) as maximum packing is approached, along with best fit curves of Eq. (8) calculated for various $\alpha$. Total available area at the start of the RSA process is $A = 1$.

![Graph showing Available Space evolution](image)

We can see that using extrapolation (based on the results from FastRSA) with a power-law $\tau^{-1/3}$ systematically overestimates the maximum packing. Instead, a power-law $\tau^{-1/2}$ gives extrapolation predictions which are much closer to the actual packing results we obtained with FastRSA.

4.5. Algorithm performance and results

In the following we present some results on the performance of the FastRSA algorithm. Configurations were created with particles of aspect ratio from 1 to 1024 and involving $N$ particles placed in a periodic square unit cell. For each $\alpha$, $N$ was selected to be high enough in order to achieve a uniform orientation distribution of the particles; it typically was $N > 10^7$ for small $\alpha$ ($< 10$) and it was increasing as $\alpha$ was increasing so that the length $l$ of each particle would be less than $10^{-3}$ of the total area length (typically $N > 10^8$ for $\alpha \geq 100$). Periodic conditions were used to avoid wall effects due to particle alignment and/or particle exclusion at the borders of the unit cell.

In Figs. 17 and 18 we show the maximum packing achieved with FastRSA at various particle aspect ratios. Our algorithm consistently achieves a maximum packing that is almost 2 percentage points higher (depending on $\alpha$) than the values reported in the literature so far. It should be noted that such literature estimates are not actually achieved packings but rather extrapolations based on a power exponent of $(-1/3)$. Besides the difference in the estimated packing values, FastRSA can achieve configurations that are by a margin of less than 1% from the theoretical jamming limit even for large $\alpha$. This can be explained if we consider that the FastRSA works by calculating $p^*$ which can either take the value of 1 or 0. So if a point is selected for particle placement a particle will either be placed, at the first attempt, or not placed,
Fig. 17. Log-log plot of $\theta_j$ vs. $\alpha$. The slope of the line is $-0.20582$.

Fig. 18. Packing achieved by FastRSA for various $\alpha$ as given in Table 1.

depending on whether $p^*$ is one or zero, and no further iterations are required. Classic RSA works not only by trying to find an acceptable point $C_p$ but by also finding, randomly, an acceptable angle over the entire arc space from 0 to 180°. As explained in Section 4, this becomes extremely difficult especially in large $\alpha$ because the formation of nematic structures can allow only for a very limited range of angles at every point. So, in the ClassicRSA algorithm, even if the same point is selected many times, successful particle placement is not certain, while, in contrast, the FastRSA algorithm will place (or not place) a particle at the first attempt. Therefore FastRSA fulfills one the basic assumptions of Swendsen’s analysis in which every point has to have an equal probability to be selected for the placement of a particle. By using Classic RSA two points $C_{p1}$ and $C_{p2}$ that could be used for particle placement ($p^*_1 = p^*_2 = 1$) do not have the same probability $p$ because they also have to pass the random process of selecting an acceptable angle $\omega$ and thus

$$p_1 = p^*_1 \cdot \text{Available Arc space}_{p1} \neq p^*_2 \cdot \text{Available Arc Space}_{p2} = p_2$$

In addition, ClassicRSA will always produce a lower packing because of the nature of the probability field on which it operates. Specifically, as a probability field is created as a result of the placement of pre-existing particles, surrounding points have a probability $p$ that decreases as we move closer to the particles. It is as if the pre-existing particles repel the new ones since the field that has been created around them gives a smaller $p$ and so it becomes harder for ClassicRSA to use the points nearby existing particles. As a result ClassicRSA will fill the empty spaces first, where $p$ is larger and as a consequence bigger interaction and overlapping areas will be created at the initial stages of a RSA configuration which will lead inevitably to a less efficient use of space and a smaller maximum packing.

5. Conclusions

A new RSA algorithm (FastRSA) with the ability to achieve very dense 2D geometrical configurations, with high particle count and using particles of aspect ratio from 1 to 1024, is introduced and studied. Using this algorithm we shed light into the operative mechanisms of an RSA process and explain its behavior, in statistical terms, as we approach the jamming limit. We show that the total packing achieved is affected by the sum of the interaction of the spatial properties of the
area $A$. Specifically we see how the Interaction Area, the Overlap Area and the Particle Area evolve throughout the RSA process and how they interact to produce the final configuration. We can also see the shift of their importance as $\alpha$ varies. We explain how nematic structures are created in dense packings of high aspect ratio particles in an RSA process and we show the main mechanisms behind their appearance. Our results further show that this approach can achieve packings that are $\sim 2$ percentage points higher through all the range of aspect ratios $\alpha$ and we have shown how these high packings can be obtained in much faster computational times. Our algorithm follows Feder’s Law and Swendsen’s conjecture, generating very dense geometrical configurations exhibiting a behavior of $\theta_\tau \sim \tau^{-1/2}$.

References