Connectedness percolation in the random sequential adsorption packings of elongated particles

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Connectedness percolation phenomena in two-dimensional packings of elongated particles (discorectangles) were studied numerically. The packings were produced using random sequential adsorption (RSA) off-lattice model with preferential orientations of particles along a given direction. The partial ordering was characterized by order parameter $S$, with $S = 0$ for completely disordered films (random orientation of particles) and $S = 1$ for completely aligned particles along the horizontal direction $x$. The aspect ratio (length-to-width ratio) for the particles was varied within the range $\varepsilon \in [1; 100]$. Analysis of connectivity was performed assuming a core-shell structure of particles. The value of $S$ affected the structure of packings, formation of long-range connectivity and electrical conductivity behavior. The effects were explained accounting for the competition between the particles’ orientational degrees of freedom and the excluded volume effects. For aligned deposition, the anisotropy in electrical conductivity was observed and the values along alignment direction, $\sigma_x$, were larger than the values in perpendicular direction, $\sigma_y$. The anisotropy in localization of percolation threshold was also observed in finite sized packings, but it disappeared in the limit of infinitely large systems.

I. INTRODUCTION

Random packing of elongated particles onto the plane is a challenging problem that has continuously been the focus of many researchers. The particle shape may affect the packing characteristics (e.g., packing density and coordination numbers) [1][3], the aggregation [4], and gravity- and vibration-induced segregation [5]. A lot of interest to such systems is stimulated by practical problems related with preparation of advanced materials [6][7] and composite films [8][9], filled with elongated nanoparticles, e.g., carbon nanotubes [10] and silicate platelets [11].

For contradiction of random packings, a random sequential adsorption (RSA) model [12][13] is frequently used. In this model, the particles are deposited randomly and sequentially onto a two-dimensional (2D) substrate without overlapping. In the so-called “jamming limit” at saturated coverage concentration $\varphi_j$ no more particles can be adsorbed and the deposition process terminates. The saturated 2D RSA packings for different particle shapes, including disks [14], ellipses [15], squares [16], rectangles [17][18], discorectangles [19][20], polygons [21], sphere dimers, sphere polymers, $k$-mers and extended objects [22][23], and other shapes [24][25] have been studied in details. Particularly, for elongated particles, the nonmonotonic dependencies of values $\varphi_j$ versus the aspect ratio, $\varepsilon$ were observed. Similar dependencies have also been observed for saturated RSA packings of elongated particles in one-dimensional (1D) [28][30] and three-dimensional (3D) [31][33] systems. The appearance of maximums of the jamming concentration was explained by a competition between the effects of orientational degrees of freedom and excluded volume effects [31].

The formation of long-range connectivity is the primary issue to be solved for better understanding of percolation phenomena of core-shell anisotropic particle in random packings. Core-shell composite particles consist of an inner layer of one material (core) and an outer layer of another material (shell). Core-shell particles have already demonstrated promising applications in electrochemical, optical, wearable and gas adsorptive sensors [34], electrode materials [35], polymeric composites [36] and drug delivery applications [37]. Practical significance of the problem is also related description of electrical conductivity behavior of composites filled by elongated core-shell particles, e.g., carbon nanotubes and fibers, metallic nanorods and nanocables, and other core-shell particulates [36][38][47]. In general case, the inner material can covered partially or fully by one or multiple outer layers. By regulation of shell properties, the materials with enhanced optical, electrical, magnetic characteristics, and improved thermal stability or dispersibility can be obtained. For particles with core-shell structures, the electrical conductivity behavior can reflect the effects of particle ordering, packing, connectivity rules and intrinsic properties of the cores, the matrix, and the interface between particles and matrix (shells).

In this paper, we’ll concentrate on the percolation effects in 2D RSA packings of discorectangles. The hard

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core-soft shell structure of particles was assumed and anisotropic packings with preferential orientations of particles along a given direction were considered. The effects of particles aspect ratio, orientation ordering, and packing fraction on electrical conductivity of packings and critical thickness of shells required for a spanning path through the system were evaluated. The rest of the paper is organized as follows. In Sec. II the technical details of the simulations are described and all necessary quantities are defined. In order to provide a better understanding on the precision of calculations some test results are also given. Section III presents our principal findings and discussions. Finally, Section IV concludes this work.

II. COMPUTATIONAL MODEL

Discorectangle represents a rectangle with semicircles at a pair of opposite sides. The discorectangles were randomly and sequentially deposited until they reached the saturated coverage concentration $\varphi_j$. An optimized RSA algorithm, based on tracking of local regions, was used. The aspect ratio (length-to-width ratio) was defined as $\varepsilon = l/d$, where $l$ is the length of the particle and $d$ is its width. The discorectangles with $\varepsilon \in [1; 100]$ were considered.

The degree of orientation was characterized by the order parameter defined as

$$S = \langle \cos 2\theta \rangle,$$

where $\langle \cdot \rangle$ denotes the average, $\theta$ is the angle between the long axis of the particle and the direction of the preferred orientation of the particles ($x$ directions).

For generation of the aligned packings, the orientations of the deposited particles were selected to be uniformly

distributed within some interval such that $-\theta_m \leq \theta \leq \theta_m$, where $\theta_m \leq \pi/2$. For the selected model of deposition the order parameter was calculated as

$$S = \frac{\sin 2\theta_m}{2\theta_m}.$$

Figure 1 shows examples of the packing patterns in jamming state for discorectangles with aspect ratios $\varepsilon = 2$ (a) and $\varepsilon = 10$ (b). For random orientation of particles

FIG. 1. Examples of RSA packings in jamming state for discorectangles with aspect ratios $\varepsilon = 2$ (a) and $\varepsilon = 10$ (b), and at different values of the order parameters: $S = 0$ (random orientation), $S = 0.5$ (partial orientation) and $S = 1$ (complete alignment along the horizontal direction $x$).
For the connectivity analysis, each particle was assumed to be covered by a supporting square mesh of size $m \times m$ ($\theta_m = \pi/2$) we have $S = 0$ and for complete alignment of particles along the horizontal direction $x$ ($\theta_m = 0$) we have $S = 1$. For intermediate values $0 < S < 1$ during the deposition, some particle orientations may be rejected and the real order parameter in the deposit may differs from the preassigned value \cite{29, 30}.

The dimensions of the system under consideration were $L$ along both the horizontal ($x$) and the vertical ($y$) directions, and periodic boundary conditions were applied in both directions. The time was measured using dimensionless time units, $t = n/L^2$, where $n$ is the number of deposition attempts. Figure 2 shows examples of the coverage concentration $\varphi$ versus the deposition time, $t$, for the RSA packing of random ($S = 0$) and perfectly aligned ($S = 1$) discorectangles with aspect ratio $\varepsilon = 4$ at different values of $L/l$. The similar dependencies were observed for other values of $S$ and $\varepsilon$. The scaling tests with $L/l = 16, 32, 64$, and $128$ evidenced the good convergence of the data at $L/l \geq 32$. In the present work, the majority of calculations were done using $L = 32l$ and the jamming coverage was assumed to be reached after deposition time of $t = L^2 \times 10^{10}$.

The analysis of the connectivity was performed assuming a core-shell structure of particles. Each particle was covered by an outer shell with a thickness $\delta d$ (Figure 3a). Any two particles were assumed to be connected when the minimal distance between their hard cores does not exceed the value of $\delta d$. The connectivity analysis was carried out using a list of near-neighbor particles \cite{51}. The minimum (critical) value of the outer shell thickness, $\delta_c$, (hereinafter, shell thickness) required for the formation of spanning clusters along the $x$ or $y$ direction, was evaluated using the Hoshen–Kopelman algorithm \cite{52}.

To calculate the electrical conductivity, $\sigma$, the 2D plane was covered by a supporting square mesh of size $m \times m$. The mesh cells with centers located at core, shell, or pore parts were assumed to have electrical conductivity of $\sigma_c$, $\sigma_s$ and $\sigma_m$, respectively. The large contrasts in electrical conductivities were assumed, $\sigma_c \gg \sigma_s \gg \sigma_m$.

$\sigma_c = 10^{12}$, $\sigma_s = 10^6$ and $\sigma_m = 1$ in arbitrary units. To calculate the electrical conductivity of RRN the Frank–Lobb algorithm based on the Y–Δ transformation was applied \cite{53}. More detailed information on electrical conductivity calculation can be found elsewhere \cite{54, 55}.

For each given value of $\varepsilon$ and $S_0$, the computer experiments were repeated using from 10 to 1000 independent runs. The error bars in the figures correspond to the standard deviation of the mean. When not shown explicitly, they are of the order of the marker size.

![Figure 3](image-url)
III. RESULTS AND DISCUSSION

A. Intrinsic conductivity

Concept of intrinsic conductivity is useful for description of behavior of electrical conductivity in the limiting case of the infinitely diluted system. For randomly aligned and arbitrarily shaped particles with electrical conductivity $\sigma_p$ suspended in continuous medium with electrical conductivity $\sigma_m$, the generalized Maxwell model gives the following virial expansion [56, 57]

$$\frac{\sigma}{\sigma_m} = 1 + [\sigma]\varphi + O(\varphi^2),$$

where

$$[\sigma] = \frac{d \ln (\sigma/\sigma_m)}{d \varphi} \bigg|_{\varphi \to 0},$$

is called the intrinsic conductivity, and $\varphi$ is the coverage concentration.

![Figure 4](image)

**FIG. 4.** Examples of intrinsic conductivities $[\sigma]$ versus the order parameter, $S$. The data are presented along $x$ and $y$ directions for discorectangles with aspect ratios $\varepsilon = 2, 4, 10$ (a). Parameters $[\sigma]_0$, $\kappa$ (See Eq. 3) versus $\varepsilon$ dependencies (b).

![Figure 5](image)

**FIG. 5.** Normalized intrinsic conductivity $[\sigma]/[\sigma]_\infty$ along $x$ and $y$ direction versus the inverse mesh size $1/m$ at different aspect ratios $\varepsilon = 4, 20$, and $S = 1$.

The value of the intrinsic conductivity $[\sigma]$ can depend upon the electrical conductivity contrast $\Delta = \sigma_p/\sigma_m$, the particle’s aspect ratio, $\varepsilon$, order parameter, $S$, and spatial dimension.

Figure 4a demonstrates examples of intrinsic conductivities $[\sigma]$ versus the order parameter, $S$. The data are presented along $x$ and $y$ directions for discorectangles with different aspect ratios $\varepsilon$. These dependencies were obtained using the mesh parameter of $m = 4096$ and 1000 independent runs. Observed $[\sigma]$ versus $S$ were practically linear:

$$[\sigma] = [\sigma]_0 (1 \pm \kappa S),$$

where $[\sigma]_0$ is the intrinsic conductivity for the isotropic system with $S = 0$, $\kappa$ is the anisotropy coefficient, signs + or − correspond to the $x$ or $y$ directions, respectively.

Therefore, intrinsic conductivity $[\sigma]_x$ along alignment direction $x$ exceeded value $[\sigma]_y$ in perpendicular direction hence symmetric behavior with the same anisotropy coefficients $\kappa$ was observed.

Figure 4b presents values of $[\sigma]_0$ and $\kappa$ versus aspect ratio $\varepsilon$. The intrinsic conductivity for the isotropic system $[\sigma]_0$ increased with $\varepsilon$. Note, that the similar behavior was predicted by the theory for randomly aligned ellipses ($S = 0$) [50, 57]

$$[\sigma] = \frac{(\Delta^2 - 1)}{2(1 + \varepsilon \Delta)(\Delta + \varepsilon)}.$$  (6)

For $\Delta \gg 1$, this equation gives (see dashed line in Fig. 4a)

$$[\sigma] = 1 + \frac{1}{2} \left( \varepsilon + \frac{1}{\varepsilon} \right).$$  (7)

The anisotropy coefficient $\kappa$ also increased with $\varepsilon$ reaching to the value of $\kappa = 1$ in the limit $[\varepsilon]_\infty$. 

Figure 6 illustrates the effect of mesh size $m$ on precision of $|σ|$ determination at two values of aspect ratio $ε$. In the limit of the infinite large mesh $m → ∞$. The data evidenced that estimation errors of $|σ|$ increased with increasing of $ε$ and they can reach about 2% for $ε = 20$ and $m = 1024$.

B. Connectivity

For discrete rectangle, the critical shell thicknesses $δ_{c,x}$ and $δ_{c,y}$ correspond to the formation of percolation clusters along the $x$ and $y$ direction, respectively. For isotropic system with $S = 0$, the values of $δ_{c,x}$ and $δ_{c,y}$ coincide, i.e., $δ_{c,x} = δ_{c,y}$. For anisotropic systems with $S ≠ 0$ these value may be different. At fixed value of shell thicknesses, $δ$, the critical coverages $ϕ_{c,x}$ and $ϕ_{c,y}$, required for the formation of percolation clusters along the $x$ and $y$ direction, respectively, can be also defined.

Figure 7 shows the examples of the critical shell thickness $δ_c$ versus inverse systems size $1/L$ at different $ϕ$. Here, $L(= 16l, 32l, 64l, 128l)$ is the size of the system. The data are presented for aspect ratio of $ε = 4$ for completely disordered ($S = 0$, dashed lines) and completely aligned ($S = 1$, solid lines) packings. Increase in $ϕ$ resulted in decrease of $δ_c$ and the minimum values of $δ_c$ were observed at the jamming coverage ($ϕ = ϕ_j ≈ 0.557$ for $ε = 4$). For $S = 0$, the data along the $x$ and $y$ directions almost coincide. However, for finite-sized aligned systems ($S ≠ 0$), the value of $δ_{c,y}$ always exceeded the value of $δ_{c,x}$, and both these values exceeded the value $δ_c$ for isotropic systems. Figure 6 shows the similar examples of the critical coverage $ϕ_c$ versus the value of $L^{-1/ν}$ at different fixed values of shell thickness, $δ$. Here, $ν = 4/3$ is the 2D correlation length percolation exponent [58]. The data on the critical coverage $ϕ_c$ also demonstrated the presence of percolation anisotropy for the finite-sized aligned systems ($S ≠ 0$). The similar percolation anisotropy was observed in finite-sized discrete systems with aligned rods ($k$-mers) and the finite size effect were also more pronounced for systems with aligned rods [54] [59] [60]. Thus, it can be concluded that observed anisotropies in behavior the critical shell thickness, $δ_c$, and critical coverage $ϕ_c$ are the finite size scaling effect and they disappear in the limit of $L → ∞$. Moreover, the scaling behaviors of value $δ_c$ for completely disordered ($S = 0$) and of average value $δ_c = (δ_{c,x} + δ_{c,y})/2$ for aligned ($S ≠ 0$) packings were rather insignificant for $L/l ≥ 32$. Therefore, in the present work, the averaged values $δ_c$ and $ϕ_c$ in both directions were always used and all connectivity analysis tests were done using $L/l = 32$.

Figure 7 and 8 demonstrate the examples of the critical shell thickness $δ_c$ (Fig. 7) and critical coverage $ϕ_c$ (Fig. 8) versus aspect ratio $ε$ for completely disordered, $S = 0$, (a) and completely aligned, $S = 1$, (b) packings. For completely disordered systems ($S = 0$) the maximums on $δ_c(ε)$ (Fig. 7a) and $ϕ_c(ε)$ (Fig. 8a) curves, at some values of $ε_{max}$, were observed. Positions of these maximums were affected values of $ϕ$ (Fig. 7h) and $δ$ (Fig. 8h).

The observed maximums in percolation characteristics $δ_c$ and $ϕ_c$ can reflect internal structure of RSA packings of elongated particles. Particularly, the the maximums in jamming coverage $ϕ_j$ versus $ε$ dependencies were also observed for disordered packings, and they were explained by the competitions of the effects of orientation degrees of freedoms and excluded volume effects. The jamming limit decreased with $ε$ [20], and for elongated particles in the vicinity of percolation packings the terminations of curves $δ_c(ε)$ (Fig. 7h) and $ϕ_c(ε)$ (Fig. 8h) at some critical values of $ε$ were observed.

These maximums became less pronounced for partially aligned systems, and they completely disappeared for completely aligned, $S = 1$, packings (Fig. 7b and Fig. 8b). For the case of $S = 1$, the values of $δ_c$ (Fig. 7b) and $ϕ_c$ (Fig. 8b) grown with $ε$, and for relatively small shell thickness, $δ$, the termination of $ϕ_c(ε)$ was observed.

![FIG. 6. Scaling dependencies of the critical shell thickness $δ_c$ at different values of particles coverage, $ϕ_c$, (a) and of the critical particle coverage $ϕ_c$ at different fixed values of shell thickness, $δ$ (b). The data are presented for aspect ratio of $ε = 4$ for completely disordered ($S = 0$, dashed lines) and completely aligned ($S = 1$, solid lines) packings. For $S = 0$ the data along the $x$ and $y$ directions almost coincide. Here, $L(= 16l, 32l, 64l, 128l)$ is the size of the system. $ν = 4/3$ is the 2D correlation length percolation exponent [58].](image-url)
when the values \( \varphi_c \) exceed the jamming coverage, \( \varphi_j \).

C. Electrical conductivity

For each independent run the electrical conductivity \( \sigma \) displayed the jump at some percolation concentration \( \varphi_\sigma \). Figure 9 presents \( \sigma \), versus the difference, \( d \varphi = |\varphi - \varphi_\sigma| \), for RSA packings of disks (\( \varepsilon = 1 \)) at different shell thickness, \( \delta = 0.2 \) and \( \delta = 0.8 \).

In order to check for the possible non-universality of the percolation exponents, the critical conductivity indexes \( s \) and \( t \) were estimated from the scaling relations for the electrical conductivities just below, \( \sigma \propto (d \varphi)^{-s} \), and above, \( \sigma \propto (d \varphi)^{t} \), the percolation threshold [58]. The classical values for 2D percolation are \( s = t = 4/3 \).

Obtained data evidenced the satisfactory correspondence of the percolation exponents to the classical universality. Below percolation threshold the difference between curves for \( \delta = 0.2 \) and \( \delta = 0.8 \) evidently reflected the effects of shell thickness on value of \( \varphi_\sigma \). Above the percolation threshold, such effects were insignificant. Figure 10 compares \( \sigma \), versus the difference, \( d \varphi = |\varphi - \varphi_\sigma| \), dependencies, for RSA packings of discorectangles (\( \varepsilon = 4 \)) at fixed value of \( \delta = 0.2 \) for completely disordered, \( S = 0 \), (a) and completely aligned, \( S = 1 \), (b) packings. For aligned packings, the significant anisotropy in electrical conductivity was observed and the values along alignment direction, \( \sigma_x \), significantly exceeded the values in perpendicular direction, \( \sigma_y \). Importantly, the obtained data for the mesh sizes of \( m = 1024 \) and \( m = 2048 \) were approximately the same within data errors.

Finally, figure 11 compares electrical conductivity \( \sigma \), versus the difference, \( d \varphi = |\varphi - \varphi_\sigma| \) rather long discorectangles (\( \varepsilon = 10 \)). The data are presented at fixed value of \( \delta = 0.3 \) for completely aligned \( S = 1 \) RSA packings at two values of \( m \). Observed behavior for \( \varepsilon = 10 \) was similar to that observed for \( \varepsilon = 4 \) (Fig. 10b). Above percolation threshold \( (\varphi > \varphi_\sigma) \) the effect of \( m \) was insignificant. However, below percolation threshold \( (\varphi < \varphi_\sigma) \)

FIG. 7. Critical shell thickness \( \delta_c \) versus aspect ratio \( \varepsilon \) at different coverages, \( \varphi \) for completely disordered, \( S = 0 \), (a) and completely aligned, \( S = 1 \), (b) packings.

FIG. 8. Critical coverage \( \varphi_c \) versus aspect ratio \( \varepsilon \) at different shell thickness, \( \delta \), for completely disordered, \( S = 0 \), (a) and completely aligned, \( S = 1 \), (b) packings.
the electrical conductivities estimated at $m = 1024$ were systematically smaller as compared to that estimated at $m = 2048$.

### IV. CONCLUSION

Numerical study of two-dimensional RSA deposition of aligned discorectangles on a plane was done. The partial ordering was characterized by order parameter $S$, with $S = 0$ for random orientation of particles and $S = 1$ for completely aligned particles along the horizontal direction $x$. Analysis of connectivity was performed assuming a core-shell structure of particles. The values of the aspect ratio, $\varepsilon$, and order parameter, $S$, significantly affected the structure of packings, formation of long-range connectivity and electrical conductivity behavior. The observed effects can reflected the competition between the particles’ orientational degrees of freedom and the excluded volume effects \[22\]. For aligned systems, different anisotropies in intrinsic conductivity, long ranged connectivity, and behavior of electrical conductivity were observed. For example, the significant anisotropy in electrical conductivity was observed and the values along alignment direction, $\sigma_x$, were larger than values in perpendicular direction, $\sigma_y$. For aligned finite-size systems, the percolation thresholds along the $x$ and $y$ directions were different. However, these differences disappeared in the limit of infinitely large systems.

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**FIG. 9.** Electrical conductivity, $\sigma$, versus the difference, $d\varphi = |\varphi - \varphi_\sigma|$, for RSA packings of disks ($\varepsilon = 1$) at different shell thickness $\delta$. Here, the value of $\varphi_\sigma$ was identified from the concentration of percolation jump for each independent run, and calculation were done using the mesh size of $m = 1024$. Dashed lines corresponds to the classical exponents $s = t \approx 4/3$ \[58\].

**FIG. 10.** Electrical conductivity, $\sigma$, versus the difference, $|\varphi - \varphi_\sigma|$, for RSA packings of discorectangles at different values of aspect ratio, $\varepsilon$, for fixed shell thickness of $\delta = 0.2$ for completely disordered, $S = 0$, (a) and completely aligned, $S = 1$, (b) packings. Here, the value of $\varphi_\sigma$ was identified from the concentration of percolation jump for each independent run, and calculation were done using the mesh size of $m = 1024$. Dashed lines corresponds to the classical exponents $s = t \approx 4/3$ \[63\].
FIG. 11. Electrical conductivity, $\sigma$, versus the difference, $|\varphi - \varphi_{\sigma}|$. The data are presented for discorectangles with aspect ratio $\varepsilon = 10$, shell thickness $\delta = 0.3$ for completely aligned, $S = 1$, RSA packings. Here, the value of $\varphi_{\sigma}$ was identified from the concentration of percolation jump for each independent run, and calculation were done using the mesh size of $m = 1024$ and of $m = 2048$. Dashed lines corresponds to the classical exponents $s = t \approx 4/3$.

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