

PERCOLATION OF RANDOM CYLINDER AGGREGATES

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ABSTRACT

The percolation threshold ρ_c of Boolean models of cylinders with their axis parallel to a given direction is studied by means of simulations. An efficient method of construction of percolating connected components was developed, and is applied to one or two scales Boolean model, in order to simulate the presence of aggregates. The invariance of the percolation threshold with respect to affine transformations in the common direction of the axis of cylinders is approximately satisfied on simulations. The prediction of the model (ρ_c close to 0.16) is consistent with experimental measurements on plasma spray coatings, which motivated this study.

Keywords: Boolean model, cylinders, multi-scale, percolation, plasma spray coating, simulation.

INTRODUCTION

In many applications in material sciences, thin layers are deposited by means of a plasma spray (Beauvais *et al.*, 2004; Kang, 2005; Sharma *et al.*, 2006). This coating is used to give specific properties to a part, like hardness, thermal or electrical conductivity. When the layer is made of a mixture of various components with a high contrast of properties (like a binary mixture with an insulating and a conductive phase), its physical properties are connected to the possibility for one of the components to percolate through or along the layer. Therefore it is important to know the percolation threshold of the layer in various directions, as a function of the microstructure. In the present work, the toroidal geometry of droplets generating the layer (Beauvais *et al.*, 2004) is simplified by considering cylinders with their axis orthogonal to the layer. A fast and efficient numerical method of simulation to generate multiscale 3D complex aggregates is presented, following the approach developed for sphere aggregates (Jeulin and Moreaud, 2005) and for sphero-cylinders (Jeulin and Moreaud, 2006a,b). This method enables us to estimate the percolation threshold of the random microstructure. It is applied to multi-scale random models of cylinders, for which the percolation thresholds of the cylinders and of their complementary sets are estimated.

METHODS

NUMERICAL SIMULATION OF MULTI-SCALE RANDOM AGGREGATES

Plasma spray coatings are generated by droplets falling on a substrate. A good representation of the layer is given by a realization of a Boolean model involving random primary grains located on points of a Poisson point process (Matheron, 1967). The initial spherical droplets are spread out on the substrate, and their shape can be approximated by flat cylinders with their axis orthogonal to the layer. In what follows we consider the case of a single primary grain (cylinder with diameter D , height H , and shape factor D/H). The germs are located on the points of a Poisson point process with a constant intensity, or restricted to a random set (multi-scale model); in the second case, we obtain a Boolean model based on a Cox point process (Jeulin, 1996).

Our method of simulation can be applied to any kind of aggregates made of objects with a bounded diameter (that can be embedded inside a cube with side T , as shown on Fig. 1), with a random implantation on a multi-scale Poisson process (Jeulin, 2004; Moreaud, 2006).

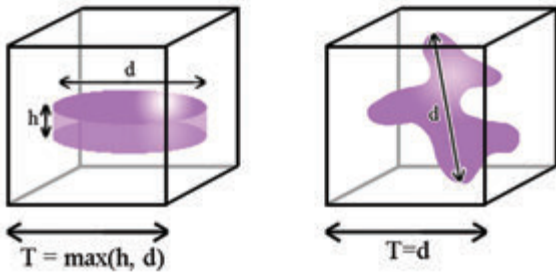


Fig. 1. *Left, cylinder ($T = \max(\text{diameter}, \text{height})$); right, any shape ($T = \text{maximum caliper diameter of the shape}$).*

SIMULATION OF A ONE-SCALE MODEL

Let us consider an implantation of centres of objects with a maximal diameter T , according to a Poisson point process with intensity θ . Every volume V contains a random number of objects N following a Poisson distribution with mean θV . Two properties of the Poisson point process are used: the number of points N_1 and N_2 contained in disjoint volumes V_1 and V_2 are independent variables. Conditionally to N_i , the points falling in V_i are uniformly distributed in V_i .

The aggregates are obtained by overlapping objects. For the simulation, the following information concerning each object is stored: coordinates of the centre, shape parameters (like height and radius for a cylinder), and an aggregate label. To get a fast method of construction, it is useful to limit the

number of tests of intersection between objects. This is made by a subdivision of the complete volume into cubes of size T , corresponding to the maximal diameter of the objects (for parallel cylinders, cubes could be replaced by englobing parallelepipeds for a faster simulation). The cubes are explored in a given order (Fig. 2). In each cube, the random number of centres of objects to locate is simulated by a Poisson point process with intensity θ . For each new centre of object to locate is generated:

- a uniform location of the centre of the object in the current cube.
- a determination of the 13 adjacent cubes of the current cube (according to the exploration order) (Fig. 2).
- For all objects already contained in these volumes is performed an intersection test, detailed later.

The next step (creation and fusion of aggregates) is explained in the “fusion of aggregates” section.

SIMULATION OF A TWO SCALES MODEL

We now consider a two-scale model built according to a Cox point process (Jeulin, 1996), where the centres of objects O_1 with maximal diameter T_1 are randomly located outside exclusion objects O_2 with maximal diameter T_2 . Objects of the small scale O_1 are stored only if their centres are outside large size objects O_2 (Fig. 3, left).

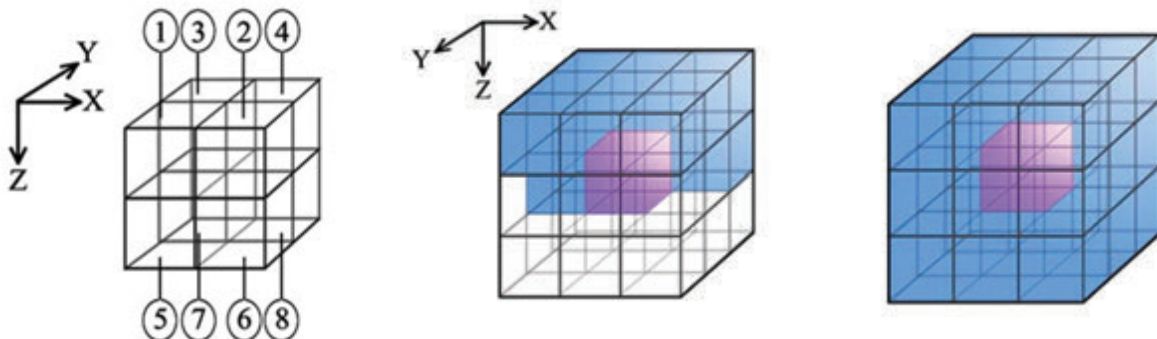


Fig. 2. *Left, division of a volume into eight parts ordered along x , y and z . Middle: the 13 adjacent volumes (light grey) to the current volume (dark grey). Right: the 26 adjacent volumes (bright grey) to the volume V_2 (dark grey).*

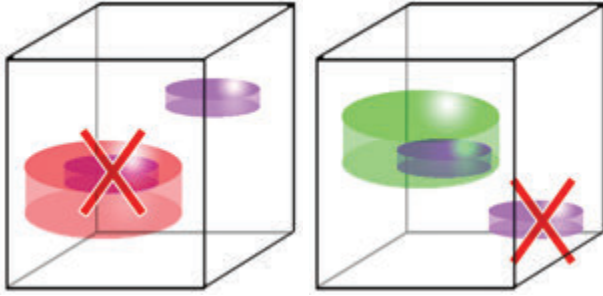


Fig. 3. Examples of two scales simulations. Left: the cylinders corresponding to the small scale objects are kept if their centers are outside of exclusion cylinders corresponding to large size objects. Right: the cylinders, corresponding to the small scale objects, are kept if their centers are inside inclusion cylinders corresponding to large size objects.

The simulation is built in two steps:

- In a first step, a one-scale simulation is performed for the large exclusion objects O_2 , without intersection test. To generate this simulation, the full volume is subdivided into cubes V_2 with side T_2 .
- In a second step, a one-scale simulation is generated for the small size objects O_1 , with addition of a new condition. For every new object O_{1i} to be implanted, the cube V_2 containing its center and its 26 adjacent cubes are extracted (Fig. 2). Then is performed a test of exclusion of the centre of O_{1i} into each large size object O_2 included in these volumes.

FUSION OF AGGREGATES

The fusion of aggregates is a critical point concerning the computation time processing. To get a fast fusion process, the aggregate labels of objects are updated at the end of the simulation by means of a labeling algorithm (Moreaud, 2006). This method avoids multiple browses of the objects during the simulation, which can take a very long time when several millions of objects are implanted.

An intermediate table “AggregateList” is used to link aggregate labels of objects during the simulation with aggregate labels of objects at the end of the simulation. Each object O_i owns an aggregate label $O_{i.ag}$. After intersection tests between a new object O_n to locate and previously located objects O_i , we have to consider several cases:

- If O_n does not intersect objects, a new aggregate is created:
 $O_{n.ag} = \text{NewLabel}$ and $\text{AggregateList}[O_{n.ag}] = \text{NewLabel}$.

- If O_n intersects an object O_i , and if $\text{AggregateList}[O_{i.ag}] \neq \text{AggregateList}[O_{n.ag}]$, then the two aggregates have to be merged.
 We update $O_{i.ag} = O_{n.ag} = \text{AggregateList}[O_{i.ag}] = \text{AggregateList}[O_{n.ag}] = \min(\text{AggregateList}[O_{i.ag}], \text{AggregateList}[O_{n.ag}])$.

When the full volume is simulated, we reflect the updates of aggregate labels only in the AggregateList , and then we update aggregate labels of the objects.

METHOD OF ESTIMATION OF THE PERCOLATION THRESHOLD

A simulation percolates when an aggregate of objects connects two opposite faces of the simulation. To test if a simulation percolates, the highest label N of objects belonging to the cubes of the first plane z is determined, and is compared to the label numbers of objects belonging to the cubes of the last plane z . If all the labels are larger than N , the simulation cannot percolate. If we only look for the percolation threshold, this test, performed for each plane along z , enables us to quickly stop the simulation when it does not percolate. With our method of construction, several realizations of 3D simulations are processed and the presence of at least an aggregate connecting two opposite faces of a field is tested. The percolation threshold p_c is obtained when 50% of the realizations percolate for a given volume fraction of objects, and is given by this volume fraction.

For two-scale simulations, it is interesting to estimate the percolation threshold of the smaller scale objects, keeping constant the volume fraction of the other objects (exclusion in the present case). A research by dichotomy is used to estimate p_c .

TIME PROCESSING

The time processing for a one scale simulation is depending of the number of tests of intersections to process. With a rough algorithm, the number S_1 of tests of intersections is given by:

$$S_1 = \sum_{i=1}^{n-1} i = \frac{n(n-1)}{2} \approx \frac{1}{2}n^2 \quad \text{with } n \text{ the number of}$$

objects. By using our method which divides the volume of simulation, the number S_2 of tests of intersections can be approximated by:

$$S_2 \approx n 14 n \frac{T^3}{v^3} \approx 14 \frac{T^3}{v^3} n^2 \quad \text{with } n \text{ the number of}$$

objects, T the diameter of the objects and V the volume of the simulation. For instance, if we process a simulation on a volume $V = v^3 = 2000^3$ with

5000000 objects with diameter $T = 10$, we obtain $S_1 = 12.5 \times 10^{12}$ tests of intersections of objects and $S_2 = 43.75 \times 10^6$.

SIMULATION OF RANDOM AGGREGATES OF CYLINDERS AND PERCOLATION THRESHOLD

For the study of coatings, we use our method with cylinders with a fixed orientation. By means of large size 3D simulations (typically 5000000 objects by simulation, and 20 realisations by shape factor), the clusters connected to the boundaries of the field are easily extracted thanks to the labellisation of the aggregates, in order to accurately estimate the percolation threshold (Moreaud, 2006).

SIMULATIONS WITH CYLINDERS

A Boolean model of cylinders (BMC) is used for a one scale structure (cf. Fig. 4a). We also use a two scales model (cf. Fig. 4b):

- an exclusion scale modelled by a Boolean model of large cylinders, corresponding to a domain containing no small size cylinders.
- a scale of aggregates made of small size cylinders. The two scales model can be used to simulate random aggregates, or to estimate the percolation threshold of the complementary set of the BMC.

To implement our method, we have to detail the tests of intersection and of exclusion between two cylinders C_1 and C_2 (with radii R_1 and R_2 , centers $M_1(x_1, y_1, z_1)$ and $M_2(x_2, y_2, z_2)$, heights h_1 and h_2):

- Test of intersection: the two cylinders overlap if the two following inequalities are satisfied: $(x_1 - x_2)^2 + (y_1 - y_2)^2 < (R_1 + R_2)^2$ and $4(z_1 - z_2)^2 < (h_1 + h_2)^2$
- Test of exclusion: the center of the cylinder C_1 is outside of the cylinder C_2 if the following condition is fulfilled: $(x_1 - x_2)^2 + (y_1 - y_2)^2 > R_2^2$ or $4(z_1 - z_2)^2 > h_2^2$

ESTIMATION OF THE PERCOLATION THRESHOLD OF THE BOOLEAN MODEL OF PARALLEL CYLINDERS

The percolation threshold of the Boolean model with various grains (spheres, spherocylinders...) can be estimated by different methods (Rintoul and Torquato, 1987; Mecke and Stoyan, 2005; Jeulin and Moreaud, 2005; 2006a,b). We used our method of simulation for a Boolean model of parallel cylinders

with the same size (a single scale) for different shape factors (ratio diameter/height), and using 20 realizations for each case. The structure being anisotropic, we considered the percolation threshold for propagation along the axis of cylinders (axis 1) and orthogonally to their axis (axis 2) (cf. Fig. 5). The simulations are periodic on the faces of the domain parallel to the direction of propagation. The results are given in table 1. The percolation threshold is slightly larger along axis 1 (close to 0.17), as compared to a propagation along axis 2 (close to 0.15). It is practically invariant with the shape factor, as could be expected: as we already indicated in (Charollais *et al.*, 1997), the connectivity properties of the Boolean model are invariant by affine transformations, which changes the shapes and the sizes, but does not introduce any new connection (this property was used to estimate the 3D connectivity number of a Boolean model of ellipsoids). In the case of parallel cylinders, changing the shape factor is equivalent to performing an affinity of the microstructure in the direction parallel to the axis of cylinders. During this transformation, we keep a Boolean model (the Poisson point process of germs being still a Poisson process) and its connectivity, and therefore its percolation threshold.

It is possible that for very large fields of simulations the percolation thresholds in the two directions would converge to the same values, as observed for spherocylinders parallel to the same plane in (Jeulin and Moreaud, 2006a), but this point should be checked with very large simulations. The percolation threshold of parallel cylinders is smaller than for spheres (close to 0.29) and higher than for an isotropic distribution of spherocylinders (between 0.01 and 0.0003 for aspect ratios h/r in the range 10-3000). It is very close to the percolation threshold of spherocylinders with their axis parallel to the same plane (up to ± 5 degrees of disorientation): in (Jeulin and Moreaud, 2006a), for $2r/h = 0.2$ the percolation thresholds along axis 1 and 2 were 0.1294 and 0.16125 respectively. More interestingly, recent experimental data on the in-plane conductivity of air plasma sprayed molybdenum coatings allowed an empirical estimation of the percolation threshold close to 0.16 (Sharma *et al.*, 2006), in an excellent agreement with our predictions with simulations.

Other estimates of the percolation threshold are available: the excluded volume (Balberg *et al.*, 1984), and the zeros of the connectivity number (Bretheau and Jeulin, 1989; Mecke and Stoyan, 2005), being only valid for isotropic Boolean models.

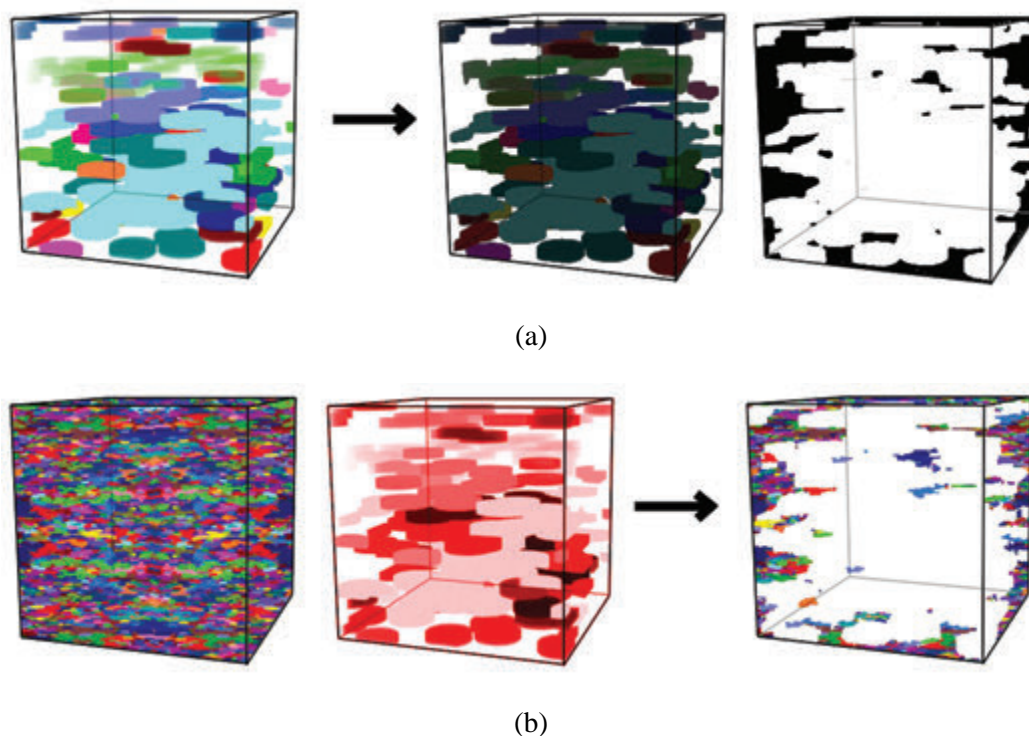


Fig. 4. (a) BMC and the complementary set of the BMC (one scale); (b) two scale Boolean model; an exclusion scale is used to simulate the complementary set of the BMC.

Tab. 1. Percolation threshold for the Boolean model of parallel cylinders for different shape factors. Axis 1 is parallel to the axis of cylinders. Axis 2 is orthogonal to axis 1.

Shape Factor ($2r/h$)	vol. size	ρ_c axis 1	ρ_c axis 2
100	4000	0.1763	0.1540
50	2000	0.1739	0.1522
10	400	0.1672	0.1497
2	200	0.1544	0.1537
1	200	0.1524	0.1543
1	100	0.1518	0.1562
1	40	0.1508	0.1531
0.1	40	0.1250	0.1596

ESTIMATION OF THE PERCOLATION THRESHOLD OF THE COMPLEMENTARY SET OF A BOOLEAN MODEL OF PARALLEL CYLINDERS

Our method of simulation of multi-scale random aggregates can be used to estimate the percolation

threshold of the complementary set of a Boolean model of parallel cylinders (cf. Fig. 4). In that case, we need to use two scales simulations (cylinders for aggregates and for exclusion zones) and to use the method of estimation of the threshold explained in a previous section with a slight change: we estimate by dichotomy the limit volume fraction of exclusion cylinders allowing a percolation of small size aggregate cylinders for 50% of realizations (the small size cylinders are used here to find paths in the complementary set of large cylinders). We get by this way an estimation of the percolation threshold of the complementary set of the Boolean model of parallel cylinders. The results are given in Table 2. The cylinders for aggregates have a diameter and a height equal to 0.1, the cylinders for exclusion zones have a diameter equals to 1 or 2 and a height equal to 1. We can notice that the percolation threshold along axis 1 is higher than for the grains of the Boolean model (compare to Tab. 1), while it is much lower for axis 2: it is much easier to find paths out of cylinders along their faces, than along their axis. To our knowledge, such results are not available in the literature.

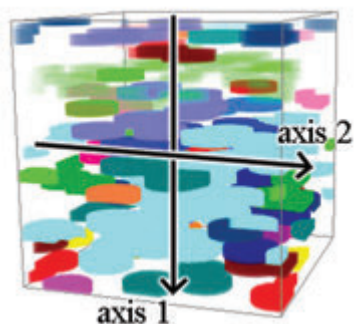


Fig. 5. Illustration of the two axis for the estimation of the percolation thresholds.

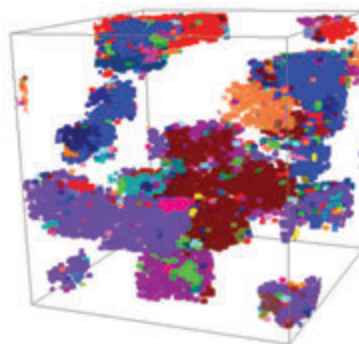


Fig. 6. Example of a two scales simulation (volume: 300^3 , cylinders for aggregates ($r = 2, h=1, V_v=0.2$), cylinders for inclusion zones ($r = 20, h = 10, V_v = 0.1$)).

Tab. 2. Percolation threshold for the complementary set of a Boolean model of parallel cylinders for different shape factors. Axis 1 is parallel to the axis of cylinders. Axis 2 is orthogonal to axis 1.

Shape Factor ($2r/h$)	vol. size	ρ_c axis 1	ρ_c axis 2
1	10	0.2167	0.0406
2	20	0.2320	0.0177

ESTIMATION OF THE PERCOLATION THRESHOLD OF A TWO SCALE BOOLEAN MODEL OF PARALLEL CYLINDERS

Two scales simulations were performed as described earlier, using large cylinders for inclusion zones, and small size cylinders for the generation of aggregates (cf. Fig. 3, right). The results of simulations are given in Tab.3. As expected, a much lower threshold is obtained, as compared to the one-scale model. The two directions give close results. They should be compared to the lower bound of the percolation threshold, asymptotically expected for a very large ratio between the two scales $\rho_c^{(2)} = (\rho_c^{(1)})^2$. In the present case, we have $\rho_c^{(2)} = 0.0256$. It turns out that the lower bound is not reached since the ratio of scales is limited to 10 in our simulations.

Tab. 3. Percolation threshold for two scales simulations. Axis 1 is parallel to the axis of cylinders. Axis 2 is orthogonal to axis 1.

cyl. agr ($2r - h$)	cyl. inc ($2r - h$)	vol. size	ρ_c axis 1	ρ_c axis 2
1 1	10 10	400	0.0367	0.0382
2 1	20 10	400	0.0377	0.0362

CONCLUSION

We presented an original method of simulation of multi scale random aggregates of parallel cylinders based on the Boolean model. This kind of model is well suited to the simulation of coatings deposited by means of a plasma spray. By subdividing the studied volume and using the properties of the Poisson point process, the computation time is significantly reduced to obtain aggregates connected to a boundary of the simulated domain, from which is numerically estimated the percolation threshold. We could check from simulations that the percolation threshold is practically invariant by affinity along an axis with the same orientation as the axis of cylinders, as expected from the invariance of the connectivity by affinity. Furthermore, the predicted percolation threshold is in agreement with experimental data obtained on plasma spray coatings. Using two-scale depositions would significantly lower the percolation threshold of the layer.

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