# Multi Scale Random Sets: From Morphology to Effective Behaviour

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**Abstract** Complex microstructures in materials and in biology often involve multi-scale heterogeneous textures, that we model by random sets derived from Mathematical Morphology. Our approach starts from 2D or 3D images; a complete morphological characterization is performed, and used for the identification of a model of random structure. Simulations of realistic microstructures are introduced in a numerical solver to compute appropriate fields (electric, elastic, velocity,...) and to estimate the effective properties by numerical homogenization, accounting for scale dependent statistical fluctuations of the fields.

## 1 Introduction

As a result of the non-homogeneous dispersion of a charge in a matrix, like carbon black in [33], many nanocomposite materials show an arrangement of aggregates at different scales. To predict the effective properties of such composites (like the dielectric permittivity or the elastic moduli), it is necessary to know the properties of the two components (charge and matrix), and their spatial distribution. For this purpose, we developed a general methodology, based on the theory of random sets [11, 22, 24, 34]: the morphology is summarized and can be simulated by multi-scale random models accounting for the heterogeneous distribution of aggregates. The identification of the model is made from image analysis. From 3D simulations of the model, overall properties like the percolation threshold or the effective properties can be estimated by numerical homogenization, or sometimes by estimations. After a reminder of the basic tools to characterize random sets and of the Boolean model, we illustrate our approach by an introduction to the multi scale combination of

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random sets, to the Cox Boolean model, and to some iterations of random media. In the last part, a prediction of effective properties is obtained by numerical simulations on representative volume elements of the microstructure.

## 2 Principle of Random Structures Modeling, and the Boolean Random Set

The heterogeneity of materials can be handled through a probabilistic approach, which enables us to generate models and simulation of the microstructures.

### 2.1 Random Set Properties

When considering two-phase materials (for instance a set of particles $A$ embedded in a matrix $A^c$), we use a model of random set $A$ [11, 22, 24, 34], fully characterized from a probabilistic point of view by its Choquet capacity $T(K)$ defined on the compact sets $K$, from (1) below, where $P$ denotes a probability:

$$T(K) = P(K \cap A \neq \emptyset) = 1 - Q(K) = 1 - P(K \subset A^c). \tag{1}$$

In practice, $T(K)$ can be estimated by area fraction measurements on 2D images (from true microstructures, or from simulations), after morphological dilation of the set $A$ by the set $K$ [11, 22, 24, 34], noted $A \oplus K$, or calculated for a given theoretical model. Equation (1) is used for the identification of a model (estimation of its parameters, and test of its validity). Particular cases of morphological properties deduced from (1) are the volume fraction $V_v$, the covariance (a useful tool to detect the presence of scales or anisotropies), the distribution of distances of a point in $A^c$ to the boundary of $A$. The access to 3D images of microstructures by means of X-ray microtomography [6] makes it possible to use 3D compact sets $K$ (like balls $B(r)$ with various radii $r$) to characterize the random set.

### 2.2 Boolean Model

Some materials (like porous media or composite materials) can be simulated by means of a basic random set model, namely by distributions of overlapping spheres. A random set model, the Boolean model, was proposed by G. Matheron [22, 24] to reproduce this situation. First, we consider a location of centres of spheres by means of a Poisson point process with intensity $\theta$. In this condition, a volume $V$ contains a random number of centres $N$ following a Poisson distribution with

average value $\theta V$. Then a random set is obtained by the union of random grains (any compact set). For this model, the Choquet capacity is given by (2), where $\overline{V}(A' \oplus \check{K})$ is the average volume of the random primary grain $A'$, dilated by $K$:

$$T(K) = 1 - \exp{-\theta\overline{V}(A' \oplus \check{K})} \tag{2}$$

As a particular case, the volume fraction $V_V$ of the grains, is given by (3):

$$q = 1 - V_V = e^{-\theta\overline{V}(A')}. \tag{3}$$

Using for $K$ a pair of points, three points, or a ball with radius $r$ gives access to the covariance, the third order moment, and to the spherical contact distribution (namely the distribution of the distance of a random point in $A^c$ to the boundary of $A$). For a given population of random grains, these morphological functions are theroretically available from (2). In the special case of spherical grains, the distribution function of the speres radii can be estimated from the covariance [3], and therefore the model can be identified from the two points statistics, which is a one-dimensional information. For more general situations, the population of grains requires higher order moments for the identification of the model.

## 2.3  Percolation of the Boolean Model

For materials made of components with a high contrast of properties, like for instance carbon black in a polymeric matrix, there is a strong effect on the macroscopic properties when a given phase percolates through the structure, inducing connected paths in the samples of the medium. Analytical estimations of the percolation of Boolean models of cylinders are available with the excluded volume model [1]. More recent analytical estimates of the percolation threshold of isotropic Boolean models with convex grains are based on the zeroes of the connectivity number [3, 15, 27]. They give an estimate of the percolation threshold of the grains $p_c^1$ and of $A^c$ . These two percolation thresholds are different, as a result of the fact that the two sets $A$ and $A^c$ are non-symmetrical. The percolation threshold can also be estimated on simulations of the microstructure. It appears that a Boolean model with anisotropic primary grains (for instance sphero-cylinders) shows a much lower percolation threshold (0.01145 for an aspect ratio $l/r = 100$ [1]) than for isotropic grains (0.2895 for spheres [16, 32]. This can explain the expected outstanding mechanical, electrical or chemical properties of composites containing carbon nanotubes, mainly due to their shape, giving a low percolation threshold. The percolation of the complementary set of a Boolean model of spheres obtained analytically from the connectivity number and by simulations are given by 0.05698  and $0.0540 \pm 0.005$ respectively [18].

# 3 Multiscale Combination of Independent Random Sets

Starting from the basic models, more complex structures, such as superposition of scales, or fluctuations of the local volume fraction $p$ of one phase can be generated in a simple way by a combination of various random sets, considering independent realizations. A convenient construction of multiscale models makes use of the union or intersection of independent random sets $A_i$ with different scales. In the case of intersections, $A = \cap_i A_i$, and it is easy to show that we have:

$$P(K) = P\{K \subset (\cap_i A_i)\} = \Pi_i P\{K \subset A_i\}. \tag{4}$$

The result (4) is exact without any approximation, whatever the independent random sets $A_i$ and their scales. For instance the overall volume fraction is the product of the volume fractions of the $A_i$. Similarly the binary covariance $C(h) = P\{x \in A, x + h \in A\}$, and more generally the $n$ points probabilities are obtained as a product of the corresponding individual $n$ points probabilities. A lower bound of the corresponding percolation threshold $p_c$ can be estimated by the products of percolation thresholds $p_c^i$ when the scales are widely separated: $p_c \simeq \Pi_i p_c^i$. This model was used to simulate the random distribution of carbon black in composites by means of the intersection of three Boolean models of spheres at different scales, reproducing the carbon black particles, aggregates, and zones of exclusion in the matrix [1]. The identification of the model is made from the measurement of the overall binary covariance on $C(h)$ images obtained by transmission elecron microscope on thin sections of the material.

# 4 The Multi Scale Cox Boolean Model

## 4.1 The Cox Point Process and the Boolean Model

In another way to account for a non homogeneous distribution of random grains, it is possible to replace the Poisson point process by a Cox point process [10]. Consider a positive random function (RF) giving a non homogenous intensity $\theta(x)$. For any realization of this RF, a Poisson point process with intensity $\theta(x)$ is generated (for any realization, the number of points in a domain $D$ follows a Poisson distribution with average $\theta(D) = \int_D \theta(dx)$. If $\varphi_K(\lambda)$ is the Laplace transform of the positive random variable $E_{A'}\{\theta(A' \oplus \check{K})\}$, where $E_{A'}$ states for the mathematical expectation with respect to the random set $A'$, we have:

$$T(K) = 1 - E_\theta\{e^{-E_{A'}\{\theta(A' \oplus \check{K})\}}\} = 1 - \varphi_K(1).$$

An interesting particular case is obtained by means of a constant intensity $\theta$ inside a first random set $A$. For instance $A$ is a Boolean model of spheres with a large

radius $R$. We then keep the points of a Poisson point process contained in $A$, as germs for centers of spheres with a smaller radius $r$ [16]. The random measure becomes $\theta(dx) = \theta 1_A(x)dx$, where $1_A(x)$ is the indicator function of the set $A$ ($1_A(x) = 1$ if $x \in A$ and $1_A(x) = 0$ if $x \in A^c$). We have

$$T(K) = 1 - \Phi_K(\theta), \tag{5}$$

where $\Phi_K(\lambda)$ is the Laplace transform of the random variable $E_{A'}\{V((\check{A}' \oplus K) \cap A)\}$ obtained on the realizations of the random set $A$, after averaging over the realizations of the primary grain $A'$. As a particular case for a deterministic grain $A'$ the Choquet capacity of the Boolean Cox model is deduced from the distribution of the change of support of the set $A$ over the compact set $\check{A}' \oplus K$ (or probability law of $V((\check{A}' \oplus K) \cap A)$). It is usually difficult to access to this law for any random set $A$, but it can be easily estimated from simulations. We can therefore estimate the theroretical covariance (or higher order moments) of the model. In the case of a large separation of scales between the two sets $A$ and $A'$ the covariance and the three points probability are asymptotically equal to the corresponding theoretical values (4) given for the intersection of independent random sets (Jeulin, 2010, "unpublished"). This result is valid for any $n$ points probability, giving an approximation of the corresponding moments for this Cox Boolean model. For thin sections in electron microscopy, we can notice that the available experimental information is a projection of the set $A$ through the thickness $e$, and therefore we can estimate $T(K \oplus e)$, from which the model can be identified, after replacing the primary grain $A'$ by $A' \oplus e$. For carbon black nanocomposites, it is usual to model the distribution of carbon black particles by means of a three scale model [7, 16, 33]: spherical carbon particles (with a possible distribution of radii $f_1(r)$) are located on Poisson points inside inclusion zones (Boolean model of spheres with a distribution of radii $f_2(r)$) and outside of exclusion zones (Boolean model of spheres with a distribution of radii $f_3(r)$). The identification of the parameters of the model is made by means of an iterative optimization process, minimizing the difference between probabilistic properties of simulations and of images of the material: in [7, 9], the multi scale model is identified from measurements of the covariance, the third order moment, and of the area fraction after 2D closings on transmission electron images.

## 4.2 Percolation of the Cox Boolean Model

As expected, the generation of aggregates produces random media with a lower percolation threshold: for a large separation of scales in a two scales model, the lower bound of the overall percolation threshold $p_c$ is given by the product $p_c^1 p_c^2$ of the corresponding thresholds, as in the case of the intersection of independent random sets. Models involving iteration of scales generate microstructure with a very low percolating threshold, and therefore with improved macroscopic properties when the percolating component presents the higher property (e.g. conductivity or

elastic moduli). It shows that a typical homogeneous distribution of grains in space (like for the standard Boolean model) does not produce a medium with optimal properties. The percolation threshold of various multi scale models was estimated in various situations [16–18] . In [18], we get $p_c = 0.0849$ for a scale factor equal to 30 in a two scale Cox Boolean model of spheres, close to $0.2897^2 = 0.0839$. In [17] the percolation threshold of sphero-cylinders (aspect ratio $l/r = 100$) with centres located in a primary Boolean model of spheres (with $V_v = 0.32$) percolates for $p_c = 0.0056$. Again, considering the case of aggregates of carbon nanotubes, high performances are expected from such a microstructure for a very low volume fraction of charge, due to the presence of clusters.

## 5 Change of Scale in Random Media

In materials sciences, one is interested by the prediction of the macroscopic behavior of a physical system from its microscopic behavior. Models of random sets as seen before can give a good description of the morphological arrangement of components, and are a crucial step towards the connexion between morhological textures and the overall physical properties. The field of prediction of the effective properties (overall properties of an equivalent homogeneous medium) of random heterogeneous media from their microstructure is usually denominated as Homogenization. Most relevant tools are based on variational principles, from which bounds of the effective properties can be derived. In some specific situations  based on multiscale random sets bounds can coincide and therefore provide an estimation of the properties, as illustrated below. In more general cases, an estimation of the effective behaviour is obtained from numerical simulations on realizations of random media on finite domains. This numerical approach is relevant, provided the notion of Representative Volume Element (RVE) can be correctly taken into account for random sets, as seen below.

## 6 Multi Scale Iterations of Random Media

Using multiscale iterations of random media gives processes to generate two components microstructures with "optimal properties", i.e. with the highest possible effective properties, given the volume fraction and the physical properties of components. The probably most famous case is given by the Hashin coated spheres assemblage [6]. It can be shown that for this isotropic morphology the effective conductivity (or effective dielectric permittivity) is given by the upper (resp. lower) Hashin–Shtrikman (H-S) bound when the material with the highest (resp. lowest) conductivity is put on the outer layer of spheres. We give here two other types of models, based on iterations of random sets.

## 6.1 Dilated Poisson Hyperplanes

Third order bounds (depending on the three-points statistics of a random medium) of effective properties (like conductivity, elastic moduli) were derived in [2, 26]. from a classical variational principle. It was shown that for an isotropic two component random set, these bounds depend on two positive functions of the volume fraction p, bounded by 1 [28]. When these functions summarizing the three points probability of a random set are equal to 1 (resp. 0), the two third order bounds coincide with the upper (resp. lower) Hashin–Shtrikman (H-S) bound. We have shown [12, 13] that for an infinite union of dilated Poisson flats with widely separated scales, we get a multi scale structure percolating for any volume fraction, and corresponding to this situation: its effective permittivity is given by the upper (resp. lower) HS bound, when attributing the highest permittivity to the union of dilated flats (resp. to their complementary set). This result is valid for a porous medium, where the upper (H-S) bound becomes the effective permittivity. The same result holds when considering the case of the bulk modulus in linear elasticity.

## 6.2 A Two Scales Hard Core Composite

In some situations, a microstructure is made of non overlapping objects (for instance spheres), described by a so-called hard-core process [34]. It turns out that for this model with spheres of a single radius, numerical simulations show that the obtained effective properties are close to the lower H-S bound for a "soft" matrix, and close to the higher H-S bound for "soft" spheres (like pores) [8, 35]. This can be explained by the fact that for this model the morphological parameter involved in the calculation of bounds is close to 1 in the first case [36], and to 0 in the second one. This point can be used to give an approximate estimate of the effective properties of a material containing non overlapping aggregates (similar to the mentioned Cox Boolean model), obtained in two steps, as illustrated in an elastomeric matrix containing carbon black particles CB [8]. Composite spherical aggregates are made of percolating CB spheres (with volume fraction $p_1$) and of the elastomeric component (with volume fraction $1 - p_1$). The elastic properties of the aggregates are estimated by the third order upper bound of a Boolean model of spheres (or equivalently of a hard core model) with volume fraction $p_1$. In a second step, these spherical aggregates (with a radius much larger than the radius of CB spheres, involving a separation of scales) are implanted according to a second hard core process with volume fraction $p_2$, giving an overall volume fraction $p = p_1 p_2$. The overall moduli are now estimated by the H-S lower bound computed for this composite. As shown in [14], the estimates of the shear modulus $G$, obtained for different values of $p_1$ are consistent with the experimental measurements obtained for various mixes, a more uniform distribution in space giving a lower $G$. For a given $p$, $G$ increases when $p_1$ decreases, making easier the percolation in the

two-scale model. A more accurate estimate is obtained by computer intensive numerical simulations [8].

# 7 Prediction of Effective Properties by Numerical Simulations

An efficient way to solve the problem of homogenization of physical properties, for instance to predict the dielectric permittivity or the effective mechanical properties of heterogeneous media, makes use of numerical solutions of the corresponding partial differential equations (PDE) solutions, before estimating the effective properties by spatial averaging of the solution. This requires the input of 3D images (real images, as obtained by confocal microscopy, or by X-ray microtomography) or simulations, so-called digital materials. In a second step a computational code (Finite Elements, Fast Fourier Transform, PDE numerical solver) is implemented.

## 7.1 Digital Materials

We use a method derived from [5, 29], to estimate the equivalent macroscopic dielectric constant $\varepsilon^*$ from the structure of a material and the properties of its constituents [30]. For this, we compute the electric field $E(x)$ inside the material, knowing the local dielectric permittivity $\varepsilon(x)$, by application of a constant electric field $E_0$, and of periodic boundary conditions. An iterative calculation in the Fourier space is used to estimate $E(x)$, giving $\varepsilon^*$ by means of a spatial average $<>$. The numerical solution is obtained by means of the Green function of a homogeneous reference medium for the corresponding PDE to the electrical field or for the elastic field problem. Using the FFT approach is very versatile and does not require any meshing of the microstructure, in contrast with other numerical methods such as Finite Elements. In elasticity, stress and strain field maps are obtained on the scale of microstructure, to get a detailed study of the effect of the microstructure on the local fields. With the Morph–Hom code developed by F. Willot [37, 38] images of large sizes (up to $1500^3$) can be handled for any contrast of properties, and therefore the electrostatic or elastic behaviour of porous media or of rigid media was studied by numerical techniques. This was applied to the Boolean model of spheres with any volume fractions [37] and to two-scales and three-scales Cox Boolean models of spheres [38]. The reinforcing effect of the iteration of scales with a large separation is clearly observed in the case of a very high conductivity or of a rigid phase, as a result of a lower percolation threshold. In the case of porous media, the reverse effect is observed, as would appear in the case of a damage at the interface between clusters of rigid particles and the matrix.

## 7.2 Statistical Approach of the RVE

When working on images of a material or on realizations of a random medium, a natural question arises [12, 20]: what is the representativity of the effective property estimated on a bounded domain of a microstructure? In other words, what is the size of a so-called "Representative Volume Element" RVE. In many industrial applications, mainly in the case of electronic parts such as MEMS, the size of the microstructure cannot be neglected with respect to the size of parts. The parts cannot be considered as an infinite homogeneous medium, and fluctuations of effective properties are observed and must be accounted for. We address this problem by means of a probabilistic approach giving size-dependent intervals of confidence, initially developed in the framework of the homogenization of the elastic moduli of random media [4, 20], and based on the size effect of the variance of the effective properties of simulations of random media. This approach was applied to the elastic properties of rigid and of porous Boolean models of spheres [37, 38].

### 7.2.1 The Integral Range

We consider fluctuations of average values over different realizations of a random medium inside the domain $B$ with the volume $V$. In Geostatistics [23], it is well known that for an ergodic stationary random function $Z(x)$, with mathematical expectation $E(Z)$, one can compute the variance $D_Z^2(V)$ of its average value $\bar{Z}(V)$ over the volume $V$ as a function of the central covariance function $\overline{C}(h)$ of $Z(x)$ by:

$$D_Z^2(V) = \frac{1}{V^2} \int_B \int_B \overline{C}(x-y) \, dxdy, \tag{6}$$

where

$$\overline{C}(h) = E\{(Z(x) - E(Z))(Z(x+h) - E(Z))\}.$$

For a large specimen (with $V \gg A_3$), equation (6) can be expressed to the first order in $1/V$ as a function of the integral range in the space $R^3$, $A_3$, by

$$D_Z^2(V) = D_Z^2 \frac{A_3}{V}, \tag{7}$$

$$\text{with } A_3 = \frac{1}{D_Z^2} \int_{R^3} \overline{C}(h) \, dh, \tag{8}$$

where $D_Z^2$ is the point variance of $Z(x)$ (here estimated on simulations) and $A_3$ is the integral range of the random function $Z(x)$, defined when the integral in equations (6) and (8) is finite. The asymptotic scaling law (7) is valid for an additive variable $Z$ over the region of interest $B$. To estimate the effective elasticity or permittivity tensors from simulations, we have to compute the average stress

$\langle\sigma\rangle$ and strain $\langle\varepsilon\rangle$ (elastic case) or electric displacement $\langle D\rangle$ and electrical field $\langle E\rangle$. For the applied boundary conditions, the local modulus is obtained from the estimations of a scalar, namely the average in the domain $B$ of the stress, strain, electric displacement, or electric field. Therefore the variance of the local effective property follows the equation (7) when the integral range $A_3$ of the relevant field is known. Since the theoretical covariance of the fields ($\sigma$ or $\varepsilon$) is not available, the integral range can be estimated according to the procedure proposed by G. Matheron for any random function [25]: working with realizations of $Z(x)$ on domains $B$ with an increasing volume $V$ (or in the present case considering subdomains of large simulations, with a wide range of sizes), the parameter $A_3$ is estimated by fitting the obtained variance according to the expression (7).

### 7.2.2   Practical Determination of the Size of the RVE

When considering a material as a realization of a random set or of a random function RF, the idea that there exists one single possible minimal RVE size must be left out. Instead, the size of a RVE can be defined for a physical property $Z$, a contrast, and a given precision in the estimation of the effective properties depending on the number of realizations that one is ready to generate. By means of a standard statistical approach, the absolute error $\epsilon_{abs}$ and the relative error $\epsilon_{rela}$ on the mean value obtained with $n$ independent realizations of volume $V$ are deduced from the 95% interval of confidence by:

$$\epsilon_{abs} = \frac{2D_Z(V)}{\sqrt{n}}; \; \epsilon_{rela} = \frac{\epsilon_{abs}}{Z} = \frac{2D_Z(V)}{Z\sqrt{n}}. \tag{9}$$

The size of the RVE can now be defined as the volume for which for instance $n = 1$ realization (as a result of an ergodicity assumption on the microstructure) is necessary to estimate the mean property $Z$ with a relative error (for instance $\epsilon_{rela} = 1\%$), provided we know the variance $D_Z^2(V)$. Alternatively, we can decide to operate on smaller volumes (provided no bias is introduced by the boundary conditions), and consider $n$ realizations to obtain the same relative error. This methodology was applied to the case of the dielectric permittivity of various random media [19], and to the elastic properties and thermal conductivity of a Voronoï mosaic[20], of materials from food industry [21], or of Boolean models of spheres [37]. Further results were obtained for the Boolean model and for the multiscale Cox Boolean model of rigid spheres (with radius 10 voxels, the larger scale being a Boolean model of spheres with radius 100 voxels). The results in [37] indicate that the highest RVE sizes for the one-scale model correspond to rigidly-reinforced media with $V_v \approx 0.49$. Accordingly, the variances $D_Z^2(V)$ and $D_Z^2$, with $Z = \sigma_m$ (mean stress or 1/3 of the trace of the stress tensor), have been computed for the two-scales Cox Boolean model when $V_{v1} = V_{v2} = 0.7$ for a contrast higher than 10,000 of the bulk and shear moduli for a scale ratio equal to 10 [38]. It is found that the integral range of the two-scales is increased by a factor close to 5.8 times, whereas the point variance

increases by a factor $32^2$. Accordingly, a 10% precision(i.e. $\epsilon = 0.1$) is achieved when $V \approx 550^3$, which corresponds to an increase of the RVE by a factor $5.7^3$. The drastic increase of the point variance in the case of the two-scale random medium underlies very large local stresses, that could induce more damage in the matrix if it contains defect, as compared to the one scale model for the same volume fraction.

## 8 Conclusion

Multi scale models of random media provide a wide variety of morphologies to simulate complex microstructures for application to real materials. These models are able to capture phenomena like the observation of very low percolation thresholds explaining the enhancement of some effective properties. Combined to predictive models by analytical means, or by numerical simulations, they give access to the optimization of microstructures with respect to the required properties.

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