LOCAL ENTROPY CHARACTERIZATION OF CORRELATED RANDOM MICROSTRUCTURES

C. Andraud[1], A. Beghdadi[2], E. Haslund[3], R. Hilfer[3,4*], J. Lafait[1], and B. Virgin[3]

[1]*Laboratoire d'Optiques des Solides, Universite Pierre et Marie Curie, CNRS, 4 Place Jussieu, 75252 Paris Cedex 05, France*

[2]*LPMTM-CNRS, Institut Galilee, Universite Paris Nord, 93430 Villetaneuse, France*

[3]*Institute of Physics, University of Oslo, P.O.Box 1048, 0316 Oslo, Norway*

[4]*Institut für Physik, Universität Mainz, 55099 Mainz, Germany*

Abstract

A rigorous connection is established between the local porosity entropy introduced by Boger et al. (Physica A **187**, 55 (1992)) and the configurational entropy of Andraud et al. (Physica A **207**, 208 (1994)). These entropies were introduced as morphological descriptors derived from local volume fluctuations in arbitrary correlated microstructures occuring in porous media, composites or other heterogeneous systems. It is found that the entropy lengths at which the entropies assume an extremum become identical for high enough resolution of the underlying configurations. Several examples of porous and heterogeneous media are given which demonstrate the usefulness and importance of this morphological local entropy concept.

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*present address: ICA-1, Universität Stuttgart, Pfaffenwaldring 27, 70569 Stuttgart

I. INTRODUCTION

An accurate geometric characterization of the correlated random microstructures found in disordered heterogeneous materials is generally difficult. One possible method consists in using globally well-defined morphological descriptors (e.g. volume fraction or specific internal surface area) as local descriptors. While local volume fraction fluctuations have been discussed previously in the context of porous media and photographic granularity [?,?,?] the idea of local morphological descriptors has only recently been applied systematically in the form of local porosity theory [?,?,?,?,?,?,?,?,?]. Local porosity theory generalizes the successful and widely used effective medium approximation to correlated random microstructures.

Microstructural information of a different kind can be obtained by applying the Gibbs-Shannon entropy concept from information theory to distributions of locally fluctuating morphological descriptors. Such "local geometry entropies" were introduced independently in [?] and [?]. It is the primary purpose of the present paper to clarify the similarities and differences between the two different local geometry concepts. Furthermore we resolve the normalization problem of [?], and provide examples of the usefulness and microstructural sensitivity of the local geometry concept. We begin with a discussion of the "local porosity entropy" introduced in [?] in the next section. Subsequently we present the definition of the "configurational entropy" as introduced in [?]. In section **??** we establish the relationship between the two concepts, and in section **??** we show that, for sufficiently high resolution, they give rise to the same "entropy length". Finally we apply our entropic analysis to several examples including percolation models, Poisson grain models, thin films and porous sandstones.

II. LOCAL POROSITY ENTROPY

Distributions of local morphological descriptors of random microstructures were introduced in [?,?]. The local geometry approach was recently reviewed and extended in [?]. In the present study we consider for simplicity only two component media in two dimensions. Such media are of practical importance as granular metal films for composite coatings. They arise also as surface morphologies when sectioning a three-dimensional medium.

Given a two-component random microstructure in the plane $\mathbb{R}^2$ let the subset $\mathbb{P} \subset \mathbb{R}^2$ denote the set occupied by one of the two components (phases), and $\mathbb{M} = \mathbb{R}^2 \setminus \mathbb{P}$ its complement occupied by the second component. For microstructures obtained by sectioning a porous medium the set $\mathbb{P}$ represents the fluid-filled pores while the set $\mathbb{M}$ is the solid mineral matrix. The local volume fraction occupied by the set $\mathbb{P}$ within an observation region $\mathbb{K}$ is then defined as [?,?,?]

$$\phi(\mathbb{K}) = \frac{V(\mathbb{P} \cap \mathbb{K})}{V(\mathbb{K})} \tag{2.1}$$

where $V(\mathbb{G})$ denotes the volume (=area) of a set $\mathbb{G} \subset \mathbb{R}^2$. The local porosity distribution is defined as the probability density [?,?,?]

$$\mu(\phi; \mathbb{K}) = \langle \delta(\phi - \phi(\mathbb{K})) \rangle \tag{2.2}$$

where $\delta(x)$ is the Dirac δ-distribution, and $\langle ... \rangle$ denotes an average over the underlying probability distribution governing the configurations of the random microstructure. Note that the local porosity distribution depends on the size and shape of the observation region $\mathbb{K}$. Recently [?] it was found that this dependence disappears in a suitable macroscopic scaling limit. In the following it will be assumed that for each $\mathbb{K}$ the function $\mu(\phi; \mathbb{K})$ is a given continuous probability density with support on the unit interval. This assumption can be justified by arguing that possible discrete components are always smeared because of finite image resolution.

Local porosity entropies $I(\mathbb{K})$ are obtained by calculating the Gibbs-Shannon entropy of the family of local porosity distributions $\mu(\phi;\mathbb{K})$ as in [?]

$$I(\mathbb{K}) = \int_0^1 \mu(\phi;\mathbb{K}) \log \mu(\phi;\mathbb{K})\ d\phi \tag{2.3}$$

which assumes, as usual, a uniform distribution as the a priori weight. Examples of local porosity entropies were given in [?] for synthetic images, and in [?] for experimental systems.

III. “CONFIGURATION” ENTROPY

In [?,?] the two-dimensional image is discretized into black and white picture elements (pixels) forming a quadratic lattice with lattice constant a. The black and white image is then analyzed using a quadratic observation region $\mathbb{K}$ (sliding cell) of sidelength L which contains $M = (L/a)^2$ pixels. The observation region is moved to N different positions, and the number $N_k(M)$ is defined as the number of cells of size M containing k active (=black) pixels. Using the relative frequencies

$$p_k(M) = \frac{N_k(M)}{N} \tag{3.1}$$

as estimators for probabilities the “configuration” entropy has been defined as [?]

$$H^*(M) = \frac{H(M)}{\log(M+1)} = -\frac{1}{\log(M+1)} \sum_{k=0}^{M} p_k(M) \log p_k(M). \tag{3.2}$$

where $H(M)$ represents the usual Gibbs-Shannon entropy of the discrete probabilities $p_k(M)$, and $M = (L/a)^2$.

The expression (**??**) does not use the Gibbs-Shannon expression $H(M)$ but divides it with $\log(M+1)$ similar to multiplicative renormalization in the theory of critical phenomena. The normalization was introduced because the underlying probability space changes when the number M of pixels inside the measurement cell is changed. The choice $1/\log(M+1)$ for the normalization factor however renders $H^*(M)$ nonadditive. The next section will show that this can be avoided by using the definition (**??**), and that a precise relation exists between $I(\mathbb{K})$ and $H^*(M)$.

IV. RELATION BETWEEN THE TWO ENTROPIES

The relationship between $I(\mathbb{K})$ and $H^*(M)$ is established by applying the definitions of section **??** to the same discretized quadratic observation region $\mathbb{K}$ of sidelength L which was used in section **??**. For such a choice of $\mathbb{K}$ the unit interval $[0,1]$ of porosities is conveniently subdivided into subintervals bounded by the porosities

$$\phi_k = \frac{k}{M+1} \tag{4.1}$$

where $0 \le k \le M+1$ and $M = (L/a)^2$. If $\mu(\phi;\mathbb{K})$ is given as a continuous function the integral in equation (**??**) can be approximated as $I(\mathbb{K}) = \lim_{M\to\infty} I(M)$ where

$$\begin{aligned} I(M) &= \sum_{k=0}^{M} \mu(\phi_k;\mathbb{K}) \log \mu(\phi_k;\mathbb{K})(\phi_{k+1} - \phi_k) \\ &= \frac{1}{M+1} \sum_{k=0}^{M} \mu(\phi_k;\mathbb{K}) \log \mu(\phi_k;\mathbb{K}). \end{aligned} \tag{4.2}$$

Identifying the probabilities

$$\frac{\mu(\phi_k;\mathbb{K})}{M+1} = p_k(M) \tag{4.3}$$

with the relative frequencies $p_k(M)$ of section **??** gives

$$I(M) = \sum_{k=0}^{M} p_k(M) \log[p_k(M)(M+1)] \tag{4.4}$$

and thus

$$I(M) = \log(M+1)(1 - H^*(M)) \tag{4.5}$$

provides a rigorous relationship between the entropies which becomes exact in the limit $(L/a) \to \infty$.

Note, however, that in the continuum limit $a \to 0$ at fixed L the normalization of $H^*(M)$ gives rise to a peculiar behaviour of this quantity. While in this limit $\lim_{a\to 0} I(M) = I(\mathbb{K})$ becomes a number depending on the local porosity fluctuations as described by $\mu(\phi;\mathbb{K})$, equation (**??**) shows that $\lim_{a\to 0} H^*(M) = 1$ which is independent of the microstructure. In other words the "configuration" entropy $H^*(M)$ should not be used to distinguish different morphologies in the continuum limit. We suggest calling $I(\mathbb{K})$ "local geometry entropy". The generalized name accounts for the fact the $I(\mathbb{K})$ is readily generalized to morphological descriptors other than porosity, such as local specific internal surface areas or local curvatures [**?**].

V. ENTROPY LENGTH

An interesting quantity associated with local geometry distributions is the entropy length. The entropy length L^* is defined as the length at which $I(M)$ becomes extremal, $\partial I(M)/\partial L|_{L=L^*} = 0$ [**?**]. Differentiating (**??**) yields

$$I^{'}(M) = \frac{1}{M+1}(1 - H^*(M)) - \log(M+1) H^{*^{'}}(M) \tag{5.1}$$

where the prime denotes the derivative with respect to M. For large M, e.g. in the limit of high resolution $a \to 0$ at fixed L, the first term becomes negligible, and thus the entropy length can be determined equally from the condition $\partial H^*(M)/\partial L = 0$. This is illustrated in Figures **??** and **??** and **??**. Figure **??** shows the microstructure of a Poisson grain model obtained by placing circles of equal radii around centers which are distributed at random and with constant number density. The corresponding entropy curves $I(M)$ and $H^*(M)$ are shown in Figures **??** where, to facilitate the comparison, the curves have been shifted along the ordinate to have the same maximum value 0. We have assumed $a = 1$ which allows one to plot the curves as function of the size L of the observation square. Note that the curves show two extrema, of which the ones at large M coincide, while the extrema at small M are different. The entropy length is an accurate measure of the typical linear size of the different phases, pores or components.

VI. APPLICATIONS

To illustrate the usefulness of the local geometry concept as a morphological descriptor we apply it to several random microstructures. We use two computer generated images of simple models for disordered systems, and two experimentally obtained disordered microstructures. The two synthetic images are generated from the percolation model on a lattice and from the Poisson grain model consisting of uniformly distributed overlapping spheres. The two experimental morphologies are observed when a thin film of gold is deposited on a glass substrate, and when slicing through a natural sandstone.

A. Simulated Bernoulli site percolation model

Perhaps the simplest model of a random microstructure is the site percolation model [**?**]. In this model, each lattice site has a probability $\langle\phi\rangle$ of being occupied and $(1 - \langle\phi\rangle)$ of being empty. The occupation of sites is assumed to be statistically independent. A configuration of such a system in which the occupied (black) sites represent pore space, and the unoccupied ones matrix space is shown in Figure **??** for $\langle\phi\rangle = 0.3$.

The local porosity distribution $\mu(\phi;\mathbb{K})$ for the percolation model depends only on $\langle\phi\rangle$ and $|\,\mathbb{K}\,| = M = (L/a)^d$, and is given by the binomial distribution [**?**,**?**]. If a hypercubic lattice with lattice constant a is considered, then the LPD for a d-dimensional hypercubic measurement cell $\mathbb{K}$ of side L reads

$$p_k(M) = \frac{\mu(\phi_k;\mathbb{K})}{M+1} = \frac{M!}{[(M+1)\,\phi_k]!\;[(M+1)\;(1-\phi_k-1/(M+1))]!}\,\langle\phi\rangle^{(M+1)\phi_k}\,(1-\langle\phi\rangle)^{(M+1)(1-\phi_k-1/(M+1))} \tag{6.1}$$

where $M = (L/a)^d$, $\phi_k = k/(M+1)$ and $k = 0, 1, \ldots, M$. Its local geometry entropy

$$I(M) = \log(M+1) - H(M) = \log(M+1) + \sum_{k=0}^{M} p_k(M)\log(p_k(M)) \tag{6.2}$$

is displayed in Figures **??** and **??** as the curve with triangular symbols. The extrema of the curve are at the boundaries. This reflects correctly the fact that the microstructure is homogenous and statistically independent at the microscopic resolution (i.e. the lattice constant). The value $I(1) = \log(2) + \langle\phi\rangle\log\langle\phi\rangle + (1-\langle\phi\rangle)\log(1-\langle\phi\rangle) \approx 0.08228$ is approached for $M = 1$. For $M \to \infty$ the entropy $I(L)$ diverges to infinity (Note that Figure **??** shows $-I(L)$ which diverges to $-\infty$), while $H^*(M)$ in Figure **??** approaches a constant.

To demonstrate the influence of finite system size and statistics we have plotted in Figures **??** and **??** also the curves $H^*(L)$ and $-I(L)$ determined directly from the image. These curves are shown using circular symbols. The agreement with the exact result is satisfactory although some deviations are apparent.

B. Simulated Poisson grain model

Another often used model for random microstructures is the Poisson grain model. In this model a constant number density of circles (spheres) is randomly placed with uniform density into continuous space. Figure **??** shows a random throw of disks with diameter of 15 pixels in two dimensions. The porosity of the image is $\langle\phi\rangle = 0.5$.

The entropy functions $-I(L)$ and $H^*(L)$ for this image differ significantly from those for the percolation image. Now the curves show a pronounced extremum while those for the percolation case were monotonic. Both curves exhibit an extremum at $L^* \approx 16$ corresponding roughly to the circle diameter of 15 pixels. From other simulations we find that the extremum changes with the circle (or sphere) diameter. Hence the position of the extremum in $-I(L)$ or $H^*(L)$ is related to a characteristic length scale for the morphology. This interpretation is also consistent for the percolation model where the extremum occurs at $L^* \approx 1$ which corresponds to the pixel diameter.

C. Experimental gold film morphology

The image of Figure **??** represents a digitized and thresholded image of a transmission electron micrograph of a thin film of gold on a glass substrate. The film was deposited by thermal evaporation under ultra-high vacuum. The evaporation was stopped before the gold phase, shown in black, begins to percolate. The surface coverage is $\langle\phi\rangle = 0.413$.

The curves for the local porosity entropy and the configuration entropy of the gold film morphology from Figure **??** are shown using square symbols in Figure **??** and **??**. Both curves exhibit again an extremum, this time at $L^* \approx 11$. This corresponds roughly to the characteristic thickness of black and white regions in the image. The length L^* is expected to decrease down to the size of the elementary metallic grain at percolation [**?**]. This is the main reason why in [?,?] it was suggested that the optical properties of these materials have to be calculated at scale L^*.

D. Experimental oolithic sandstone morphology

In Figure **??** we show a slice through a natural oolithic sandstone. The image was obtained by slicing a sample of Savonnier sandstone whose pore space was made visible by first filling it with a coloured epoxy resin. The cut surface was polished and photographed. The photgraph could then be digitized and thresholded to give a black and white image. The threshold was adjusted to match the measured bulk porosity of $\langle\phi\rangle = 0.186$. In the image shown in Figure **??** the pore space is coloured black while the rock matrix is shown in white.

The entropy functions for the image of Figure **??** are displayed as curves with cross symbols in Figures **??** and **??**. Note that there is again a maximum in $-I(L)$ at $L^* \approx 47$, but this maximum is much broader and less pronounced than for the other images. The curve for $H^*(L)$ in Figure **??** also shows a maximum at $L^* \approx 55$. It is so flat that it is difficult to distinguish from the figure.

The difference between the values of L^* obtained from maximizing $H^*(L)$ as opposed to $-I(L)$ can be understood from Eq. (**??**). The equality of the two entropy lengths is obtained in the limit $M \to \infty$ in which the first term in Eq. (**??**) becomes negligible. For finite M the lengths can in general be different. Hence we conclude that for the morphology of Figure **??**, which represents the most heterogeneous case, the resolution of the image needs to be improved before $I(M)$ and $H^*(M)$ will give the same entropy length.

VII. DISCUSSION AND CONCLUSION

In this paper we have established an exact relationship between the local porosity entropy [?] and the configurational entropy of [?]. These two morphological entropies were introduced independently by the authors to characterize random but correlated microstructures. We have discussed the advantages and disadvantages of the two entropies and suggest to call their common element local geometry entropy. It is argued that local geometry entropies are a useful morphological indicator for random correlated morphologies.

The evaluation of the local geometry entropy for selected morphologies indicates the existence of a minimum. The position L^* of the minimum in the entropy function is called the entropy length. The length L^* correlates well with a characteristic length scale of the microstructure. It is, however, different from the pixel-pixel correlation length. Our analysis and past experience indicates that the entropy length L^*, as measured here, corrresponds to that size of measurement cells at which a finite fraction of measurement cells begin to have vanishing local porosity.

Another observation concerns the width (or curvature) of the extremum. More heterogeneous microstructures such as the one in Figure **??** appear to show a wider extremum than those microstructures, such as Figures **??** or **??**, in which the black and white regions are more compact. This observation is consistent with the findings in [?] for synthetic morphologies. Hence we conclude tentatively that the width of the extremum correlates with the complexity or heterogeneity of the microstructure. More analyses of synthetic and experimental data are desirable to further elucidate the geometrical and morphological information content of the local geometry concept.

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FIGURE CAPTIONS

1. (a) FIGURE **??**

   Random microstructure of the (uncorrelated) site percolation model in which occupied (black) points represent pore space. The porosity (volume fraction of pore space) in the image is $\langle\phi\rangle = 0.3$.

   (b) FIGURE **??**

   Poisson grain model configuration with constant point density of circles. Total volume fraction of $\langle\phi\rangle = 0.5$

   (c) FIGURE **??**

   Discontinuous thin film morphology of gold at volume fraction $\langle\phi\rangle = .413$

   (d) FIGURE **??**

   Planar thin section through a Savonnier oolithic sandstone with pore space rendered black. The side length of the image corresponds to roughly 1 cm, the total porosity is $\langle\phi\rangle = .186$

2. FIGURE **??**

   Comparison of local porosity entropy $-I(L)$ and configuration entropy $H^*(L)$ for the Poisson grain model shown in Figure **??** with disks diameter 15 pixels. Note that the curves show extrema at the same length $L \approx 26$. The curves are shifted to have the same ordinate $-I(L) = H^*(L) = 0$ at the extremum.

3. FIGURE **??**

   Local porosity entropy $-I(L)$ [**?**] as a function of the side length of the measurement cell calculated for all random microstructures shown in Figures **??**, **??**, **??** and **??**. The circles correspond to the site percolation image shown in Figure **??**. The triangles are the theoretical values for an infinitely large image at the same bulk porosity. The diamonds are the result for the Poisson grain model shown in Figure **??**. The squares represent the experimentally observed gold film morphology of Figure **??**, while the crosses correspond to the sandstone cross section shown in Figure **??**.

4. FIGURE **??**

   Configuration entropy $H^*(L)$ [**?**,**?**] as a function of the side length of the measurement cell calculated for all random microstructures shown in Figures **??**, **??**, **??** and **??**. The symbol usage is the same as in Figure **??** and is explained in the figure caption there.