



**HAL**  
open science

# Random Auxetic Porous Materials from Parametric Growth Processes

Jonàs Martínez

► **To cite this version:**

Jonàs Martínez. Random Auxetic Porous Materials from Parametric Growth Processes. 2020. hal-02911788v2

**HAL Id: hal-02911788**

**<https://inria.hal.science/hal-02911788v2>**

Preprint submitted on 10 Nov 2020 (v2), last revised 5 Jul 2021 (v3)

**HAL** is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

# Random Auxetic Porous Materials from Parametric Growth Processes

Jonàs Martínez

*Université de Lorraine, CNRS, Inria, LORIA  
615, rue du Jardin Botanique, 54600 Villers-lès-Nancy, France*

---

## Abstract

We introduce a computational approach to optimize random porous materials through parametric growth processes. We focus on the problem of minimizing the Poisson's ratio of a two-phase porous random material, which results in an auxetic material. Initially, we perform a parametric optimization of the growth process. Afterward, the optimized parametric growth process implicitly generates an auxetic random material. Namely, the growth process intrinsically entails the formation of an auxetic material. Our approach enables the computation of large-scale auxetic random materials in commodity computers. We also provide numerical results indicating that the computed auxetic materials have close to isotropic linear elastic behavior.

*Keywords:* Porous media, inverse design, random auxetics, growth processes

---

## 1. Introduction

Auxetic structures are materials with negative Poisson's ratio: when stretched they expand perpendicularly to the applied force [23, 26], a seemingly counterintuitive property. Auxetic materials find applications in multiple fields thanks to their excellent shock absorption, fracture toughness, or acoustical and vibrational absorption [55, 46, 22, 27, 44, 40] among other reasons. A substantial amount of research is devoted to the design of auxetic mechanical materials [22, 11, 52] that derive their physical properties from the particular arrangement of their small-scale geometry rather than from the material from which they are made. Recent manufacturing technologies can fabricate complex small-scale structures, and therefore manufacture auxetic materials.

Random materials offer some significant advantages compared to the more widespread periodic materials. In particular, they are more resilient to fabrication-related symmetry-breaking imperfections [39], are capable to smoothly and seamlessly grade material properties and conform to a surface [25], and allow to compute the material geometry in an efficient and scalable way [31]. While most auxetic materials are defined by a repeating periodic structure a distinct line of research is otherwise interested in random auxetic materials [33] since they offer certain advantages over periodic structures [41, 56, 24]. Auxetic polymeric foams [26, 8] were reported in the 80s and are widely used in industrial applications. The most common process to obtain

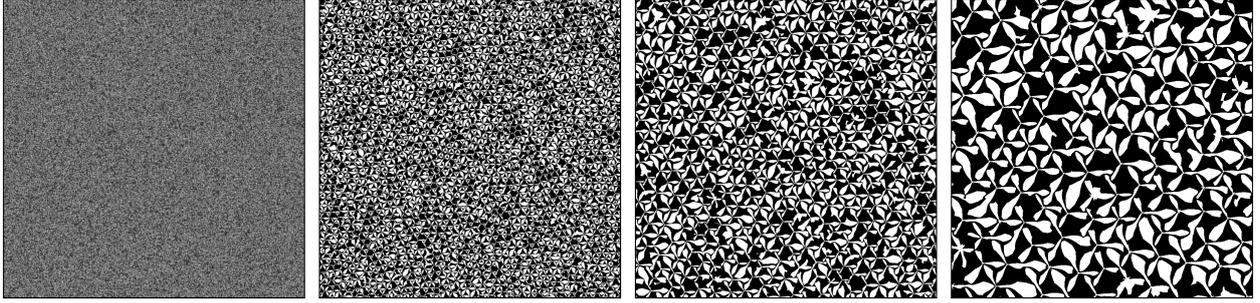


Figure 1: From left to right: large-scale random auxetic porous material with increasing close-up views. The growth process has 43590 nuclei and the computation took 26 minutes on a laptop using a single CPU core.

auxetic foams consists of compressing a conventional flexible cellular foam to force the cell ribs to buckle, producing a re-entrant structure which is then heated to its softening temperature [9, 1]. The geometry of cellular foams is usually idealized and modeled with Voronoi diagrams [16], and some works studying auxetic foams start by modeling a compressed Voronoi diagram [29, 14]. Alternative methods to produce random auxetic materials consider sheets with random perforations [17] or random fiber networks that can be produced by electrospinning [12].

A recent research direction tackles the minimization of the Poisson's ratio of finite two-dimensional random networks consisting of nodes connected by bonds. Different optimization approaches have been proposed, ideally assuming that the elastic behavior of the network remains isotropic during the iterative optimization process. Reid et al. [43] proposed to iteratively prune network bonds, further improved in [42] by modifying, in addition, the position of the network nodes and the stiffness of the network bonds. Hagh et al. [18] starts from a planar triangulation and iteratively removes bonds while avoiding the creation of reentrant polygons. Liu et al. [30] proposed an iterative adjustment of the bond stiffness. Pashine et al. [38] presented a network aging process that decreases the Poisson's ratio.

We advocate for a different perspective on the problem of computing two-dimensional auxetic random materials that is computationally efficient. We propose to compute a porous material [47] with a solid and void phase through a parametric growth process. The growth process is compactly defined by a random point process and two parametric functions controlling the growth law. Our goal is to optimize the parameters of these two functions in order to minimize the Poisson's ratio of the resulting porous material. Importantly, this optimization is only carried out once and allows afterward the immediate generation of random auxetic materials through the process of growth, without requiring any further optimization process. This is in opposition to prior methods [43, 42, 18, 30, 38] that proceed with an iterative optimization, rendering the computation infeasible for very large materials. We enable the computation of large-scale random auxetic porous materials in commodity computers with close to isotropic behavior (see Figure 1).

## 2. Growth process

We consider a growth process [10] in which nuclei are born at the same time. Each nucleus, a point  $x \in \mathbb{R}^2$ , is the origin of a cell. Cells evolve according to a growth law and are forbidden to overlap. At the end of the growth, the union of all cells corresponds to the void phase of the porous material, while its complement in  $\mathbb{R}^2$  corresponds to the solid phase. We start by describing how the nuclei are placed in space and how the law of growth is defined.

A two-dimensional point process  $\Phi \subset \mathbb{R}^2$  defines the nuclei. We restrict ourselves to two different types of point processes: a homogeneous Poisson point process with intensity  $\lambda > 0$ , and a hard-core point process in which the points are forbidden to lie closer together than a minimum distance  $D > 0$  (e.g. the centers of a jammed packing of disks). In particular, we consider the random sequential adsorption (RSA) model [54], where disks with radius  $\frac{D}{2}$  are iteratively added as long as they do not overlap any previously added disk. The center of the disks correspond to the point process and its intensity is  $\lambda = \frac{4A}{\pi D^2}$ , where  $A \approx 0.547$  is the area fraction where saturation occurs [10].

Cells grow through a process of uniform scaling of a compact set  $\mathcal{S} \subset \mathbb{R}^2$  centered around each nucleus. The growth ceases whenever and wherever a cell comes into contact with another one (see Figure 2). This type of growth process has been well studied in the literature when  $\mathcal{S}$  is a convex set and is also known as the Voronoi growth model [37]. Our central idea is to allow  $\mathcal{S}$  to be non-convex, enabling the growth of porous materials with more diverse shapes.

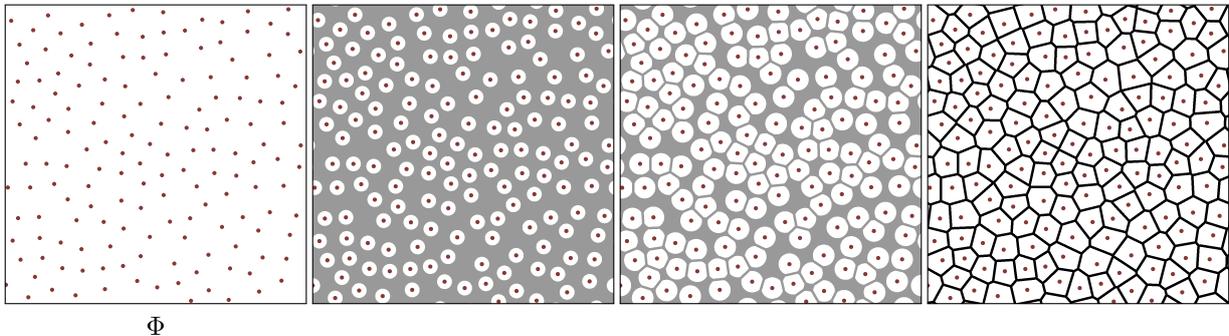


Figure 2: Given a point process  $\Phi$ , the cells grow according to the uniform scaling of a Euclidean disk.

In addition, we control how close to each other cells can grow. This is done by restricting the growth process with what we call a maximum Euclidean radius of growth (see Figure 3). Let  $B_r \subset \mathbb{R}^2$  be a closed Euclidean disk with radius  $r$ , and centered at the origin. Let  $x$  be a point in the boundary of a growing cell. The growth ceases at  $x$  if  $x + B_r$  intersects another growing cell.

We start by defining  $\mathcal{S}$ . Let  $\mathcal{S} \subset \mathbb{R}^2$  be a compact star-shaped set with respect to the origin  $O = (0, 0)$ . That is, for any point  $x \in \mathcal{S}$ , the line segment  $[x, O]$  is contained in  $\mathcal{S}$ . The boundary of  $\mathcal{S}$  can be parameterized

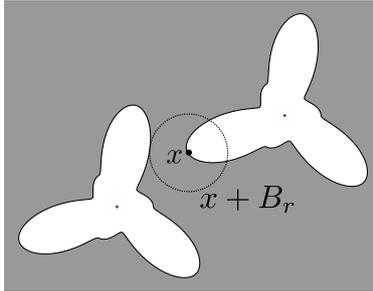


Figure 3: Growth of two cells. The growth ceases at  $x$  since the disk  $x + B_r$  intersects another growing cell.

in polar coordinates by a continuous and periodic function  $\psi_{\mathcal{S}} : [0, 2\pi] \mapsto [r_{\min}, r_{\max}]$ , where  $0 < r_{\min} \leq r_{\max}$  are the minimum and maximum Euclidean distance from  $O$  to the boundary of  $\mathcal{S}$ .

Following [32] we parameterize  $\mathcal{S}$  with a few known values  $(\alpha_i, l_i)_{i>0}$  of the function  $\psi_{\mathcal{S}}(\alpha_i) = l_i$ , and we call them *radial spans*. For simplicity, we always consider  $m > 0$  equally spaced radial spans separated by an angle of  $\alpha_m = \frac{m}{2\pi}$ , and starting at an angle of zero  $\alpha_1 = 0$ . Let  $\alpha_{jk} \in [0, 2\pi]$  be an angle that we seek to interpolate, lying between  $(\alpha_j, l_j)$  and  $(\alpha_k, l_k)$ , such that  $k = \lfloor \alpha_{jk} \alpha_m \rfloor$  and  $j = (k + 1) \bmod m$ . The relative position  $t$  of  $\alpha_{jk}$  between  $\alpha_k$  ( $t = 0$ ) and  $\alpha_j$  ( $t = 1$ ) is given by the fractional part  $t = \alpha_{jk} \alpha_m - \lfloor \alpha_{jk} \alpha_m \rfloor \in [0, 1)$ . Then, we interpolate the value of  $\psi_{\mathcal{S}}(\alpha_{jk})$  by considering the following cubic Hermite spline on the unit interval

$$\psi_{\mathcal{S}}(\alpha_{jk}) = (2t^3 - 3t^2 + 1)l_k + (-2t^3 + 3t^2)l_j \quad (1)$$

The above interpolation ensures that  $\psi_{\mathcal{S}}(\alpha_{jk})$  lies in between  $l_k$  and  $l_j$  by setting the starting and ending tangents of the spline to zero. In addition, we can impose rotational symmetries to the interpolation. An illustration of the interpolation procedure is shown in Figure 4.

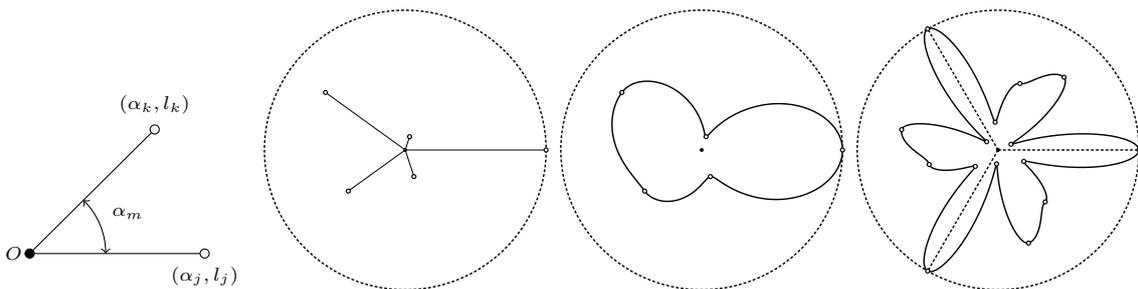


Figure 4: Star-shaped set parameterization and interpolation. Five equally spaced radial spans and the interpolation result. Leftmost figure: imposing three-fold rotational symmetry.

The distance induced by  $\mathcal{S}$  from a point  $p$  to a point  $x$  is [32]

$$d_{\mathcal{S}}(p, x) = \frac{\|x - p\|}{\psi_{\mathcal{S}}(\angle(x - p))}, \quad (2)$$

where  $\angle(\cdot)$  is the angle of a vector with respect to the horizontal axis, and  $\|\cdot\|$  is the Euclidean norm. We

always consider that  $p$  is a cell nucleus and  $x$  is any point of the cell emanating from  $p$ . Importantly,  $d_{\mathcal{S}}(p, x)$  is the factor by which we have to uniformly scale  $\mathcal{S}$ , centered around  $p$ , such that  $x$  lies on its boundary. Therefore, the distance  $d_{\mathcal{S}}$  allows us to compactly define the growth process of a cell.

The maximum Euclidean radius of growth is parameterized by a star-shaped set  $\mathcal{S}^*$ . We consider a polar function  $\psi_{\mathcal{S}^*} : [0, 2\pi] \mapsto [r_{\min}^*, r_{\max}^*]$ , where  $0 < r_{\min}^* \leq r_{\max}^*$  are the minimum and maximum Euclidean radius of growth. Let  $x$  be a point in the boundary of a growing cell emanating from  $p \in \Phi$ . The growth ceases at  $x$  if  $x + B_{\psi_{\mathcal{S}^*}(\angle(x-p))}$  intersects any other growing cell.

The minimal distance between two points in a Poisson point process can be arbitrarily small. Thus, a cell can be empty as its growth was forbidden through  $\mathcal{S}^*$  from the very beginning of the growth process. For a hardcore point process, the minimal distance between two points is at least  $D$  and it suffices to enforce  $D > r_{\max}^*$  in order to guarantee that all cells are allowed to grow.

It is challenging to formulate a continuous growth law [3] since both  $\mathcal{S}$  and  $\mathcal{S}^*$  are not necessarily convex, and  $\mathcal{S}^*$  imposes an atypical constraint on the growth process. We instead resort to a discrete growth process formulation [2], where the cells grow in discrete steps. Therefore, our growth process produces what is known as a *discrete random set*. The general principle is to assign each cell a disjoint discrete set of points.

More precisely, we consider an integer lattice  $L_W = \mathbb{Z}^2 \cap W$  where  $W$  is a bounded subset of  $\mathbb{R}^2$ . Let  $\Phi_W = \Phi \cap W = (p_1, \dots, p_g)$  be the subset of nuclei in  $W$ . The set of lattice points belonging to the cells is given by the union  $C_W = \bigcup_{i=1}^g C_W^i \subset L_W$ , where  $C_W^i$  corresponds to the lattice cell points associated with the nuclei  $p_i$ . Lattice points are incrementally added to  $C_W$  throughout the discrete growth process. Let  $S_1 \subset \mathbb{R}^2$  be the unit square centered at the origin, and let  $\oplus$  denote the Minkowski sum operator. We consider that the void phase of the porous material is given by  $C_W \oplus S_1$ , while the solid one is given by  $(L_W \setminus C_W) \oplus S_1$ . If  $W$  is a rectangle, both phases can be compactly represented with a binary digital image. In all illustrations, the void phase is colored in white and the solid phase in black. Figures 5 and 6 show some examples of the growth process. It is worth recalling that the size of the solid phase can be regulated by  $\mathcal{S}^*$ . For instance, lower values of  $r_{\min}^*$  will lead to narrower porous materials.

In the following, we describe in more detail Algorithm 1 that defines the process of discrete growth. An ordered set  $Q$  contains tuples  $\{x, p\}$  where the first element is a lattice point of  $x \in L_W$  and the second one is a nucleus point of  $p \in \Phi_W$ .  $Q$  is ordered according to the distance  $d_{\mathcal{S}}(p, x)$ . The tuples in  $Q$  identify the points where cell growth can potentially take place. The growth process is divided into two parts, the nucleation phase, and the growth phase.

*Nucleation.* Before the growth starts, for all  $p_i \in \Phi_W$  we insert in  $Q$  the tuple  $\{x_i, p_i\}$ , where  $x_i \in L_W$  is the closest lattice point to  $p_i$ . Ideally,  $L_W$  should be sufficiently large so no two points in  $\Phi_W$  are closest to

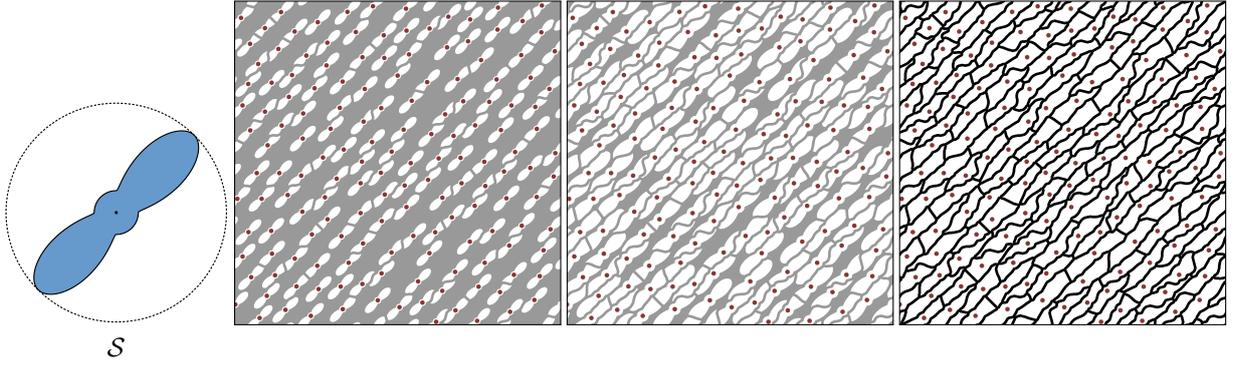


Figure 5: Growth process given by the point process of Figure 2 and the uniform scaling of  $\mathcal{S}$  at the left. In this example  $\mathcal{S}^*$  is a Euclidean disk.

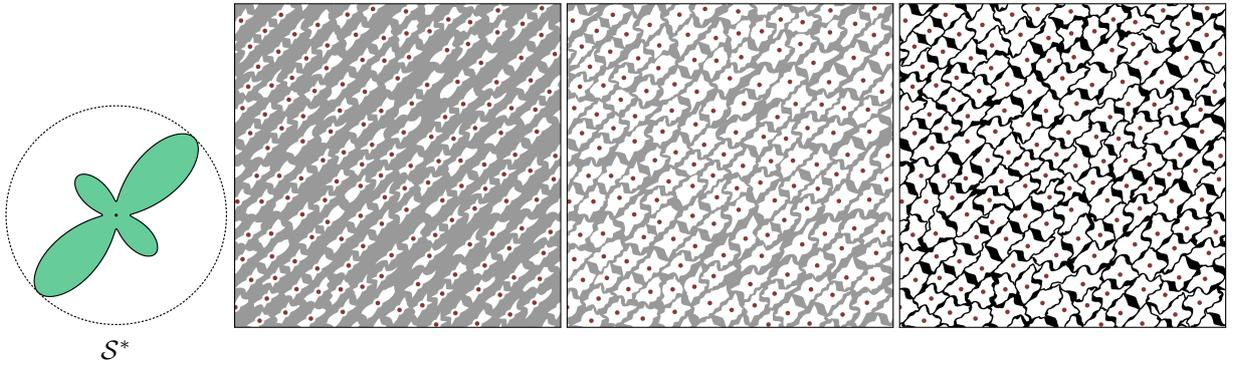


Figure 6: Growth process given by the point process of Figure 2, the uniform scaling of  $\mathcal{S}$  in Figure 5, and the maximum Euclidean radius of growth  $\mathcal{S}^*$  at the left.

the same lattice point in  $L_W$ .

*Growth.* The discrete growth process is simulated as follows. Let  $\{x, p_i\}$  be the tuple in  $Q$  with the smallest distance  $d_{\mathcal{S}}(p_i, x)$  among all tuples.

- (Line 7) First, we remove  $\{x, p_i\}$  from  $Q$ .
- (Lines 8–11) Second, if  $x$  does not already belong to any cell and the Euclidean disk  $B_{\psi_{\mathcal{S}^*}(\angle(x-p_i))}$  does not contain any other cell point we proceed to add  $x$  in  $C_W^i$ . That is, the lattice point  $x$  now belongs to the cell emanating from  $p_i$ .
- (Lines 12–14) Finally, candidate points of growth around a local neighborhood of  $p$  are inserted in  $Q$ . More precisely, we insert in  $Q$  the four tuples  $\{x_n, p_i\}$ , where  $x_n$  is a lattice point in the 4-connected neighborhood of  $x$  in  $L_W$ .

The previous steps are done iteratively until  $Q$  is empty.

---

**Algorithm 1** discreteGrowthProcess( $L_W, \Phi_W, \mathcal{S}, \mathcal{S}^*$ )

---

```
1:  $C_W^i \leftarrow \emptyset$  for all  $C_W^i \in C_W$ 
2: for  $p_i \in \Phi_W$  do ▷ Nucleation
3:   Let  $x_i \in L_W$  be the closest lattice point to  $p_i$ 
4:   Insert in  $Q$  the tuple  $\{x_i, p_i\}$ 
5: end for
6: while  $Q \neq \emptyset$  do ▷ Growth
7:   Let  $\{x, p_i\} \in Q$  with smallest distance  $d_{\mathcal{S}}(p_i, x)$ 
8:   Remove  $\{x, p_i\}$  from  $Q$ 
9:   if  $x \notin C_W$  then
10:     $N \leftarrow L_W \cap \left(x + B_{\psi_{\mathcal{S}^*}}(\angle(x-p_i))\right)$ 
11:    if  $(y \notin C_W) \vee (y \in C_W^i \text{ for all } y \in N)$  then
12:      $C_W^i \leftarrow C_W^i \cup \{x\}$ 
13:     for  $x_n \in \text{neighborhood of } x \text{ in } L_W$  do
14:      if  $x_n \notin C_W$  then
15:       Insert in  $Q$  the tuple  $\{x_n, p_i\}$ 
16:      end if
17:     end for
18:    end if
19:   end if
20: end while
```

---

*Computational complexity of Algorithm 1.* Let  $n$  be the number of points of  $L_W$  and  $g$  be the number of nuclei of  $\Phi_W$ . Since we assume that  $n > g$  the number of iterations of the nucleation loop (lines 2–5) and the growth loop (lines 6–20) is of order  $n$ . The size of  $Q$  is  $O(n)$ . If  $Q$  is implemented as a heap priority queue then the insertion and removal of elements in  $Q$  can be done in logarithmic time (lines 4, 7 and 15). The loop of line 10 (i.e. for all  $y \in N$ ) has  $O(1)$  time complexity, since it depends on the constant size of  $\mathcal{S}^*$ . The loop of line 13 also has  $O(1)$  time complexity (four iterations). Thus, Algorithm 1 has  $O(n \log(n))$  worst-case time complexity. In practice, we observe a quasi-linear runtime with respect to  $n$  (see Section 4.3) primarily because the size of the queue  $Q$  remains comparatively small in proportion to  $n$ .

*Periodic boundary conditions.* Optionally, when  $W$  is a rectangle periodic boundary conditions can be easily imposed by considering that  $L_W$  is a periodic rectangular lattice as well. If  $\Phi$  is an RSA point process, the random adsorption of disks has to be performed considering the periodicity. All results shown in this article feature periodic boundary conditions.

*Cell regularization.* Algorithm 1 is guaranteed to produce cells  $C_W^i$  that are 4-connected sets. However, we observe that when  $\mathcal{S}^*$  is not convex a cell may have a genus higher than zero (i.e. has “holes”). This leads to a solid phase composed of more than one connected component, which is undesirable. To resolve this we fill any hole that a cell may have. Formally, we consider the complement of the unbounded component of the complement of a cell (see Figure 7). We denominate this last step *cell regularization*. Experimental results indicate that cell regularization has on average a very small impact (see Section 4.1).

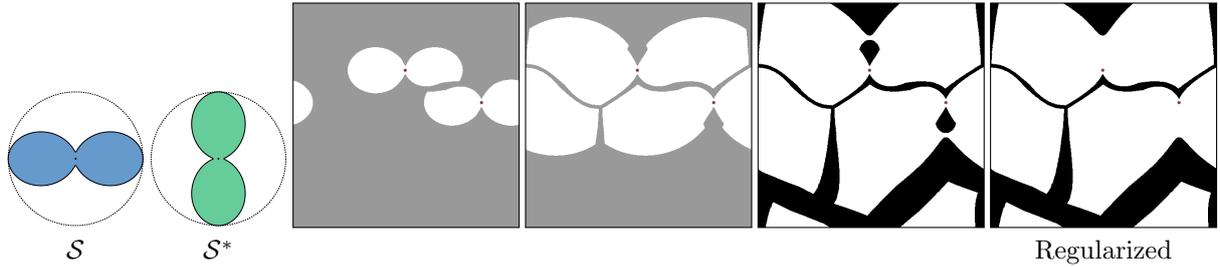


Figure 7: Left:  $\mathcal{S}$  and  $\mathcal{S}^*$  governing the growth process of two nuclei. Middle: evolution of the growth, the holes arise due to the high anisotropy of both  $\mathcal{S}$  and  $\mathcal{S}^*$ , and the particular arrangement of the two nuclei. Right: cell regularization, the two cell holes are filled.

### 3. Elastic behavior of random porous materials

We study the elastic behavior of the random porous materials given by our growth process. We restrict our analysis to linear elasticity which is useful to model the behavior of materials under small deformations. In particular, we apply a procedure known as *stochastic homogenization* [6] that seeks to approximate the average elastic behavior of a random material.

Hooke's law represents the elastic behavior of materials with the linear relation  $\sigma = C\epsilon$ , where  $\sigma$  is the stress,  $\epsilon$  is the strain, and  $C$  is the elasticity tensor (a symmetric and positive definite matrix), in Voigt notation [53]

$$\begin{bmatrix} \sigma_1 \\ \sigma_2 \\ \sigma_{12} \end{bmatrix} = \begin{bmatrix} c_{11} & c_{12} & c_{13} \\ c_{12} & c_{22} & c_{23} \\ c_{13} & c_{23} & c_{33} \end{bmatrix} \begin{bmatrix} \epsilon_1 \\ \epsilon_2 \\ 2\epsilon_{12} \end{bmatrix} \quad (3)$$

*Periodic homogenization* [4] seeks to find the elasticity tensor characterizing a periodic composite material defined from a periodic cell, as the length of the cell tends to zero. In the periodic case, the homogenization is defined on a finite domain (periodic cell) while in the random case is defined on the whole space  $\mathbb{R}^2$ , and cannot be reduced to a problem posed on a finite domain [7].

We approximate the homogenized elasticity tensor coefficients of a random porous material, by means of the so called ‘‘cut-off’’ techniques. Consider a periodized cut  $[0, s]^2$  (a square) of a stationary random material and its corresponding homogenized elasticity tensor  $C^s$ . It was shown in [7] that  $\lim_{s \rightarrow \infty} C^s$  converges, almost surely. Nevertheless,  $s$  needs to be bounded for practical purposes of computation.

In our discrete setting, the size of the cut  $s$  is a positive integer that corresponds to the pixel resolution  $s \times s$  of the porous material image. In all results, we consider a constant  $D = 80$  for the RSA point process and  $\lambda = \frac{4A}{\pi D^2} \approx 0.00011$  for the Poisson point process. Therefore, the two resulting point processes have similar intensity.

We compute the homogenized elasticity tensor  $C^s$  with a publicly available numerical homogenization method

based on the finite element method [5]. The input is an image with values identifying each distinct phase of the periodic material. We impose periodic boundary conditions on the growth process to produce a suitable input image. The solid phase is composed of a linear elastic isotropic material with unit Young's modulus  $E = 1.0$  and Poisson's ratio  $\nu = 0.3$ . Let us recall that the Young's moduli  $E$  is a measure of the tensile stiffness and that the Poisson's ratio is a measure of the deformation (expansion or contraction) of a material in directions perpendicular to the direction of loading.

Let  $\mathbb{E}[\cdot]$  be the expectation of a random variable. We estimate the value of  $\mathbb{E}[C^s]$  with the average of  $k$  independent realizations of the random porous material giving  $(C^s)_i$ ,  $1 < i \leq k$

$$\mathbb{E}[C^s] \approx \overline{C^s} = \frac{1}{k} \sum_{i=1}^k (C^s)_i \quad (4)$$

This is an approximation (Monte Carlo method) frequently used in the context of stochastic homogenization [6]. Let  $std(C^s)$  be the standard deviation of the  $k$  independent realizations. A range of plausible values for  $\mathbb{E}[C^s]$  is  $\left(\overline{C^s} \pm 1.96 \frac{std(C^s)}{\sqrt{k}}\right)$  for a confidence interval with probability 95% (see Figure 8).

In order to evaluate the accuracy of the stochastic homogenization [21], we examine the statistical fluctuations of the material porosity  $p \in [0, 1]$  (the area fraction of the void phase) for a given  $k$  and  $s$ . In particular, we consider the coefficient of variation  $c_v(p) = \frac{std(p)}{\overline{p}}$  that indicates the extent of variability of the porosity in relation to the mean. Increasing the value of  $k$  and  $s$  most likely decreases  $c_v(p)$ . Following [49] we consider that the approximation is sufficiently accurate if  $c_v(p)$  is below a maximum predefined one. In all the following results we always consider  $k = 60$  realizations.

### 3.1. Isotropic elasticity

Since our goal is to minimize the Poisson's ratio of an isotropic material, we give here a brief presentation of how do we determine if the average tensor  $\overline{C^s}$  is close to isotropic, and if so, what are its defining parameters  $E$  and  $\nu$ .

An isotropic material has an elasticity tensor  $C^{iso}$  with only two independent components  $c_{11}^{iso}$  and  $c_{12}^{iso}$

$$C^{iso} = \begin{bmatrix} c_{11}^{iso} & c_{12}^{iso} & 0 \\ c_{12}^{iso} & c_{11}^{iso} & 0 \\ 0 & 0 & \frac{c_{11}^{iso} - c_{12}^{iso}}{2} \end{bmatrix} \quad (5)$$

The relation between the tensor components  $c_{11}^{iso}, c_{12}^{iso}$  and the Young's moduli  $E > 0$  and the Poisson's ratio  $\nu \in [-1, 1]$  is given by

$$E = \frac{(c_{11}^{iso})^2 - (c_{12}^{iso})^2}{c_{11}^{iso}} \quad \nu = \frac{c_{12}^{iso}}{c_{11}^{iso}} \quad (6)$$

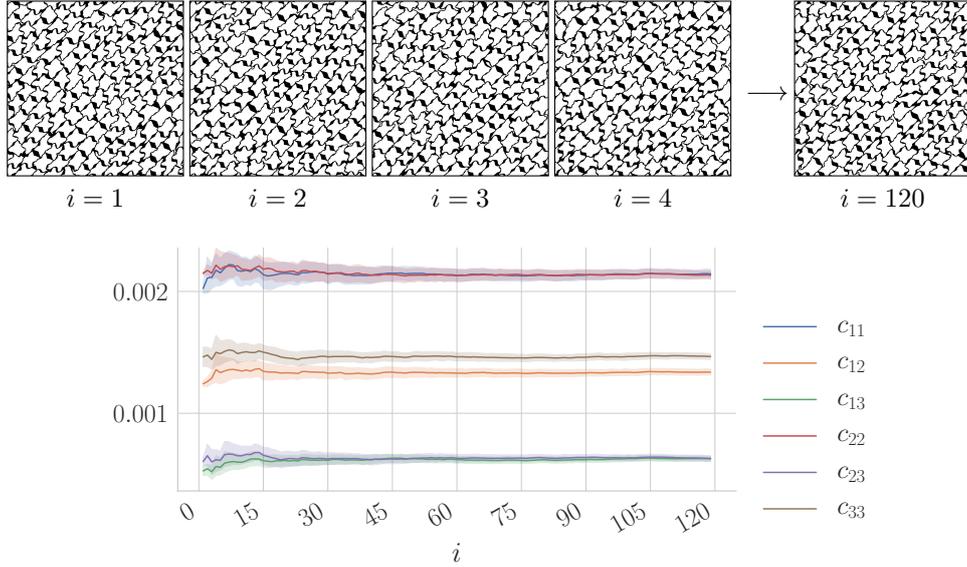


Figure 8: Approximation of the elasticity tensor of the random porous material of Figure 6 in a square with size  $s = 1200$ , and a total of 120 independent realizations. Top: Some realizations. Bottom: mean and confidence interval of each tensor component. As the number of realizations  $i$  increases, the mean stabilizes and the confidence interval becomes narrower. The coefficient of variation of porosity is  $c_v(p) = 0.00409$ .

In practice, it never occurs that the average tensor  $\overline{C^s}$  is perfectly isotropic. Thus, we seek to determine its closest isotropic tensor under some norm [34]. We consider the Frobenius norm for a  $3 \times 3$  matrix  $A$

$$\|A\|_F = \sqrt{\sum_{i=1}^3 \sum_{j=1}^3 |a_{ij}|^2} \quad (7)$$

and minimize the Frobenius norm between  $C^{iso}$  and the average tensor  $\overline{C^s}$

$$\min_{c_{11}^{iso}, c_{12}^{iso}} \|\overline{C^s} - C^{iso}\|_F \quad (8)$$

We provide in Appendix A the closed-form expression of  $c_{11}^{iso}, c_{12}^{iso}$  from  $C^{iso}$  minimizing Equation 8.

We also need a measure of the deviation of  $\overline{C^s}$  from perfect isotropy. We consider the normalized measure  $\delta_{iso}$  used in practice [51, 56]

$$\delta_{iso} = \frac{\|\overline{C^s} - C^{iso}\|_F}{\|\overline{C^s}\|_F} \geq 0 \quad (9)$$

where  $\delta_{iso} = 0$  indicates perfect isotropy, and increasing values indicate divergence from isotropy.

## 4. Results

### 4.1. Three-fold symmetry and isotropy

It is known that two-dimensional periodic structures having three-fold rotational symmetry (invariant to rotations of  $\frac{2\pi}{3}$ ) lead to an isotropic linear elastic behavior [13]. We hypothesize that a resembling property holds in our instance of random growth.

A two-dimensional point process  $\Phi$  is *stationary* if  $\Phi + x$  has the same probability distribution than  $\Phi$ , for all  $x \in \mathbb{R}^2$ .  $\Phi$  is *isotropic* if  $R(a)\Phi$  has the same probability distribution than  $\Phi$ , for all rotations  $R(a)$  of angle  $a \in \mathbb{R}$  around the origin. Both the Poisson point process and the RSA point process are stationary and isotropic point processes [10, 48]. The notion of stationarity and isotropy extends to general random sets, as well.

Let us consider a rotation of  $R(\frac{2\pi}{3}n)$ , for  $n \in \mathbb{Z}$ . The probability distribution of  $\Phi$  is invariant to a rotation  $R(\frac{2\pi}{3}n)$ . Moreover, if  $\mathcal{S}$  and  $\mathcal{S}^*$  are three-fold symmetric then  $R(\frac{2\pi}{3}n)\mathcal{S} = \mathcal{S}$  (same for  $\mathcal{S}^*$ ). Given a point  $p \in \Phi$  then we have

$$R(\frac{2\pi}{3}n)(p + \mathcal{S}) = R(\frac{2\pi}{3}n)p + R(\frac{2\pi}{3}n)\mathcal{S} = R(\frac{2\pi}{3}n)p + \mathcal{S} \quad (10)$$

i.e., the rotation of  $\mathcal{S}$  centered at  $p$  around the origin keeps  $\mathcal{S}$  and  $\mathcal{S}^*$  invariant. This suggests that the probability distribution of the discrete random set given by the growth process is close to being stationary and invariant to rotations  $R(\frac{2\pi}{3}n)$ , which leads us to the following hypothesis:

**Hypothesis 1.** *Let  $\Phi$  be a stationary and isotropic point process. Let  $\mathcal{S}$  and  $\mathcal{S}^*$  be three-fold rotational symmetric. For  $s \rightarrow \infty$  the discrete growth process described in this article gives a porous material with  $\delta_{iso}$  close to zero. Namely, the porous material has close to isotropic linear elastic behavior.*

We have numerically evaluated Hypothesis 1 for a large set of porous materials with different sets  $\mathcal{S}$  and  $\mathcal{S}^*$ , and increasing size  $s$  (see Figure 9). There is strong evidence that as  $s$  increases the average value of  $\delta_{iso}$  decreases and it appears to tend towards a value close to zero.

In all the subsequent results considering three-fold rotational symmetric sets  $\mathcal{S}$  and  $\mathcal{S}^*$  we consider a square image of side length  $s = 1200$  pixels. This size gave among all the tests performed in Figure 9 a maximum coefficient of variation of porosity of  $c_v(p) = 0.03519$  for the Poisson point process, and of  $c_v(p) = 0.01118$  for the RSA point process.

We also evaluate the impact of the cell regularization over the porosity. More precisely, we measure the relative amount of cell holes that are filled during the cell regularization. Let  $\hat{p}$  be the porosity of the porous material before performing the cell regularization. We consider the relative measure  $\zeta_{p,\hat{p}} = \left| \frac{\hat{p}-p}{\hat{p}} \right| \geq 0$ . For all the results in Figure 9 the maximum value of  $\zeta_{p,\hat{p}}$  is 0.06157 and the mean is 0.00433 for the Poisson point process, and maximum 0.01948 and mean 0.00041 for the RSA point process. This indicates that the

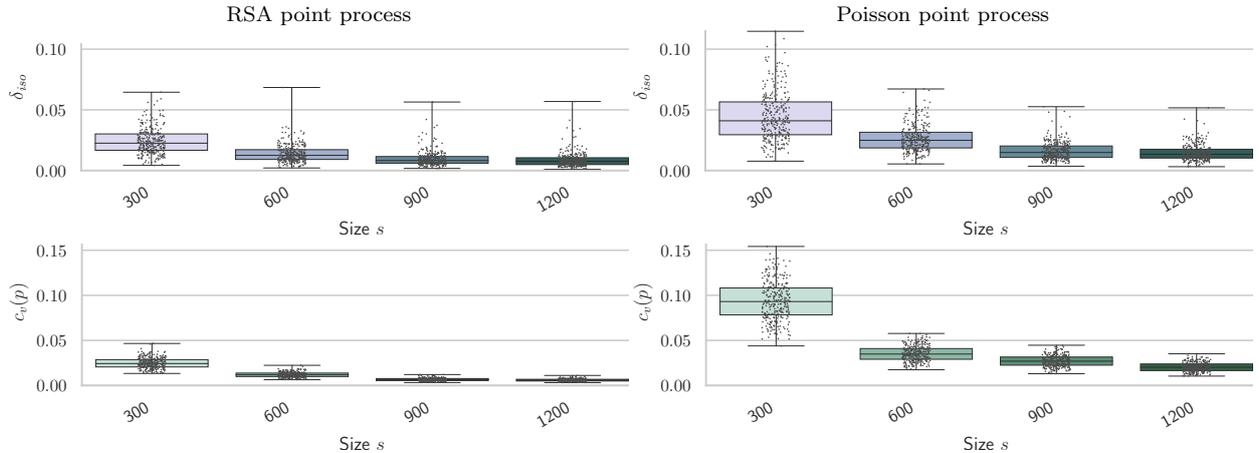


Figure 9: Analysis of the deviation of isotropy  $\delta_{iso}$  and coefficient of variation of the porosity  $c_v(p)$  for a large number of porous materials, and two different point processes (left and right columns).  $\mathcal{S}$  and  $\mathcal{S}^*$  are three-fold rotational symmetric. The colored boxes encompass the interquartile range of values of  $\delta_{iso}$ . We consider 4 different sizes  $s$ , going from 300 to 1200. For each point process and size we enumerate 300 different random radial spans in the range  $\psi_{\mathcal{S}} \in [0.05, 1.0]$  and  $\psi_{\mathcal{S}^*} \in [4.0, 40.0]$ , and 8 different equally spaced angles in  $[0, \frac{2\pi}{3}]$  (three-fold symmetry) considering a total of  $2 \cdot 4 \cdot 300 = 2400$  different porous materials. In general, we observe a clear trend of the average value of  $\delta_{iso}$  and  $c_v(p)$  getting closer to zero as the size  $s$  of the domain increases. We also notice lower average values with the RSA point process compared to the Poisson point process.

effect of the cell regularization on the porosity is almost negligible, and is particularly small for the RSA point process.

#### 4.2. Parametric optimization

By this point, we have all the ingredients to formulate the parametric optimization of the growth process. Let  $\overline{\mathcal{C}^s}(\psi_{\mathcal{S}}, \psi_{\mathcal{S}^*})$  be the average elasticity tensor of a porous material parameterized by the functions  $\psi_{\mathcal{S}}$  (star-shaped distance) and  $\psi_{\mathcal{S}^*}$  (maximum Euclidean radius of growth). Let  $\mathbb{F} : M \mapsto \mathbb{R}$  be the objective function, where  $M$  is a  $3 \times 3$  symmetric matrix. Our objective is to minimize

$$\arg \min_{\psi_{\mathcal{S}}, \psi_{\mathcal{S}^*}} \mathbb{F}(\overline{\mathcal{C}^s}(\psi_{\mathcal{S}}, \psi_{\mathcal{S}^*})) = \frac{c_{12}^{iso}}{c_{11}^{iso}} \quad (11)$$

where  $c_{11}^{iso}, c_{12}^{iso}$  are derived from  $\overline{\mathcal{C}^s}(\psi_{\mathcal{S}}, \psi_{\mathcal{S}^*})$  considering Equation A.4. In order to obtain a physical behavior close to isotropic elasticity, we constrain  $\mathcal{S}$  and  $\mathcal{S}^*$  to be three-fold rotational symmetric (see Hypothesis 1).

The input of function  $\mathbb{F}$  is expensive to evaluate and has no closed-form. In this case, Bayesian optimization [35] is often a good choice to tackle the optimization. In particular, we have used Bayesian optimization using Gaussian processes as implemented in the Python library *scikit-optimize* [50]. Both functions  $\psi_{\mathcal{S}}$  and  $\psi_{\mathcal{S}^*}$  are parameterized with  $m = 8$  radial spans. We set the first radial span of  $\psi_{\mathcal{S}}$  to a constant value of 1, removing an unnecessary degree of freedom. For the Bayesian optimization, we set a maximum number of 150 evaluations of function  $\mathbb{F}$  and 50 random initial guesses.

Figures 10 to 12 provide minimization results of the Poisson's ratio with increasingly larger optimization domains of  $\psi_{\mathcal{S}^*}$ , and a constant optimization domain of  $\psi_{\mathcal{S}} \in [0.05, 1.0]$ . We distinguish two clear trends from the optimization results. First, the RSA point process is able to achieve a lower Poisson's ratio  $v$  compared to the Poisson point process. Second, increasing the domain of function  $\psi_{\mathcal{S}^*}$  leads to lower Poisson's ratio, since we expand in turn the space of achievable porous materials. Overall, we recommend the use of a hardcore point process instead of a Poisson point process to reach a lower Poisson's ratio and lower variance of the material properties using the same area (see Figure 9).

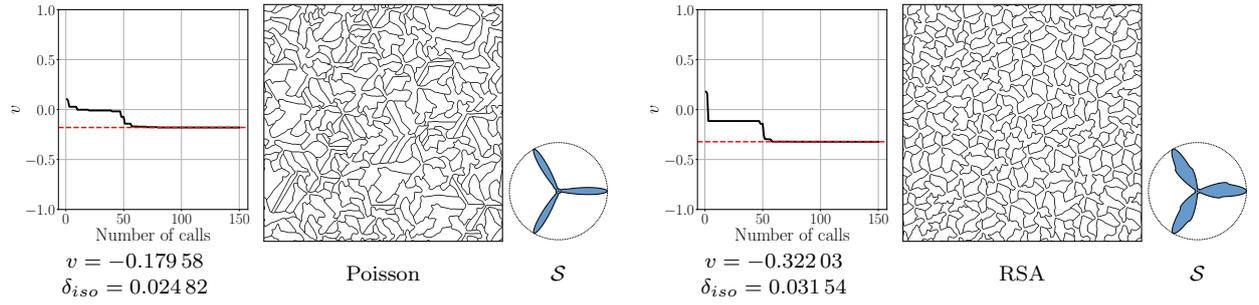


Figure 10: Optimization with constant  $\psi_{\mathcal{S}^*} = 4.0$ .

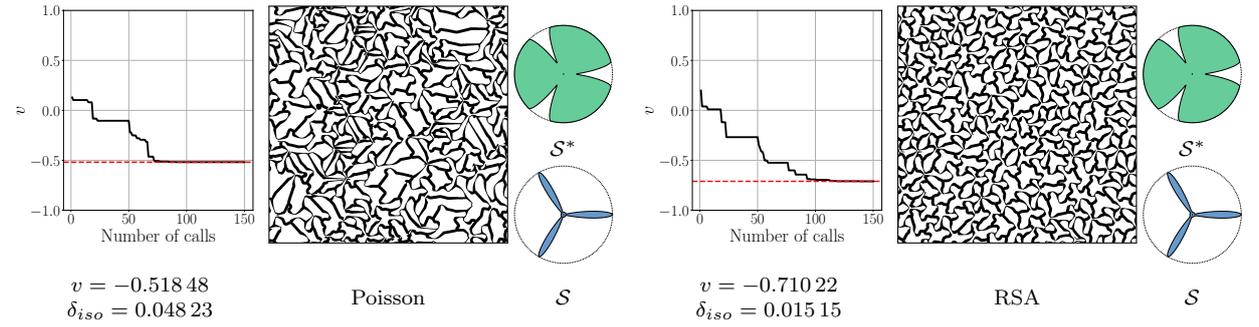


Figure 11: Optimization with variable  $\psi_{\mathcal{S}^*} \in [4.0, 15.0]$

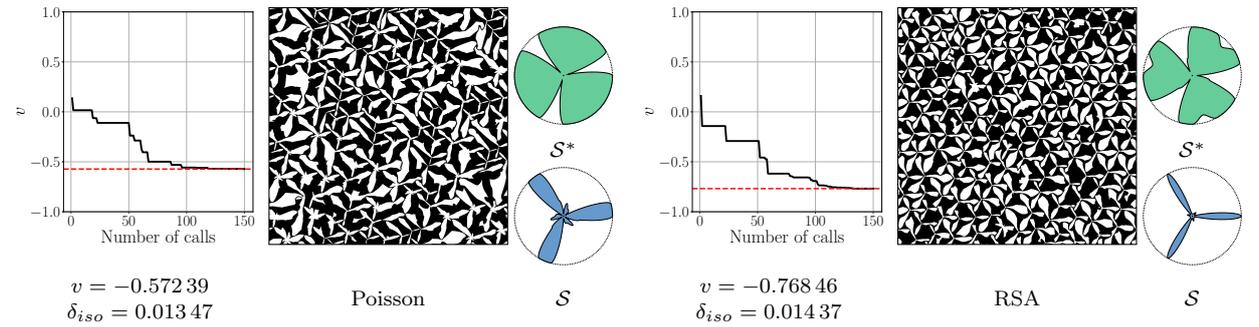
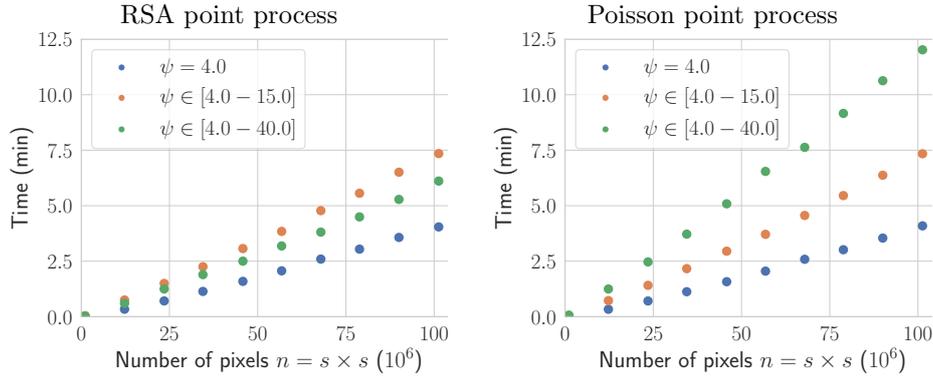


Figure 12: Optimization with variable  $\psi_{\mathcal{S}^*} \in [4.0, 40.0]$

#### 4.3. Computational efficiency

We evaluate the efficiency of the growth process computation. To this end, we performed a series of timing tests shown in Table 1 where we gradually increase the number of pixels  $n$  of the output image. For a range



$\psi_{\mathcal{S}^*}$	RSA				Poisson			
	(a)	(b)	(c)	(d)	(a)	(b)	(c)	(d)
4.0	0.995%	453%	3.07%	79.4	0.998%	456%	3.19%	79.2
[4.0, 15.0]	0.367%	342%	2.98%	314	0.402%	355%	2.59%	294
[4.0, 40.0]	0.363%	226%	2.52%	759	0.171%	226%	1.47%	2930

Table 1: Timing tests performed on images with size ranging  $n = [1, 100]$  million pixels. We compute the growth process with a laptop equipped with an Intel i7-8650U. For each optimization result in Figures 10 to 12 we perform 10 tests with gradually increasing size  $n$  and plot the corresponding time in minutes. The table below shows the following additional statistics (considering the average value between all the tests). (a): Relative time spent computing the regularization with respect to the total time of computation. (b): Number of iterations in the main loop (line 6 of Algorithm 1) divided by  $n$ . (c): Average size of the queue  $Q$  divided by  $n$ . (d): Average number of iterations in line 11 of Algorithm 1.

of  $n = [1, 100]$  million pixels, we observe a nearly linear correlation between  $n$  and the time of computing. Based on the statistics shown in Table 1, we consider that this is mainly explained by the fact that the relative average size of the queue  $Q$  is below 4% (column (c)), amortizing the logarithmic cost of insertion and deletion of elements in  $Q$ . In addition, the relative time spent computing the cell regularization is below 1% (column (a)) indicating that the bulk of the computational cost is in the growth algorithm.

Finally, we observe among the tests different scaling laws, depending on the input sets  $\mathcal{S}$  and  $\mathcal{S}^*$  and type of point process. We infer that these differences are mostly explained by a trade-off between the number of iterations in the main loop (column (b)) and the average number of iterations in line 11 of Algorithm 1 (column (d)). Both statistics are tightly related to  $\mathcal{S}^*$ : a larger area of  $\mathcal{S}^*$  implies fewer iterations of (b) at the expense of more iterations of (d).

## 5. Conclusions and future work

We have presented a computational approach based on parametric growth processes to minimize the Poisson's ratio of random porous materials. The results attain a negative Poisson's ratio while providing a numerical analysis showing close to linear elastic isotropic behavior. Increasing the number of parameters of  $\psi_{\mathcal{S}}$  and  $\psi_{\mathcal{S}^*}$  and their domain further improves the minimization. In practice, our approach is able to compute large-scale porous materials in a reasonable time. This capability is most valuable for applications that need to generate large quantities of random porous materials.

With a parametric growth process, it is possible to generate heterogeneous materials by considering that both functions  $\psi_{\mathcal{S}}$  and  $\psi_{\mathcal{S}^*}$  are varying in the space (see Figure 2). One of the main advantages of our approach over periodic structures is that the transitions between different material properties do not need to be handled [15, 20], as they are intrinsically captured by the parameters governing the growth process.

Our approach also opens up different opportunities for future work. First, to parallelize Algorithm 1 and significantly increase its performance. Second, to extend Algorithm 1 to the 3D case by considering a 3D growth process and adapting the parameterization of  $\mathcal{S}$  and  $\mathcal{S}^*$ . This may be of interest to the inverse design of random 3D porous materials, e.g. random foams are of interest for acoustic absorption [19] or tissue engineering [28]. Finally, our approach may open the door to other applications besides the minimization of the Poisson’s ratio of linearly elastic materials. For example, to tackle the inverse design and optimization of porous materials [45, 25] using our growth process. We conjecture that in most cases it will be sufficient to replace the objective function of Equation 11 with another one of interest.

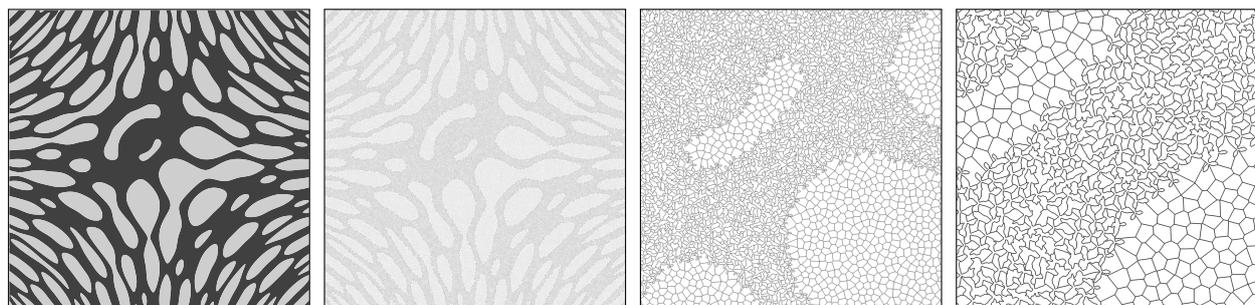


Figure 13: Spatially varying growth process with two different growth functions. Left: Input heterogeneous field, the darker regions correspond to the optimized  $\psi_{\mathcal{S}}$  in Figure 10, while the rest correspond to growth according to the Euclidean disk.

## 6. Code and data availability

An open source implementation of our approach can be found at <https://future-url-code>. We provide an script to automate the generation of all paper results and figures, apart from the manually done Figure 3. All the results of this article can be downloaded from <https://future-url-data>.

## Acknowledgements

This work was partly supported by ANR MuFFin (ANR-17-CE10-0002).

## References

- [1] Kim Alderson, Andrew Alderson, Naveen Ravirala, Virginia Simkins, and Philip Davies. Manufacture and characterisation of thin flat and curved auxetic foam sheets. *physica status solidi (b)*, 249(7):1315–1321, 2012.
- [2] Giacomo Aletti, Enea G Bongiorno, and Vincenzo Capasso. Statistical aspects of fuzzy monotone set-valued stochastic processes. application to birth-and-growth processes. *Fuzzy Sets and Systems*, 160(21):3140–3151, 2009.
- [3] Giacomo Aletti, Enea G Bongiorno, and Vincenzo Capasso. Integration in a dynamical stochastic geometric framework. *ESAIM: Probability and Statistics*, 15:402–416, 2011.

- [4] Grégoire Allaire. Homogenization and two-scale convergence. *SIAM Journal on Mathematical Analysis*, 23(6):1482–1518, 1992.
- [5] Erik Andreassen and Casper Schousboe Andreassen. How to determine composite material properties using numerical homogenization. *Computational Materials Science*, 83:488–495, 2014.
- [6] Xavier Blanc, Claude Le Bris, and Frédéric Legoll. Some variance reduction methods for numerical stochastic homogenization. *Philos. T. R. Soc. A*, 374(2066):20150168, 2016.
- [7] Alain Bourgeat and Andrey Piatnitski. Approximations of effective coefficients in stochastic homogenization. *Annales de l’IHP Probabilités et statistiques*, 40(2):153–165, 2004.
- [8] BD Caddock and KE Evans. Microporous materials with negative Poisson’s ratios. I. Microstructure and mechanical properties. *Journal of Physics D: Applied Physics*, 22(12):1877, 1989.
- [9] N Chan and KE Evans. Fabrication methods for auxetic foams. *Journal of materials Science*, 32(22):5945–5953, 1997.
- [10] Sung Nok Chiu, Dietrich Stoyan, Wilfrid S Kendall, and Joseph Mecke. *Stochastic geometry and its applications*. John Wiley & Sons, 2013.
- [11] John Dagdelen, Joseph Montoya, Maarten de Jong, and Kristin Persson. Computational prediction of new auxetic materials. *Nature communications*, 8(1):1–8, 2017.
- [12] S. Domaschke, A. Morel, G. Fortunato, and AE. Ehret. Random auxetics from buckling fibre networks. *Nature communications*, 10(1):1–8, 2019.
- [13] Sandra Forte and Maurizio Vianello. Symmetry classes for elasticity tensors. *Journal of Elasticity*, 43(2):81–108, 1996.
- [14] Ruicong Gao, Dong Li, Liang Dong, and Xin Wang. Numerical analysis of the mechanical properties of 3D random Voronoi structures with negative Poisson’s ratio. *physica status solidi (b)*, page 1800539, 2019.
- [15] Eric Garner, Helena MA Kolken, Charlie CL Wang, Amir A Zadpoor, and Jun Wu. Compatibility in microstructural optimization for additive manufacturing. *Additive Manufacturing*, 26:65–75, 2019.
- [16] Lorna J Gibson and Michael F Ashby. *Cellular solids: structure and properties*. Cambridge university press, 1999.
- [17] Joseph N Grima, Luke Mizzi, Keith M Azzopardi, and Ruben Gatt. Auxetic perforated mechanical metamaterials with randomly oriented cuts. *Advanced materials*, 28(2):385–389, 2016.
- [18] Varda F Hagh and MF Thorpe. Disordered auxetic networks with no reentrant polygons. *Physical Review B*, 98(10):100101, 2018.
- [19] Fusheng Han, Gary Seiffert, Yuyuan Zhao, and Barry Gibbs. Acoustic absorption behaviour of an open-celled aluminium foam. *Journal of Physics D: Applied Physics*, 36(3):294, 2003.
- [20] Jingqiao Hu, Ming Li, Xingtong Yang, and Shuming Gao. Cellular structure design based on free material optimization under connectivity control. *Computer-Aided Design*, page 102854, 2020.
- [21] Toufik Kanit, Samuel Forest, Isabelle Galliet, Valérie Mounoury, and Dominique Jeulin. Determination of the size of the representative volume element for random composites: statistical and numerical approach. *Int. J. Solids Struct.*, 40(13-14):3647–3679, 2003.
- [22] H MA Kolken and AA Zadpoor. Auxetic mechanical metamaterials. *RSC Advances*, 7(9):5111–5129, 2017.
- [23] AG Kolpakov. Determination of the average characteristics of elastic frameworks. *J. Appl. Math. Mech.*, 49(6):739–745, 1985.
- [24] Jagdish A Krishnaswamy, Federico C Buroni, Roderick Melnik, Luis Rodriguez-Tembleque, and Andres Saez. Design of polymeric auxetic matrices for improved mechanical coupling in lead-free piezocomposites. *Smart Materials and Structures*, 29(5):054002, 2020.
- [25] Siddhant Kumar, Stephanie Tan, Li Zheng, and Dennis M Kochmann. Inverse-designed spinodoid metamaterials. *npj Computational Materials*, 6(1):1–10, 2020.
- [26] Roderic Lakes. Foam structures with a negative Poisson’s ratio. *Science*, 235:1038–1041, 1987.
- [27] Roderic S Lakes. Negative-Poisson’s-ratio materials: auxetic solids. *Annual review of materials research*, 47:63–81, 2017.
- [28] Robert Lanza, Robert Langer, Joseph P Vacanti, and Anthony Atala. *Principles of tissue engineering*. Academic press, 2020.
- [29] Dong Li, Liang Dong, Jianhua Yin, and Roderic S Lakes. Negative Poisson’s ratio in 2D Voronoi cellular solids by biaxial compression: a numerical study. *Journal of materials science*, 51(14):7029–7037, 2016.
- [30] Jun Liu, Yunhuan Nie, Hua Tong, and Ning Xu. Realizing negative Poisson’s ratio in spring networks with close-packed lattice geometries. *Physical Review Materials*, 3:055607, 2019.
- [31] Jonàs Martínez, Jérémie Dumas, and Sylvain Lefebvre. Procedural Voronoi foams for additive manufacturing. *ACM Transactions on graphics*, 35(4):44:1–44:12, 2016.
- [32] Jonàs Martínez, Mélina Skouras, Christian Schumacher, Samuel Hornus, Sylvain Lefebvre, and Bernhard Thomaszewski. Star-shaped metrics for mechanical metamaterial design. *ACM Transactions on Graphics*, 38(4):1–13, 2019.
- [33] MJ Mirzaali, H Pahlavani, and AA Zadpoor. Auxeticity and stiffness of random networks: Lessons for the rational design of 3D printed mechanical metamaterials. *Applied Physics Letters*, 115(2):021901, 2019.
- [34] Maher Moakher and Andrew N Norris. The closest elastic tensor of arbitrary symmetry to an elasticity tensor of lower symmetry. *Journal of Elasticity*, 85(3):215–263, 2006.
- [35] Jonas Moćkus. On Bayesian methods for seeking the extremum. In *Optimization techniques IFIP technical conference*, pages 400–404. Springer, 1975.
- [36] Andrew Norris. The isotropic material closest to a given anisotropic material. *Journal of Mechanics of Materials and Structures*, 1(2):223–238, 2006.
- [37] Atsuyuki Okabe, Barry Boots, Kokichi Sugihara, and Sung Nok Chiu. *Spatial tessellations: concepts and applications of Voronoi diagrams*. John Wiley & Sons, 2000.
- [38] Nidhi Pashine, Daniel Hexner, Andrea J Liu, and Sidney R Nagel. Directed aging, memory, and nature’s greed. *Science*

- advances*, 5(12), 2019.
- [39] Carlos M Portela, A Vidyasagar, Sebastian Krödel, Tamara Weissenbach, Daryl W Yee, Julia R Greer, and Dennis M Kochmann. Extreme mechanical resilience of self-assembled nanolabyrinthine materials. *Proceedings of the National Academy of Sciences*, 117(11):5686–5693, 2020.
  - [40] Guangzhao Qin and Zhenzhen Qin. Negative Poisson’s ratio in two-dimensional honeycomb structures. *npj Computational Materials*, 6(1):1–6, 2020.
  - [41] Daniel Rayneau-Kirkhope, Silvia Bonfanti, and Stefano Zapperi. Density scaling in the mechanics of a disordered mechanical meta-material. *Applied Physics Letters*, 114(11):111902, 2019.
  - [42] Daniel R Reid, Nidhi Pashine, Alec S Bowen, Sidney R Nagel, and Juan J de Pablo. Ideal isotropic auxetic networks from random networks. *Soft Matter*, 15(40):8084–8091, 2019.
  - [43] Daniel R Reid, Nidhi Pashine, Justin M Wozniak, Heinrich M Jaeger, Andrea J Liu, Sidney R Nagel, and Juan J de Pablo. Auxetic metamaterials from disordered networks. *Proceedings of the National Academy of Sciences*, 115(7):1384–1390, 2018.
  - [44] Xin Ren, Raj Das, Phuong Tran, Tuan Duc Ngo, and Yi Min Xie. Auxetic metamaterials and structures: A review. *Smart materials and structures*, 27(2):023001, 2018.
  - [45] Henrik Ronellenfitsch, Norbert Stoop, Josephine Yu, Aden Forrow, and Jörn Dunkel. Inverse design of discrete mechanical metamaterials. *Physical Review Materials*, 3:095201, 2019.
  - [46] Krishna Kumar Saxena, Raj Das, and Emilio P Calius. Three decades of auxetics research – materials with negative Poisson’s ratio: a review. *Advanced Engineering Materials*, 18(11):1847–1870, 2016.
  - [47] Craig Schroeder, William C Regli, Ali Shokoufandeh, and Wei Sun. Computer-aided design of porous artifacts. *Computer-aided design*, 37(3):339–353, 2005.
  - [48] Dietrich Stoyan and Martin Schlather. Random sequential adsorption: relationship to dead leaves and characterization of variability. *Journal of Statistical Physics*, 100(5-6):969–979, 2000.
  - [49] M Stroeven, H Askes, and LJ Sluys. Numerical determination of representative volumes for granular materials. *Computer Methods in Applied Mechanics and Engineering*, 193(30-32):3221–3238, 2004.
  - [50] Head Tim, Louppe Gilles, and Shcherbatyi Iaroslav. scikit-optimize 0.7.4.
  - [51] VP Tran, J Guilleminot, Sébastien Brisard, and Karam Sab. Stochastic modeling of mesoscopic elasticity random field. *Mechanics of Materials*, 93:1–12, 2016.
  - [52] Panagiotis Vogiatzis, Shikui Chen, Xiao Wang, Tiantian Li, and Lifeng Wang. Topology optimization of multi-material negative poisson’s ratio metamaterials using a reconciled level set method. *Computer-Aided Design*, 83:15–32, 2017.
  - [53] Woldemar Voigt. *Lehrbuch der kristallphysik*, volume 962. Teubner Leipzig, 1928.
  - [54] B Widom. Random sequential addition of hard spheres to a volume. *The Journal of Chemical Physics*, 44(10):3888–3894, 1966.
  - [55] Wei Yang, Zhong-Ming Li, Wei Shi, Bang-Hu Xie, and Ming-Bo Yang. Review on auxetic materials. *Journal of materials science*, 39(10):3269–3279, 2004.
  - [56] O Zerhouni, MG Tarantino, and K Danas. Numerically-aided 3D printed random isotropic porous materials approaching the Hashin-Shtrikman bounds. *Composites Part B: Engineering*, 156:344–354, 2019.

## Appendix A. Derivation of $c_{11}^{iso}$ and $c_{12}^{iso}$ from Equation 8

For the three-dimensional case, the minimizing values of  $c_{11}^{iso}$ ,  $c_{12}^{iso}$  were derived, for instance, in [36]. We were not able to find any previous work concerning the simpler two-dimensional case. Nevertheless, the values minimizing Equation 8 can be derived by simple calculus. Consider the function  $f$  with two arguments  $c_{11}^{iso}$  and  $c_{12}^{iso}$

$$\begin{aligned}
 f &= \|C - C^{iso}\|_F^2 \\
 &= (c_{11} - c_{11}^{iso})^2 + 2(c_{12} - c_{12}^{iso})^2 + (c_{22} - c_{11}^{iso})^2 + 2c_{13}^2 + 2c_{23}^2 + \left(c_{33} - \frac{c_{11}^{iso} - c_{12}^{iso}}{2}\right)^2
 \end{aligned} \tag{A.1}$$

whose first-order partial derivatives are

$$\begin{aligned}\frac{\partial f}{\partial c_{11}^{iso}} &= -c_{33} - 2(c_{22} - c_{11}^{iso}) + \frac{c_{11}^{iso} - c_{12}^{iso}}{2} - 2(c_{11} - c_{11}^{iso}) \\ \frac{\partial f}{\partial c_{12}^{iso}} &= c_{33} - 4(c_{12} - c_{12}^{iso}) - \frac{c_{11}^{iso} - c_{12}^{iso}}{2}\end{aligned}\tag{A.2}$$

The Hessian matrix  $H$  of  $f$

$$H = \begin{bmatrix} \frac{\partial^2 f}{\partial (c_{11}^{iso})^2} & \frac{\partial^2 f}{\partial c_{11}^{iso} \partial c_{12}^{iso}} \\ \frac{\partial^2 f}{\partial c_{12}^{iso} \partial c_{11}^{iso}} & \frac{\partial^2 f}{\partial (c_{12}^{iso})^2} \end{bmatrix} = \begin{bmatrix} \frac{9}{2} & -\frac{1}{2} \\ -\frac{1}{2} & \frac{9}{2} \end{bmatrix}\tag{A.3}$$

is positive definite everywhere and it follows that  $f$  is a convex function. Since  $\frac{\partial^2 f}{\partial (c_{11}^{iso})^2} = \frac{\partial^2 f}{\partial (c_{12}^{iso})^2} > 0$  the function  $f$  has a global minimum at

$$\begin{aligned}c_{11}^{iso} &= \frac{1}{20}(9(c_{11} + c_{22}) + 2c_{12} + 4c_{33}) \\ c_{12}^{iso} &= \frac{1}{20}(c_{11} + c_{22} + 18c_{12} - 4c_{33})\end{aligned}\tag{A.4}$$

found by solving the system of linear equations from Equation A.2 being equal to zero.