SuMo BIOMATERIALS CHALMERS

Spatial modeling of micro-structure in porous material

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Goal

Characterizing and **modeling** micro-structures of porous materials in order to better understand and control their properties

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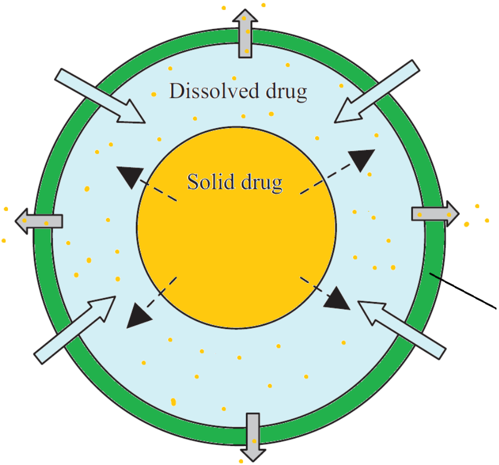
Taking germ-grain models to the next level!?

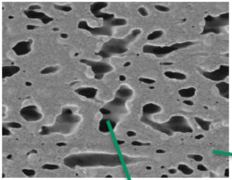
Motivation: Controlled drug release

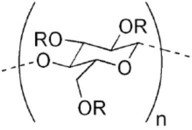


Pharmaceutical coating

[KMBA11]

Polymer blended coating





Ethyl cellulose (EC)

R=H or CH_2CH_3

Hydroxypropyl cellulose (HPC)

R=H or $CH_2CH(OH)CH_3$

Summary: Important data features

- From images
  - isotropic surface, but **anisotropic** cross-section
- Results from experiments
  - film is permeable $\rightarrow$ percolation path through sample
  - pore connectivity is essential
- Results from 2D image analysis
  - HPC area fraction and **pore shape** significant factors for permeability
  - number of endpoints in skeleton good measure of pore shape

Conclusions prior to modeling

3D modeling with marked point processes

Step I Germ-grain models for the surfaces

- stack of marked point processes

$$\Phi = \{[X_i, M_i] : \; i = 1, \; 2, \ldots\}$$

- assign shape parameter(s) to points as marks M_i defining the grains

Step II Link surfaces by displacement vectors capturing the chain like HPC structure in z-direction

- skeleton
- mark correlation

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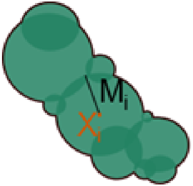
Germ-Grain models

Based on marked point process $\Phi_M = \{[X_i, M_i] : \; i = 1, \; 2, \ldots\}$

$$\Xi = \bigcup_{i=1}^{\infty} (\underbrace{G_{M_i}}_{grain} + \underbrace{X_i}_{germ})$$



- $\Phi = \{X_i\}$ point process
- G_{M_i} (random) compact set defined by parameter(s) M_i
- $(G_{M_i} + X_i) = \{x + X_i : \; x \in G_{M_i}\}$

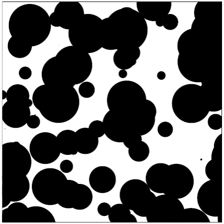
Boolean model

Overlapping sphere system $\Xi = \bigcup_{i=1}^{\infty} (\underbrace{G_{M_i}}_{grain} + \underbrace{X_i}_{germ})$

- $\{X_i\}$ (in)homogeneous Poisson process
- $G_{M_i} = b(o, M_i)$ with fixed or random radii $M_i : \ \Omega \to (0, \infty)$

\+ Well known properties

— Limited variety of generated structures



Quermass-interaction model

Adding morphological interaction between grains s.t. absolutely continuous w.r.t. marked Poisson process with density $\propto exp(-H_{\Phi}^{\theta})$

$$H_{\Phi_W}^{\theta} = \theta_1 \mathcal{A}(\Phi_W) + \theta_2 \mathcal{L}(\Phi_W) + \theta_3 \chi(\Phi_W)$$

$\theta = (\theta_1, \theta_2, \theta_3) \in \mathbb{R}^3$ for Quermass functionals

$\mathcal{A}$ area
$\mathcal{L}$ perimeter
χ Euler number (# objects - # wholes)

$\Phi_W = \{X_i\}$ Poisson process in observation window $W \in \mathbb{R}^d$

Quermass-interaction model: Examples

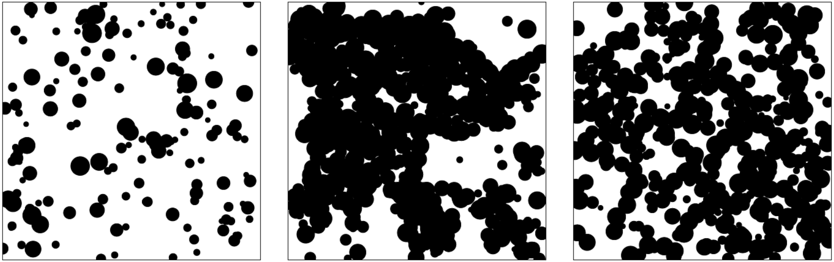


Figure : Examples based on marked Poisson process with intensity λ and spheres with radii $M_i \sim U[0.5, 2]$: left $\lambda = 0.1$, $\theta = (0.2, 0, 0)$, middle $\lambda = 0.2$, $\theta = (0, 0.4, 0)$, right $\lambda = 0.1$, $\theta = (0, 0, 1)$

[DLS14]

Quermass-interaction model

Adding morphological (Quermass) interaction between grains via

$$H^{\theta}_{\Phi_W} = \theta_1 \mathcal{A}(\Phi_W) + \theta_2 \mathcal{L}(\Phi_W) + \theta_3 \chi(\Phi_W)$$

+ Large variety of generated structures

− Complicated parameter estimation based on K chosen test functions f_k for Takacs-Fiksel method

$$(\hat{\lambda}_\Phi,\ \hat{\theta}) := arg \min_{\lambda_\Phi, \theta} \sum_{k=1}^{K} (\underbrace{C_W^{\lambda_\Phi, \theta}(\Phi; f_k)}_{r.v.})^2$$

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Multi-layer modeling of pore connectivity

Task: Capture heterogeneity and correlation of chain like HPC

- Continuum percolation
  - Boolean model with dependent growth of radii: spheres grow until they have touched k neighbors
  - random-connection model based on a connection function

Multi-layer modeling of pore connectivity

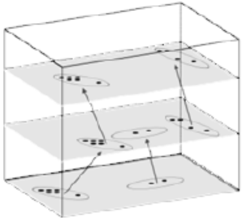
Task: Capture heterogeneity and correlation of chain like HPC

- Continuum percolation
  - Boolean model with dependent growth of radii: spheres grow until they have touched k neighbors
  - random-connection model based on a connection function
- **Markov chain** as stationary process in 3D based on displacement vectors as well as birth and death rates

Example: Matérn process Markov chain

Spatial birth-and-death process with random displacement[1]

- Stack of $s \geq 1$ elliptical Matérn cluster processes $\{S_n^{(s)},\ s \geq 1\}$ in $\mathbb{R}^2$
- High correlation of center locations in z-direction
  - stationary Markov chain with small displacements between $0 < r_{min} < r_{max} \in \mathbb{R}$ of clusters
  - births and deaths of clusters



[1][SHT+11]

Example: Matérn process Markov chain ctd.

Underlying Poisson process $\{\Phi_n^{(s)},\ n \geq 1\}$ with intensity λ defined by

- **Births**: $\{B_n^{(s)}\}$ Poisson process with intensity λ_B
- **Deaths**: $\{\delta_n^{(s)}\}$ $i.i.Ber(p)$ s.t. $\lambda p + \lambda_B = \lambda$, independent of $\{B_n^{(s)}\}$
- **Displacement vectors**: $\{D_n^{(s)}\}$ $i.i.U$ on $b(o, r_{max}) \setminus b(o, r_{min})$, independent of $\{B_n^{(s)}\}$ and $\{\delta_n^{(s)}\}$

$$\{\Phi_n^{(s+1)}\} = \bigcup_{i:\delta_i^{(s)}=1} \{\Phi_j^{(s)} + D_j^{(s)}\} \cup \{B_n^{(s)}\}$$

Example: Matérn process Markov chain ctd.

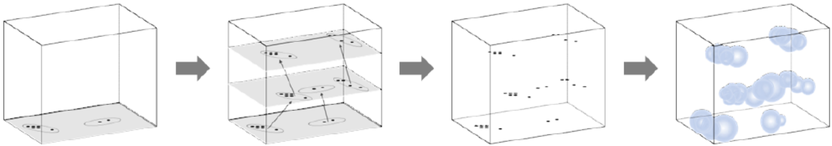


Figure : Modeling germs with Markov chain of Matérn cluster processes and spherical grains with random radii

[SHT^{+}11]

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Main challenge: Germ-grain model

Question: What kind of grains should we model?

- Overlapping sphere system
  - Poisson process
  - Matérn cluster process
  - Gibbs process

  } — limited structure — fitting unobserved germs
- Quermass-interaction model — tricky parameter estimation

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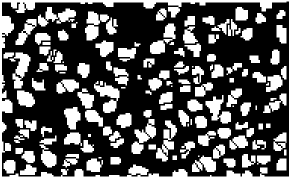
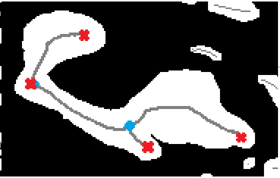
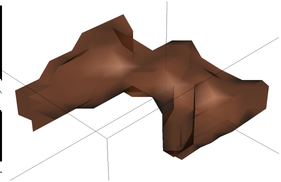
- System of overlapping compact but **more complex sets**
  + better control over pore shape
  + germs as midpoints of Watershed objects
  — unclear how to capture pore connectivity

More complex sets as grains

Possible candidates

- Watershed
- skeleton endpoints
- Fourier transform
- tensors
- set of basic shapes
- ...

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Summary

- **Task**: 3D model of porous micro-structure
  - pore shape and connectivity VERY important
- **Step I**: Extraction of marked point pattern
  - overlapping sphere system
  - system of overlapping more complex sets
- **Step II**: Connectivity in z-direction
  - continuum percolation
  - Markov chain
- **Question**: Use none of the above and base model on, e.g., a dilated skeleton?

Important references

Dereudre, D., Lavancier, F., and Staňková Helisová, K.
Estimation of the intensity parameter of the germ-grain quermass-interaction model when the number of germs is not observed.
Scandinavian Journal of Statistics, 41:239–246, 2014.

Kaunisto, Erik, Marucci, M., Borgquist, P., and Axelsson, A.
Mechanistic modelling of drug release from polymer-coated and swelling and dissolving polymer matrix systems.
International Journal of Pharmaceutics, 418:54–77, 2011.

Stenzel, O., Hassfeld, H., Thiedmann, R., Koster, L.J.A., Oosterhout, S.D., van Bavel, S.S., Wienk, M.M., Loos, J., Janssen, R.A.J., and Schmidt, V.
Spatial modeling of the 3d morphology of hybrid polymer-zno solar cells, based on electron tomography data.
The Annals of Applied Statistics, 5(3):1920–1947, 2011.

Thank you for your attention and let us have a nice discussion!