Supporting information of

"Quantifying local heterogeneities in the 3D morphology of X-PVMPT battery electrodes based on FIB-SEM measurements"

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Details of image segmentation

For image segmentation, i.e., for classifying the voxels as pores or solid, the following features are used: Gaussian smoothing, Laplacian of Gaussian, Gaussian gradient magnitude, difference of Gaussians, structure tensor eigenvalues, and Hessian of Gaussian eigenvalues, each one for $\sigma \in \{0.3, 0.7, 1.0, 1.6, 3.5, 5.0, 10.0\}$. The output of the trained random forests is a probability map, indicating the probability whether a voxel belongs to the pore space or to the solid phase. Each voxel that belongs to the solid phase with probability greater than 0.4 is assigned to solid phase and otherwise to background, leads to a segmentation into the solid phase and its complement. Note that this threshold has been manually determined based on visual inspection, where we used the open-source software Fiji.

Computation of morphological descriptors

In the following, we describe the computation of morphological descriptors considered for structure characterization by statistical image analysis. For a given sampling window, the thickness of the electrode δ is computed as follows. For each line of voxels, which is orthogonal to the aluminum foil, the corresponding thickness is defined as the maximum distance between two voxels on this line belonging to the solid phase of the electrode. The average of these distances over all lines contained in the sampling window is considered to be the thickness of the electrode in this sampling window. The porosity ε is determined by the point-count method,[S1] i.e., the porosity is the ratio of pore voxels over all voxels of the electrode within the sampling window. The surface area per unit volume S is computed as the surface area between solid and pores in the considered sampling window divided by the volume of the sampling window. For the computation of surface areas from voxelized image data, we use the algorithm proposed in Ohser and Schladitz.[S2] Finally, we consider the mean geodesic tortuosity which is a purely geometrical descriptor quantifying the windedness of shortest transportation paths through a given phase. To compute the mean geodesic tortuosity of a phase in a given sampling window, the shortest pathways through this phase are determined by applying the Dijkstra algorithm[S3] on the voxel grid. When computing mean geodesic tortuosity on local cutouts as sampling windows, the starting points of the paths are located within these local cutouts, while the paths themselves are allowed to leave the sampling window in order to avoid a strong influence of edge effects.

Univariate probability density functions

The probability density functions $f : \mathbb{R} \to [0, \infty)$ of the parametric distributions used for modeling the univariate distributions of local morphological descriptors are provided in the following.

1. Mixture of two Beta distributions $\mathsf{Beta}(\alpha_1, \alpha_2, \beta_1, \beta_2, p_B)$

$$f\left(x\right) = p_B \; \frac{\Gamma\left(\alpha_1 + \beta_1\right) x^{\alpha_1 - 1} \left(1 - x\right)^{\beta_1 - 1}}{\Gamma\left(\alpha_1\right) \Gamma\left(\beta_1\right)} + \left(1 - p_B\right) \; \frac{\Gamma\left(\alpha_2 + \beta_2\right) x^{\alpha_2 - 1} \left(1 - x\right)^{\beta_2 - 1}}{\Gamma\left(\alpha_2\right) \Gamma\left(\beta_2\right)},$$

   for each $x \in [0, 1]$, where $\alpha_1, \alpha_2, \beta_1, \beta_2 > 0$ and $0 \leq p_B \leq 1$.

2. Mixture of two Gaussian distributions $\mathsf{N}(\mu_1, \mu_2, \sigma_1, \sigma_2, p_N)$

$$f\left(x\right) = p_N \; \frac{1}{\sigma_1 \sqrt{2\pi}} \exp\left(\frac{\left(x - \mu_1\right)^2}{2\sigma_1^2}\right) + \left(1 - p_N\right) \; \frac{1}{\sigma_2 \sqrt{2\pi}} \exp\left(\frac{\left(x - \mu_2\right)^2}{2\sigma_2^2}\right)$$

   for each $x \in \mathbb{R}$, where $\mu_1, \mu_2 \in \mathbb{R}, \sigma_1, \sigma_2 > 0$ and $0 < p_N < 1$.

3. Maxwell-Boltzmann distribution $\mathsf{MB}(\mu_M, \sigma_M)$

$$f\left(x\right) = \sqrt{\frac{2}{\pi}} \frac{\left(x - \mu_M\right)^2}{\sigma_M^2} \exp\left(-\frac{\left(x - \mu_M\right)^2}{2\sigma_M^2}\right),$$

   for each $x > \mu_M$, where $\mu_M \in \mathbb{R}$ and $\sigma_M > 0$.

4. Shifted Gamma distribution $\Gamma(a_\Gamma, \sigma_\Gamma, \mu_\Gamma)$

$$f\left(x\right) = \frac{1}{\Gamma\left(a_\Gamma\right)} \left(\frac{x - \mu_\Gamma}{\sigma_\Gamma}\right)^{a_\Gamma - 1} \exp\left(-\frac{x - \mu_\Gamma}{\sigma_\Gamma}\right),$$

   for each $x > \mu_\Gamma$, where $a_\Gamma, \sigma_\Gamma > 0$ and $\mu_\Gamma \in \mathbb{R}$.

5. Rayleigh distribution $\mathsf{R}(\mu_R, \sigma_R)$

$$f\left(x\right) = \frac{x - \mu_R}{\sigma_R} \exp\left(-\frac{\left(x - \mu_R\right)^2}{2\sigma_R^2}\right),$$

   for each $x > \mu_R$, where $\mu_R \in \mathbb{R}$ and $\sigma_R > 0$.

Brief introduction to copulas

We briefly recall the concept of copulas. Let (U, V) be a two-dimeensional random vector taking values in the unit square $[0,1]^2$, where U and V are uniformly distributed on the unit interval $[0,1]$. Then, the joint probability distribution function $C : [0,1]^2 \to [0,1]$ of (U, V) is called a two-dimensional copula, where $C(u,v) = P(U \leq u, V \leq v)$ with $P(U \leq u) = u$ and $P(V \leq v) = v$ for any $u, v \in [0,1]$. From Sklar's representation formula[S4] we get that for any two-dimensional random vector (X, Y), its joint probability distribution function $H : \mathbb{R}^2 \to [0,1]$ with $H(x,y) = P(X \leq x, Y \leq y)$ can be written in the form

$$H(x,y) = C\left(F\left(x\right), G\left(y\right)\right)$$

for all $x, y \in \mathbb{R}$, where $F : \mathbb{R} \to [0,1]$ with $F\left(x\right) = P\left(X \leq x\right)$ for each $x \in \mathbb{R}$ and $G : \mathbb{R} \to [0,1]$ with $G\left(y\right) = P\left(Y \leq y\right)$ for each $y \in \mathbb{R}$ are the univariate distribution functions of X and Y, respectively, and $C : [0,1]^2 \to [0,1]$ is a certain copula. Moreover, if the functions F, G and C are differentiable, then the joint probability density $h : \mathbb{R}^2 \to [0,\infty)$ of (X, Y) can be written as

$$h(x,y) = f(x)g(y)\left(\frac{\partial^2}{\partial x \partial y}C\right)(F(x), G(y))$$

for all $x, y \in \mathbb{R}$, where $f : \mathbb{R} \to [0,\infty)$ and $g : \mathbb{R} \to [0,\infty)$ are the univariate probability densities of X and Y, respectively. From the joint probability density, we directly obtain the conditional probability density $h_{Y=y} : \mathbb{R} \to [0,\infty)$ of X given $Y = y$ for each $y \in \mathbb{R}$ fulfilling $g(y) > 0$. It reads as

$$h_{Y=y}(x) = f(x)\left(\frac{\partial^2}{\partial x \partial y}C\right)(F(x), G(y))$$

for each $x \in \mathbb{R}$.

References

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