

## Finite Element Simulation of the Performance of a Structural Electrolyte

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**Summary.** This contribution concerns the multi-scale and multi-physics finite element analysis of structural power composites, i.e. multifunctional composites with simultaneous load bearing and energy storing functionality. We are particularly interested in obtaining the effective macro-scale properties of the structural electrolyte by employing computational homogenization to capture the effects of micro-heterogeneities on the sub-scale. The sub-scale problem is defined by a statistical volume element that is numerically generated, and the effective properties are obtained by conducting virtual material testing on the synthetic microstructure.

*Keywords:* structural electrolyte, artificial microstructure, statistical volume element, computational homogenization, virtual material testing

### Structural power composites

Carbon fibers are not only widely used as structural reinforcement materials; they are also attractive for use in electrode components due to their excellent intercalation properties. Combining and utilizing both of its functionalities, strength and ionic storage capacity, at once results in new multifunctional materials called structural power composites, such as the structural battery and structural supercapacitor that have simultaneous load bearing and energy storing functionality [1, 2].

If two different needs are addressed with a multifunctional material instead of two separate subsystems, then significant weight and volume savings can be achieved [2]. In short, this innovation enhances the system performance in various applications, and addresses the demand for more efficient and sustainable systems. However, due to the infancy of this technology, more studies need to be conducted.

In this contribution, we are particularly interested in the structural electrolyte, which is a crucial component for all structural power composites. The structural electrolyte's function is to be electrically insulating, but ionically conductive while being able to carry mechanical loads. The considered structural electrolyte consists of a highly porous solid polymer matrix, where the pores are interconnected and form a complex channel system that contains liquid electrolyte. Since both the solid phase and the pore space are continuous, the structural electrolyte forms a bicontinuous system. The aim of this contribution is to study the ionic conductivity and the stiffness of the structural electrolyte with finite element analysis.

## Two-scale modeling by computational homogenization

In order to include the effects of micro-heterogeneities in the structural electrolyte microstructure, we employ computational homogenization [3] to separate the sub-scale and the macro-scale. Doing this allows us to compute the effective properties for the macro-scale via homogenization of the sub-scale problem. The sub-scale problem is defined by a Statistical Volume Element (SVE) that characterizes the material heterogeneity. The SVE is given by an artificial microstructure that is numerically generated in a fashion that enables us to use periodic boundary conditions. Different approaches to the artificial microstructure generation are used depending on the desired morphology of the structural electrolyte microstructure, see Figure 1.

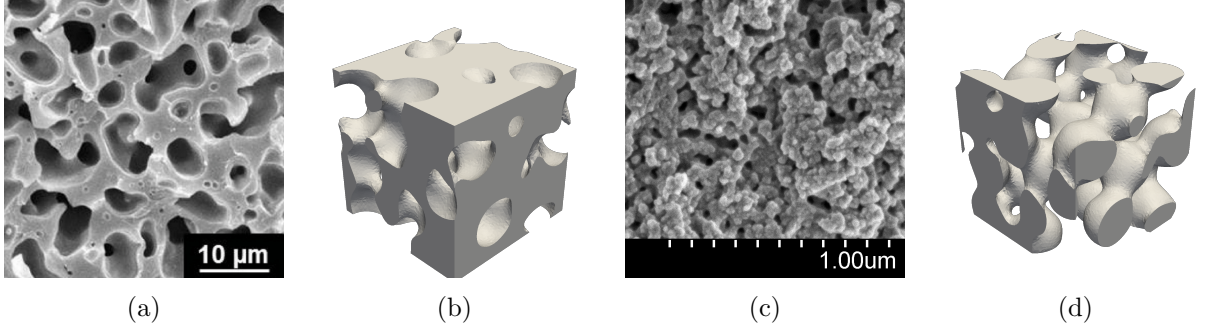


Figure 1: SEM scans of the solid polymer matrix and corresponding artificial structures. (a) SEM scan 50MTM57/2.3M\_1PC [4], and (b) its artificial counterpart. (c) SEM scan AB/0.65 [5]<sup>1</sup>, and (d) its artificial counterpart.

Since both the mechanical properties and the electro-chemical conductivity are of interest, we solve the linear elasticity and linear diffusion problem on the sub-scale. Here, we assume that there is no mechanical resistance from the liquid electrolyte in the pore channels when the structural electrolyte is loaded mechanically, i.e. the structural electrolyte is considered to be a drained system. Furthermore, we assume that diffusion cannot take place in the solid polymer matrix, and that the diffusion problem is stationary and decoupled from the mechanical problem.

The strong form of the sub-scale linear elasticity and diffusion problem in the SVE domain  $\Omega_{\square}$  is given as

$$-\boldsymbol{\sigma} \cdot \boldsymbol{\nabla} = \mathbf{0} \text{ in } \Omega_{\square}, \quad (1)$$

$$\boldsymbol{\sigma} = \mathbf{E} : \boldsymbol{\epsilon}, \quad (2)$$

$$\boldsymbol{\epsilon} := (\mathbf{u} \otimes \boldsymbol{\nabla})^{\text{sym}}, \quad (3)$$

$$\boldsymbol{\nabla} \cdot \mathbf{j} = 0 \text{ in } \Omega_{\square}, \quad (4)$$

$$\mathbf{j} = -\mathbf{D} \cdot \boldsymbol{\nabla} \mu, \quad (5)$$

where  $\boldsymbol{\sigma}$  is the sub-scale stress,  $\mathbf{u}$  is the displacement field,  $\boldsymbol{\epsilon}$  is the linear symmetric sub-scale strain and  $\mathbf{E}$  is the sub-scale isotropic stiffness tensor for the linear elasticity problem. For the diffusion problem  $\mathbf{j}$  is the sub-scale ion mass flux,  $\mu$  is the sub-scale electro-chemical potential and  $\mathbf{D}$  is the isotropic sub-scale conductivity tensor which is, in the simplest case of isotropy, defined as  $\mathbf{D} = DI$ .

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While formulating the corresponding weak forms is straightforward, the scale separation needs to be addressed properly. The prolongation rule employs an additive split of the displacement field and the electro-chemical potential field

$$\mathbf{u}(\mathbf{x}) = \mathbf{u}^M(\mathbf{x}) + \mathbf{u}^s(\mathbf{x}), \quad \mathbf{x} \in \Omega_\square, \quad (6)$$

$$\mu(\mathbf{x}) = \mu^M(\mathbf{x}) + \mu^s(\mathbf{x}), \quad \mathbf{x} \in \Omega_\square. \quad (7)$$

Here,  $\mathbf{u}^M(\mathbf{x})$  and  $\mu^M(\mathbf{x})$  are the smooth displacement and electro-chemical potential fields, while  $\mathbf{u}^s(\mathbf{x})$  and  $\mu^s(\mathbf{x})$  are the sub-scale displacement and electro-chemical potential fluctuation fields. The smooth fields are prescribed according to the assumption of first order homogenization as

$$\mathbf{u}^M(\mathbf{x}) = \bar{\boldsymbol{\epsilon}} \cdot [\mathbf{x} - \bar{\mathbf{x}}], \quad \mathbf{x} \in \Omega_\square, \quad (8)$$

$$\mu^M(\mathbf{x}) = \nabla \bar{\mu} \cdot [\mathbf{x} - \bar{\mathbf{x}}], \quad \mathbf{x} \in \Omega_\square, \quad (9)$$

where the imposed macro-scale strain  $\bar{\boldsymbol{\epsilon}}$  and electro-chemical potential gradient  $\nabla \bar{\mu}$  serve as the driving forces for respective problem. Rigid body motion on the macro-scale does not affect the mechanical response, and a constant macro-scale electro-chemical potential  $\bar{\mu}$  is omitted since it does not change the diffusion process in the stationary case. The reference point  $\bar{\mathbf{x}}$  is chosen arbitrarily, but a common choice is the SVE centre.

The next step is to specify the boundary conditions. The strongly periodic boundary conditions (SPBC) are mainly defined by the micro-periodicity assumption. Micro-periodicity assumes that the sub-scale fluctuation fields  $\mathbf{u}^s$  and  $\mu^s$  are periodic on the boundary of the SVE. Before defining the micro-periodicity assumption formally, it is first necessary to introduce the boundary split  $\Gamma_\square = \Gamma_\square^+ \cup \Gamma_\square^-$ , where  $\Gamma_\square^+$  is the image boundary ("positive" side) and  $\Gamma_\square^-$  is the mirror boundary ("negative" side). The consequence of introducing such a boundary split is that every point on a boundary gets a partner point on the opposite side. The next step is to introduce the periodic mapping operator  $\varphi_{\text{per}} : \Gamma_\square^+ \mapsto \Gamma_\square^-$  such that  $\mathbf{x}^- = \varphi_{\text{per}}(\mathbf{x}^+)$ . Finally, the micro-periodicity of the displacement and ion concentration fluctuation field can be expressed as

$$\mathbf{u}^s(\mathbf{x}) = \mathbf{u}^s(\varphi_{\text{per}}(\mathbf{x})), \quad \forall \mathbf{x} \in \Gamma_\square^+, \quad (10)$$

$$\mu^s(\mathbf{x}) = \mu^s(\varphi_{\text{per}}(\mathbf{x})), \quad \forall \mathbf{x} \in \Gamma_\square^+. \quad (11)$$

Moreover, the symmetry condition of the sub-scale stress and ion mass flux are defined as

$$\boldsymbol{\sigma}(\mathbf{x}) \cdot \mathbf{n} = \boldsymbol{\sigma}(\varphi_{\text{per}}(\mathbf{x})) \cdot \mathbf{n}, \quad \forall \mathbf{x} \in \Gamma_\square^+, \quad (12)$$

$$\mathbf{j}(\mathbf{x}) \cdot \mathbf{n} = \mathbf{j}(\varphi_{\text{per}}(\mathbf{x})) \cdot \mathbf{n}, \quad \forall \mathbf{x} \in \Gamma_\square^+, \quad (13)$$

where  $\mathbf{n}$  denotes the outward-pointing normal on  $\Gamma_\square^+$ . Hence, the surface fluxes  $\mathbf{t} := \boldsymbol{\sigma} \cdot \mathbf{n}$  and  $\mathbf{j} := \mathbf{j} \cdot \mathbf{n}$  are anti-periodic.

Although the SPBC are defined here, they are implemented in a weak sense. Nevertheless, all of this gives a solvable sub-scale problem. Lastly, we introduce the homogenization of the sub-scale quantities which is defined by the SVE volume average operator

$$\langle \bullet \rangle_\square := \frac{1}{|\Omega_\square|} \int_{\Omega_\square} \bullet \, d\Omega, \quad (14)$$

which gives the macro-scale stress and ion mass flux in a post-processing step as

$$\bar{\sigma} = \langle \sigma \rangle_{\square}, \quad (15)$$

$$\bar{j} = \langle j \rangle_{\square}. \quad (16)$$

Since the linear elasticity problem is solved in a strain-controlled setting, and the macro-scale ion concentration gradient is prescribed in the linear diffusion problem, it becomes a trivial task to compute the corresponding effective properties based on the macro-scale constitutive equations.

### Preliminary numerical results

The effective Young's modulus  $\bar{E}$  (normalized) and the effective diffusion coefficient  $\bar{D}$  (normalized) are computed for a wide range of porosities  $\phi$  and structures, see Figure 2. Note that the different structures only exist in certain porosity ranges since each structural electrolyte must be a bicontinuous system.

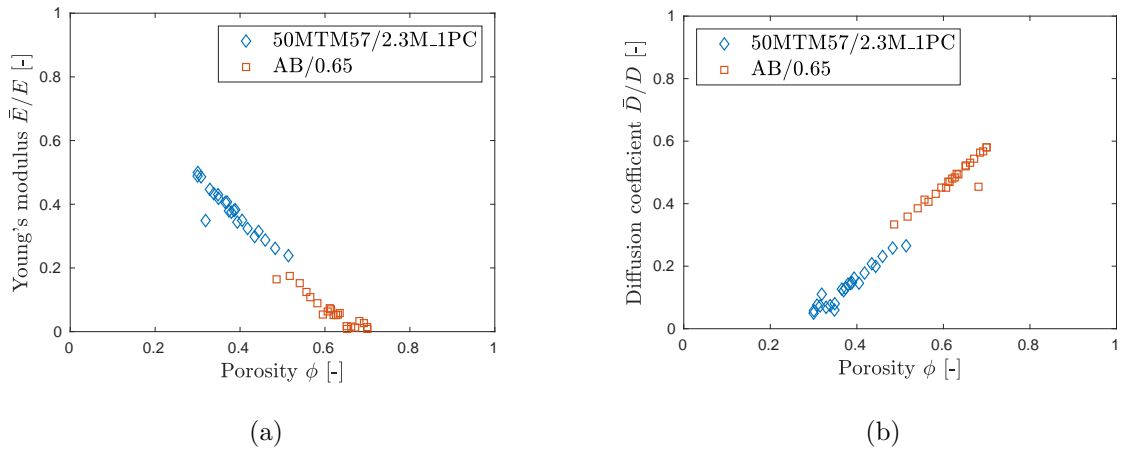


Figure 2: Effective properties for different microstructures and their relevant ranges of porosity. (a) Effective Young's modulus (normalized). (b) Effective diffusion coefficient (normalized).

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