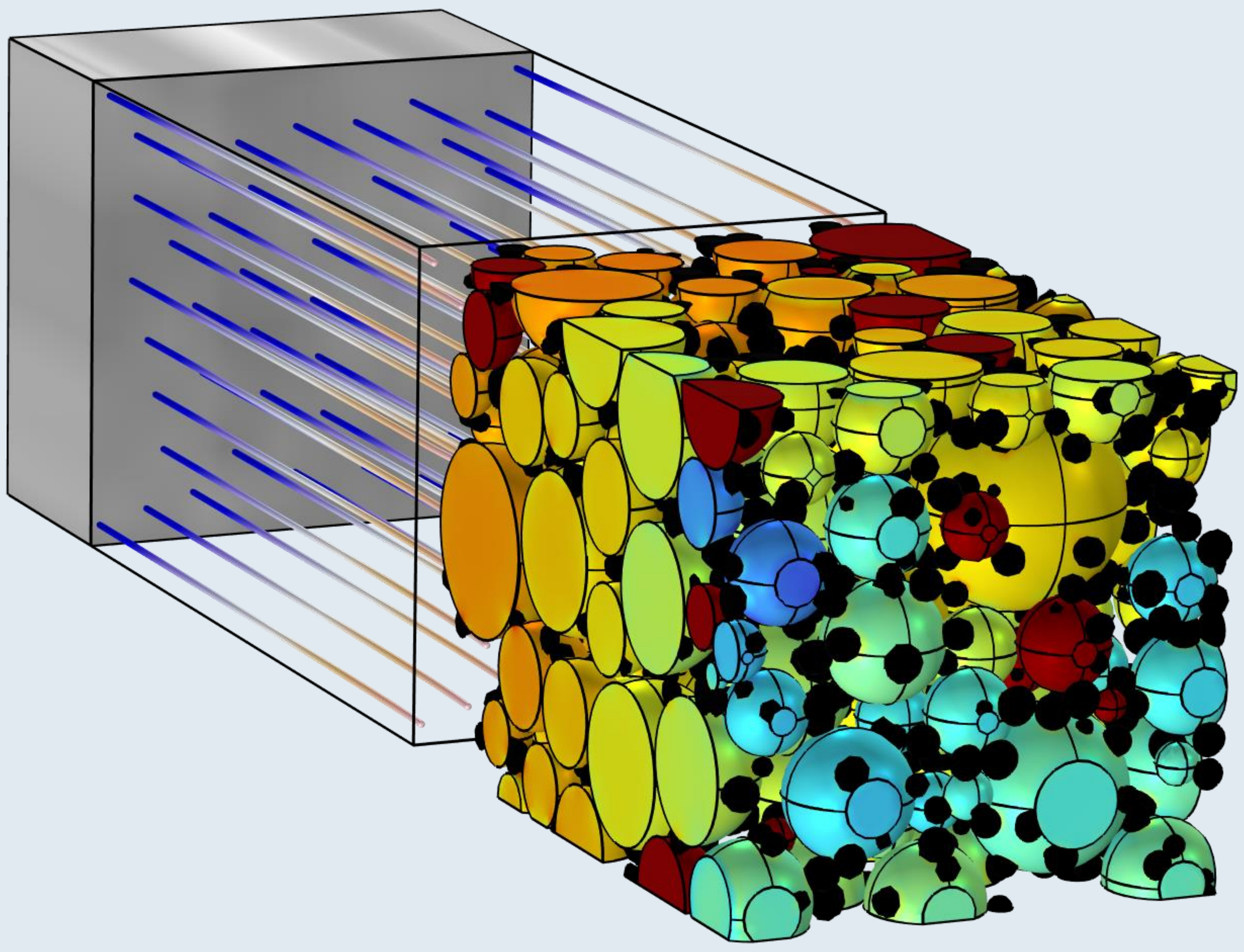

Electrochemomechanical Simulations of 3D-Resolved Solid-State Lithium-Ion Battery Cells

Comparison between a novel microstructure model for solid-state battery, coupling electrochemistry and mechanics, and a standard simplified setup.

G. Blanco[1], M. Alberghini[1], A. Bertinetti[1], A. Tommasi[1]

1. Gemmate Technologies, Buttigliera Alta, Italy

Introduction

A novel model for particle-based electrodes was compared with a standard setup [1-2] by measuring their tortuosity, electrical conductivity and electrochemical transport, with the aim of understanding the impact of microstructure heterogeneities and the involved physical phenomena on the cell performance. The cathode was modelled as a mixture of NMC811, carbon-based additive SuperP, and argyrodite sulfide (Li_6PS_5Cl) as solid electrolyte, mimicking a real-world solid-state cell architecture.

In particular, mechanical stresses were coupled with electrochemistry, as they affect the equilibrium potential and introduce a convective term in the Li transport equation. Balancing accuracy and simplicity, the proposed approach aims to provide a more accurate model of battery cathodes, resulting in a more physical behavior that prevents to overestimate the battery performance.

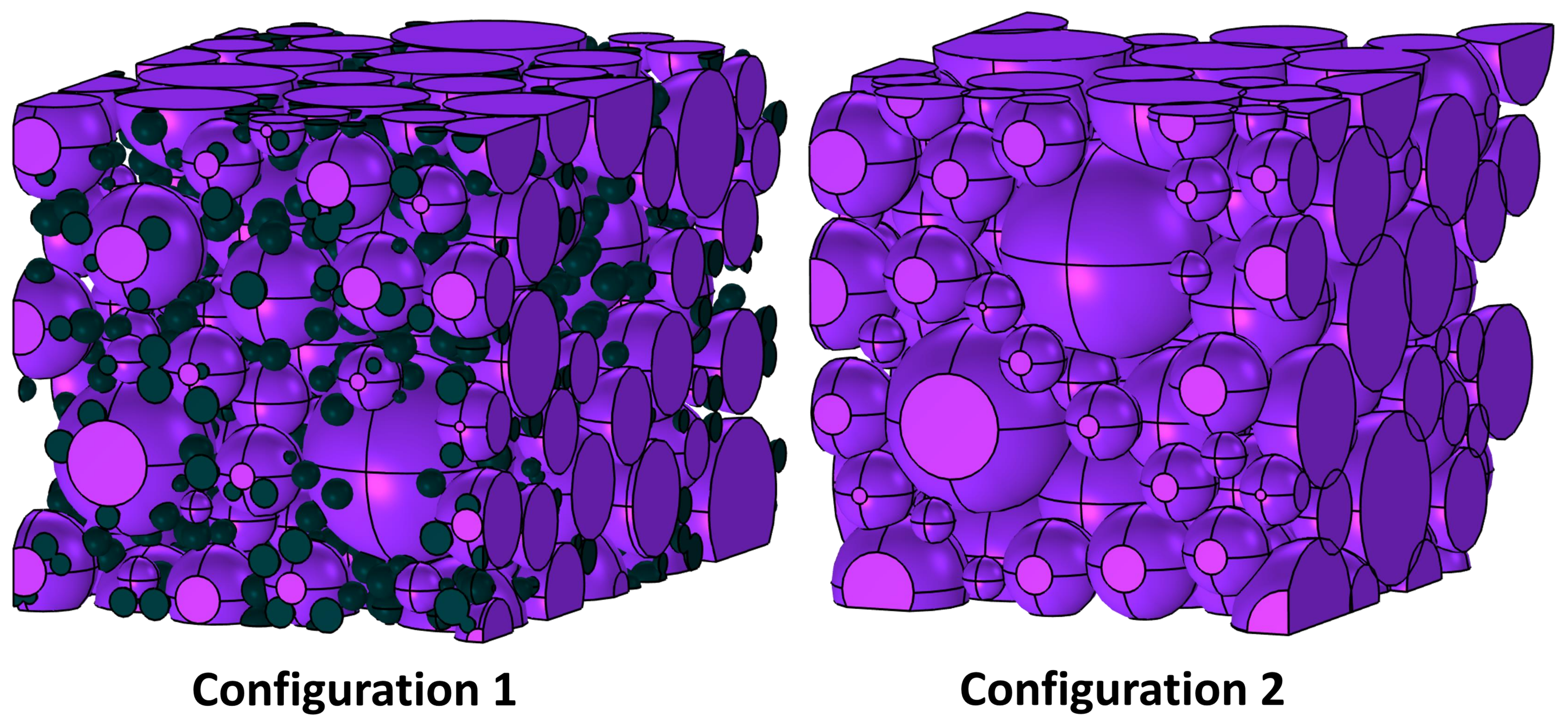

FIGURE 1. Left: Novel microstructure configuration. Right: Standard microstructure configuration.

Methodology

The proposed model (Conf.1) avoids contact between NMC particles (purple spheres) and explicitly includes SuperP aggregates (black spheres); the standard setup (Conf.2) consists of overlapped NMC particles, while SuperP is homogenously dispersed in the electrolyte.

In Conf.1 electrochemistry and mechanics were coupled: the transport of lithium within the NMC, with concentration c, is influenced both by its diffusion coefficient D and the gradient of the hydrostatic stress σ_h, generated by de/intercalation.

$$\frac{\partial c}{\partial t} + \nabla \cdot \left(-D\nabla c + \frac{D\Omega}{RT} \nabla \sigma_h c \right) = 0$$

Results

The inclusion of carbon-based particles results in a lower electrical conductivity of the cathode compared to the standard setup, in agreement with experimental results [3]. This decrease the amount of stored and released energy in the cell.

Incorporating mechanically-driven Li transport within the active material particles (Fig.2, left) results in a more homogeneous Li distribution respect to conventional model (Fig.2, right), with a consequential significant reduction in the cell overpotential. It also leads to a higher and more homogeneous distribution of the stress within the NMC particles.

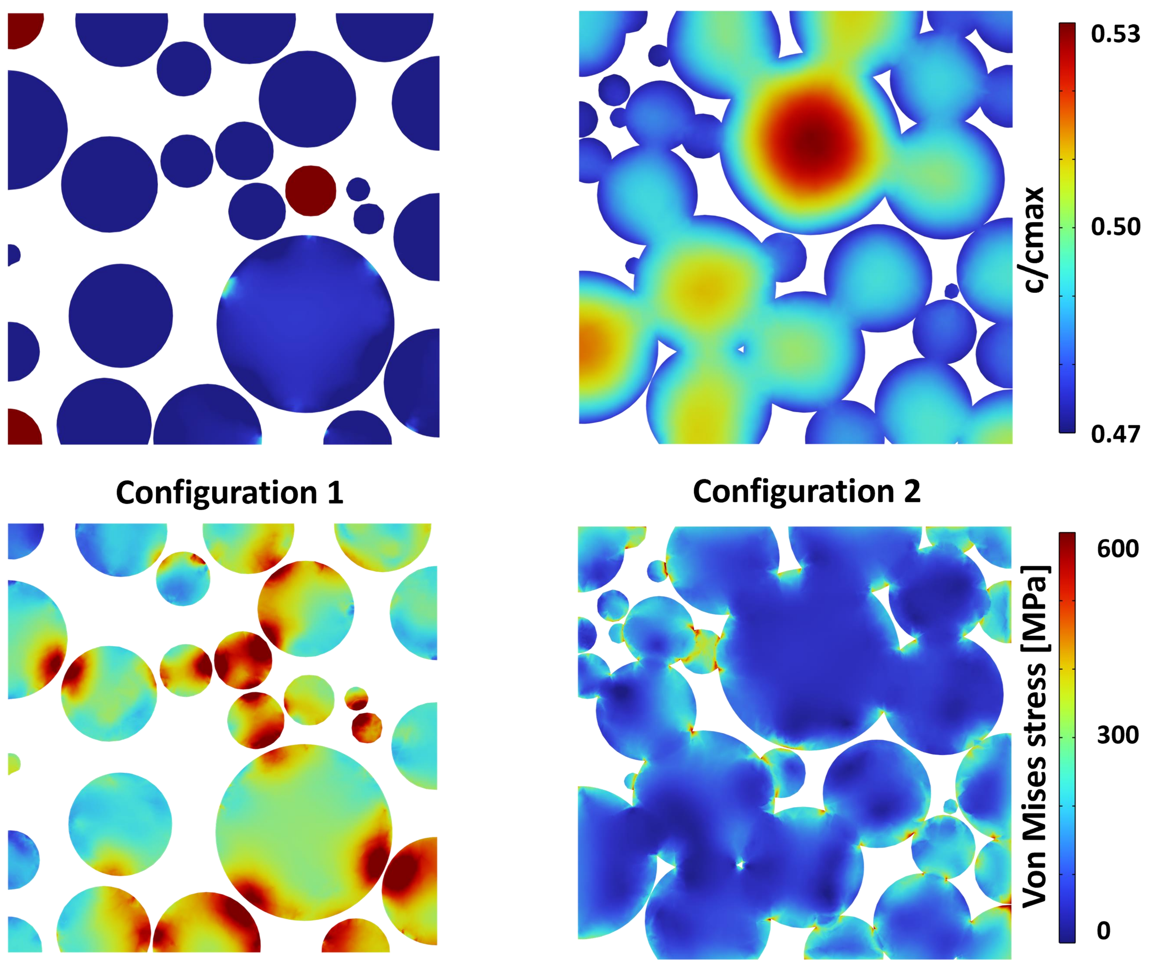

FIGURE 2. Comparison of the relative lithium concentration within the active particles of the two microstructures at C/20.

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