A new approach for calculation of ion diffusion characteristics based on topological analysis of procrystal electron density

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Understanding the mechanisms of ion migration at the atomic level is crucial for developing high-performance electrochemical devices. First-principles simulations are powerful tools for calculating ion diffusion characteristics, but they are computationally intensive and cannot handle complex structures, such as interfaces. To overcome these limitations, we present a new method based on topological analysis of procrystal electron density. This approach enables a more efficient exploration of ion migration pathways. We demonstrate the effectiveness of the methodology through three case studies: Li-ion transport across Li/Li_6PS_6Cl solid electrolyte interface and grain boundaries in Li_3ClO electrolyte, and computation of diffusion coefficients in spinel $LiTiS_2$ electrode material. The presented approach is not limited by these specific cases and can be applied for calculating ion diffusion characteristics in various crystalline ion conductors.

Phase field simulation of Li dendrite growth

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In our quest to advance rechargeable lithium metal electrodes for broader applications, we have developed a sophisticated grand potential-based non-linear phase-field model. Utilizing the open-source software package MOOSE, this model meticulously studies the electrodeposition of lithium, pertinent to lithium metal anodes, under varying conditions of overpotential and different operating temperatures. Through meticulous two-dimensional simulations, we observed dynamic morphological evolutions that alternately reveal dendrite growth or stable deposition patterns, contingent upon the applied overpotential magnitude and varying operating temperature. This comprehensive study elucidates the temporal and spatial distributions of overpotential, highlighting the interplay between ion transport and electrochemical reactions. These interactions culminate in diverse dendritic growth patterns, fundamentally influenced by the operating conditions.

P2D model of lithium-ion battery with solid-state electrolyte

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To predict and study the electrochemical performances of a solid-state cell a pseudo-two-dimensional (P2D) battery model was developed considering single-ion conducting electrolyte. The P2D model considers two separated but connected one-dimensional domains: the first one is constituted by the thickness of the cell components, and the second one represents the active material particle domain. In the first domain charge and mass transport equations are solved for the solid and electrolyte phases and the Butler-Volmer equation is used to simulate the electrochemical reactions. In the active material particle, the conservation of lithium is described by Fick's law and the equation is solved along the radial dimension. The two domains are coupled based on the law of conservation of species.

Considering the solid-state cell, in contrast with the classical P2D model, the concentration of lithium-ion in the electrolyte is assumed to be constant and diffusion and convection processes within the electrolyte phase are ignored [1]. Therefore, in the solid electrolyte only charge transport is considered and modelled using Ohm's Law:

$$i_l = -\sigma_l \nabla \phi_l$$

i_l is the current density in the solid electrolyte [A/m^2]
σ_l is the conductivity of the solid electrolyte [S/m]
ϕ_l is the potential in the solid electrolyte [V]

Furthermore, in this model the lithium electrode is considered as an infinite reservoir of lithium supply without any dimension; therefore, it is modelled as a boundary [2].

[1] Wolff N. et al., "Model based assessment of performance of Lithium-Ion batteries using single ion conducting electrolytes", *Electrochimica Acta*, **2018**, vol. 284, pp. 639
[2] Sahapatsombut U. et al., "Modelling the micro-macro homogeneous cycling behaviour of a lithium-air battery", *Journal of Power Sources*, vol. 227, **2013**, pp. 143

P4D modelling approach to predict BoL and EoL behaviour of lithium-ion battery with solid-state electrolyte

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The P4D model is realized as an approach in which the mathematical equations describing the underlying physical phenomena are solved in the domains with their real dimensions. This feature distinguishes this approach from the P2D model, developed by POLITO, which simplifies the geometry of the cell and represents the cell in two dimensions. In P4D, however, the equations are formulated in four dimensions: cell height, cell width, cell thickness, and the dimension related to the spherical particle of the active material. Accordingly, similar to the P2D model, four sub-domains can be identified in the P4D model: the metallic lithium as the anode, the argyrodite as the solid electrolyte, NMC622 as the cathode and aluminium as the current collector. While the electrolyte, cathode and current collector domains are modelled as cylinders, the lithium anode is implemented as a boundary.

While the parameters obtained and used in the P2D model were used in the P4D to reproduce BoL, the P4D model requires further parametrization for EoL. For this purpose, the specific surface area of the electrode (α_v) is parameterized. As α_v gradually decreases during cycling, the contact loss between the active material and the solid electrolyte increases, as often reported in the literature. The increasing contact loss during cycling leads to limited reaction kinetics and therefore a higher overpotential is achieved. As a result, the capacity retention during cycling is reduced.