**Physics-based modeling opportunities to overcome critical challenges in lithium-ion batteries**

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**1.Introduction**

The commercialization of electric vehicles powered by lithium-ion batteries is one of the main strategies towards a future society based on renewable energy systems. However, the effective and sustainable use of lithium-ion batteries still faces several critical challenges.

One of the desirable requirements is the capability of the automotive battery to be charged in less than 15 min. Such a difficulty comes mostly from the microscopic electrochemical phenomena occurring at the anode, namely the sluggish intercalation of lithium within a phase-separating material like graphite, leading to degradation phenomena such as lithium plating. Understanding the interplay among phase separation, plating and other parasitic phenomena can stimulate significant advancements in the field.

A second related challenge stems from the possibility to early detect such degradation phenomena during operation. This would allow the battery management system to actively control the charge/discharge of single cells, prolonging their cycle life and reduce risk concerns. At the moment only semi-empirical indicators of battery state of health are used in the battery management system.

Another critical aspect regarding the sustainable use of lithium-ion batteries refers to their recyclability. The European Union is setting up a new regulation, pushing towards an increased content of recycled materials in new lithium-ion batteries. Although achieving such a target will likely reduce the environmental impact associated to battery production from raw materials, it is still unclear whether a recycled battery material, with potentially reduced intrinsic properties, can be effectively used for demanding automotive applications.

In this study we show how all these critical challenges can be quantitatively addressed by adopting a physics-based microscopic modeling approach to lithium-ion battery research, taking into account the interplay among different electrochemical phenomena across multiple length scales.

**2. Methods**

The modelling approach is based on porous electrode theory, which describes a single-repeating unit of a battery cell by means of mass and charge conservation equations in a pseudo-2D fashion (Figure 1). Along the through-thickness coordinate the model solves for the charge and species transport in the liquid electrolyte as well as for the conservation of electrons in the electron-conducting phase. A second dimension is added to solve for the transport of intercalated lithium across the radius of active material particles. At the negative electrode side, both intercalation and Li plating kinetics are enforced at the interface between graphite particles and the electrolyte. Details of the model derivation can be found in previous dedicated papers [1–4].

**3. Results and discussion**

Figure 2 summarizes the main results of the application of the modeling framework. Figure 2a shows the graphite state of lithiation across the electrode thickness and the particle radius upon fast charge as predicted by the model (solid and dashed lines) and as obtained by optical experiments. The comparison shows that the model correctly captures the phase transitions of graphite during lithiation, as denoted by the different colors, indicating that a significant plating risk is present when the graphite surface is saturated by intercalated lithium.



**Figure 1.** Physics-based modeling framework for the simulation of lithium-ion batteries.

Figure 2b reports the comparison of simulated (line) and experimental (symbol) discharge curves after a current pulse. During relaxation the model predicts the system dynamics, enabling the identification of the characteristic time of each microscopic process: intercalation kinetics takes place at short time scales (< 1 min), the relaxation of electrolyte concentration gradient occurs at medium time scales (ca. 5 min) while solid-state diffusion within active material particles occurs at long time scales (> 30 min). Thus, based on the shift of the characteristic time of the process, the model can efficiently detect any early signal of degradation.

Finally, Figure 2c shows practical indications about the compensatory measures of electrode design when recycled cathode active materials are used. Compared to the electrode thickness and particle diameter of a reference battery cell, a decrease in gravimetric capacity of the recycled cathode materials can be compensated by a small increase in electrode thickness while a reduction in solid-state diffusivity can be mitigated by a reduction in particle diameter. All these modifications indicate that compensatory measures are technically feasible, thus opening for a large share of recycled materials in the manufacturing of lithium-ion-batteries.



**Figure 2.** Potential of physics-based model in predicting a) intercalation and degradation phenomena during fast charge, b) time scale of processes for early detection of degradation, c) guidelines for recycled materials.

**4. Conclusions**

This study shows how physics-based electrochemical modelling of coupled transport and reaction phenomena in lithium-ion batteries has the potential of guiding the research towards viable solutions to the critical challenges of this technology. In particular, fast charge protocols, algorithms for early detection of degradation and guidelines for recycled battery designs can be effectively explored by using this approach.

**References**

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