Multiscale Modelling of Li-Ion Battery Volume Expansion

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Abstract—Mechanical characteristics of Li-ion batteries play an important part in the design of electric devices or energy storage systems. Proper mechanical design can increase the safety and the longevity of devices. Apart from external forces, Li-ion batteries also experience internal volume changes due to inherent physical processes, especially volume changes caused by intercalation and deintercalation of lithium ions into electrode materials. Currently, these changes are mainly studied experimentally using complex methods, while numerical simulations of these processes are surprisingly rare. This works presents different numerical approaches for studying internal volume changes in Li-ion batteries. The results show that it is possible to study volume changes at different length scales, but it is necessary to apply a suitable simplification of the model to obtain macroscopic deformation.

Keywords—Li-ion battery, numerical simulation, volume expansion, intercalation, electrochemistry

1. INTRODUCTION

In recent years, mechanical behavior of Li-Ion batteries became an interesting area due to their importance in the design and longevity of energy storage systems. The structural integrity and damage resistance are important factors in the transportation industry and especially for emerging electrical vehicles. One specific property of Li-ion battery is its sensitivity to external and internal mechanical loads. These loads can lead to internal short circuits, which could result in thermal runaway and rapid dissipation of energy [1, 2].

Furthermore, volume changes are directly related to inherent electrochemical processes. These changes can be caused by lithium migration, in which electrode materials change their volume due to lithium intercalation and deintercalation. This process seems to be the most significant one as graphite can expand up to 13%, while common cathode materials up to 5 % [3]. Additionally, effects such as gas formation due to side reactions or lithium plating can lead to further volume changes. Both reversible and irreversible volume changes increase with the degradation of the battery cell. As a result, accurate prediction of volume changes is necessary to ensure safety and longevity of batteries designed for long term use [4]. Experimental investigation of these changes is often difficult, as the maximum deformation is usually in tenths of millimeters. To estimate the internal deformation of the layers, it is necessary to use techniques such as micro-CT or destructive methods such as dissembling the battery [1]. The disadvantage of experimental methods is that they require a lot of time and usually they can be carried out only on a smaller scale.

On the other hand, numerical simulations can provide a suitable alternative to the experiments. Currently, most research on internal volume changes is carried out on the microscopic scale [5-7]. While this approach is the most accurate, as it directly describes the electrochemical nature of the process, its complexity makes it nearly impossible to apply it on the macroscopic scale. Currently, most of the macroscopic models focus on the effects of external forces, while the internal volume changes remain suppressed [8].

The presented work shows a possibility of numerical modelling of internal volume changes in Li-ion batteries caused by elastic volume change caused by intercalation and deintercalation at different length (particles, layers, and whole battery) scales. The simulations show that a simplification of the model based on the geometry is necessary to obtain precise results in appropriate time.

2. NUMERICAL MODEL

As it is possible to study volume changes of Li-ion batteries from different perspective, it is necessary to use appropriate models for each case based on the geometry scale. The simulations presented in this work focused on a description of volume changes at different scales (particles, electrode layers and the whole battery). Electrochemical and mechanical properties of an 18650 NCA battery were used for simulations [2, 3, 9]. The volume changes were caused only by lithiation and delithiation as it seemed to be the most important mechanism. For all simulations, it was assumed that the anode volume was increased by 10%, while the cathode volume was decreased by 5% [1]. This process represented a discharge of the battery. The structural simulations were carried out in Ansys Mechanical software, while the electrochemical simulation was carried out in Ansys Fluent.

Microscopic simulation used a numerical model based on the models presented in [5] and [6], which describe in detail the transport and intercalation of lithium ions into the cathode/anode host materials. The transport of lithium inside the particles is only governed by diffusion, while the transport of lithium ions in the electrolyte is derived from a concentrated solution theory, which considers both solute-solvent and solute-solute interaction. The charge transfer chemistry at the solid-electrolyte interface is described through a Butler-Volmer equation. In this model, the volume expansion can be directly related to lithium concentration in a particle, as the initial and maximum concentrations are known parameters [6]. The simplified geometry is shown in Fig. 1. The radius of anode and cathode particles was set to 1 μm.

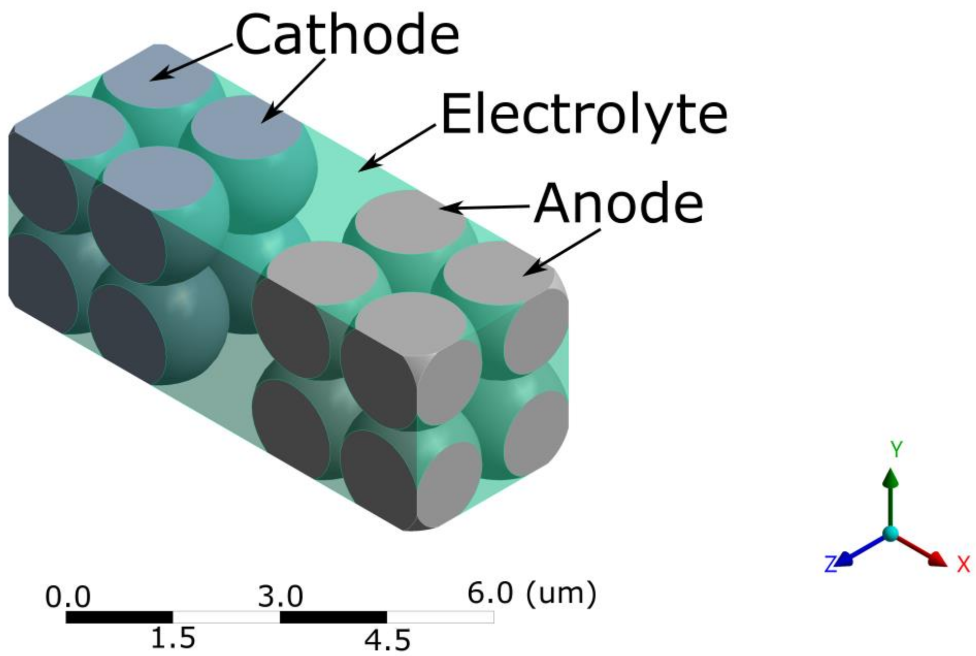

Figure 1: Simplified microstructure of a Li-Ion battery. Each particle has a radius of 1 μm

Simulations which focused on large scale volume changes were described by a simplified approach. Even a commonly used physics-based Newman's P2D model [10] does not readily holds information about lithium concentration in the electrodes. In these models the anode, the cathode and the electrolyte are usually considered as single joint zone, so it would not be possible to define individual volume changes for different materials. For this reason, it was not necessary to resolve whole electrochemical model. Instead, only estimated volume changes were set for the anode (+10%) and the cathode (-5%) materials. The simulation was carried out in Ansys Mechanical software, as the problem became purely mechanical. The thickness of layers was defined as: anode – 80 μm, aluminium foil – 30 μm, cathode – 55 μm and copper foil – 30 μm. The thickness of steel casing was set to 250 μm. The height of the jelly roll was set to 60 mm. Mechanical properties of materials were obtained from [2]. The geometry for the whole battery simulation consisted of a steel casing with a width of 18 mm and a height of 65 mm. Inside the casing, there was a 60 mm tall domain, which represented the jelly roll structure. A similar approach to the method shown in [11] was used to obtain average material properties of the simplified continuum domain.

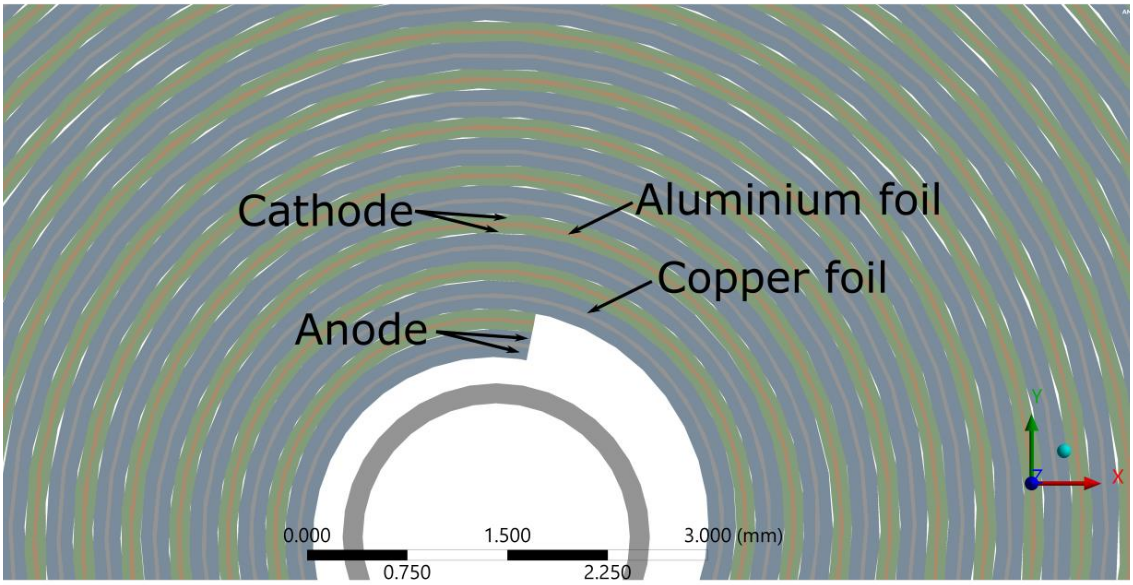

Figure 2: Detail of the jelly roll layer structure with visible individual layers.

3. RESULTS

Results from microscopic simulation show that it is possible to track lithium concentration inside the electrode particles. As lithium ions migrate from the anode, they are intercalated into the cathode structure, which is shown by the increased lithium concentration near the particle surface. Lithium ions inside the particles are then transported by a diffusion mechanism. In this case the volume changes are not very significant. This is caused by the fact that the actual active layer of the electrode consists of many particles, and the overall volume change is the sum of individual contributions. In order to increase the particle volume by 10%, its radius has to be only increased approximately 1.032 times. This means that the original radius of 1 μm would be 1.032 μm at the end of the simulation, so the change is not very noticeable.

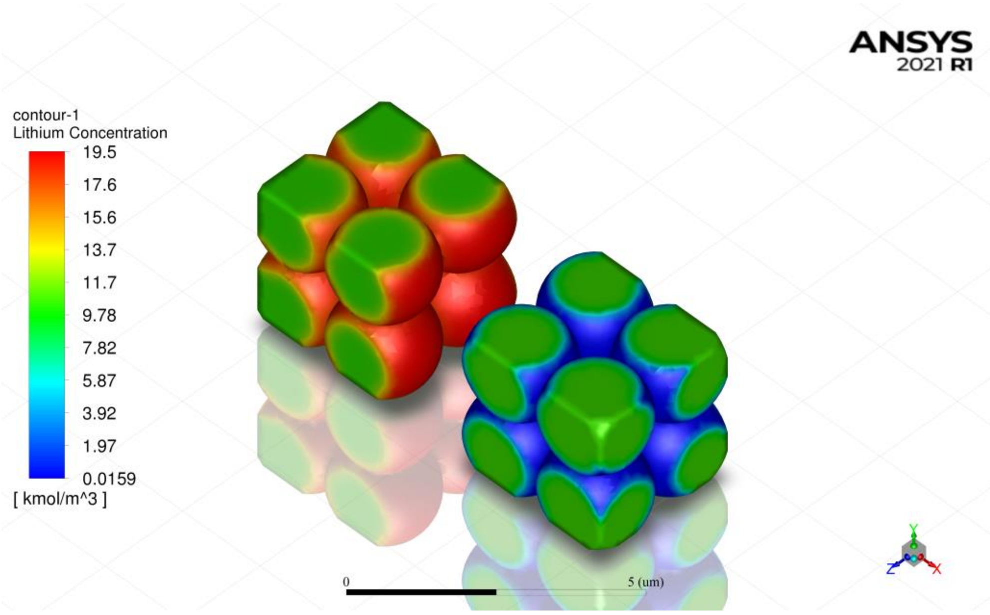

Figure 3: Lithium concentration inside microscopic structure of Li-Ion battery.

Fig. 4 shows the macroscopic deformation of the jelly roll structure. In this case, the volume change was set to individual cathode and anode layers. The maximum deformation was visible at the end of the cathode layer (top of the geometry) and the top left area, which was empty. The average deformation of the steel casing was approximately 0.18 mm. Generally, the inner layers experienced more strain as they had more free space to expand. This effect was also reported in the work of Willenberg et al. [1].

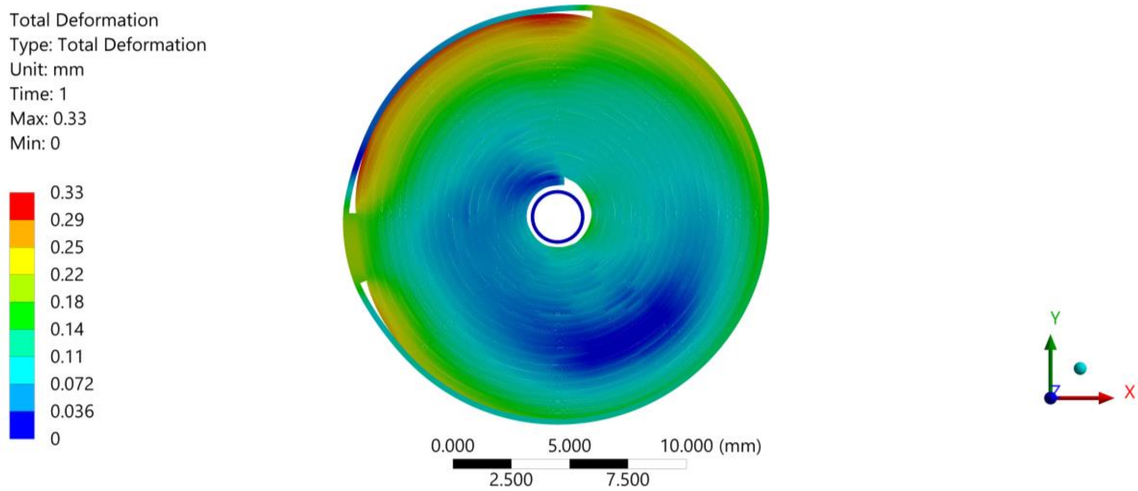

Figure 4: Deformation of a jelly roll structure with resolved individual layers.

Even the resolution of individual layers may be too detailed for the study of multiple batteries in battery packs. Fig. 5 shows the result of volume changes in the whole battery. Even though, the jelly roll structure was considered as a continuum with averaged values, the results generally agree with the previous simulation with resolved layers. The average outward deformation of the steel casing was approximately 0.17 mm. The main difference, in contrast to the previous simulation, is that in this case, the inner layers experienced significant deformation.

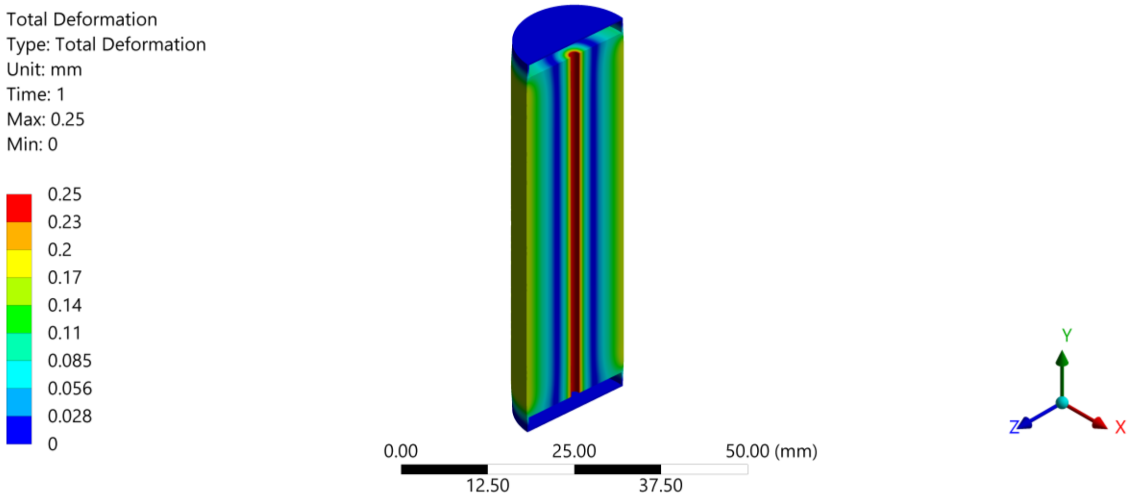

Figure 5: Deformed geometry of an 18650 NCA battery.

4. CONCLUSION

Presented simulations show that it is possible to study volume changes of Li-ion batteries at different length scales. However, it is necessary to choose an appropriate model for the desired application. While the simulation at the microscopic scale should be the most precise, the volume changes can be studied only at this scale. To study macroscopic effects by this method, it would require resolving complex and large geometries, which would be extremely computationally expensive. On the other hand, simplified methods show good accuracy while being relatively quick. The resolution of individual layers is an effective method to study processes inside individual batteries. To study volume changes of batteries in battery packs, defining of individual layers may become too complicated, so a connection of layers into a single zone may seem to be the best option. Generally, these simulations can help with identifying critical areas in the design of batteries or even battery packs, which might lead to a mechanical failure. These models can be also extended to include the effects of thermal expansion, Joule heating, inelastic volume changes or gasification, which would increase their accuracy.

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REFERENCES

[1] L. K. Willenberg, P. Dechent, G. Fuchs, D. U. Sauer, and E. Figgemeier, “High-Precision Monitoring of Volume Change of Commercial Lithium-Ion Batteries by Using Strain Gauges”, *Sustainability*, vol. 12, no. 2, 2020, doi 10.3390/su12020557.

[2] C. Zhang, J. Xu, L. Cao, Z. Wu, and S. Santhanagopalan, “Constitutive behavior and progressive mechanical failure of electrodes in lithium-ion batteries”, *Journal of Power Sources*, vol. 357, pp. 126-137, 2017, doi 10.1016/j.jpowsour.2017.04.103.

[3] S. Schweidler, L. de Biasi, A. Schiele, P. Hartmann, T. Brezesinski, and J. Janek, “Volume Changes of Graphite Anodes Revisited: A Combined Operando X-ray Diffraction and In Situ Pressure Analysis Study”, *The Journal of Physical Chemistry C*, vol. 122, no. 16, pp. 8829-8835, Apr. 2018, doi 10.1021/acs.jpcc.8b01873.

[4] C. R. Birkl, M. R. Roberts, E. McTurk, P. G. Bruce, and D. A. Howey, “Degradation diagnostics for lithium ion cells”, *Journal of Power Sources*, vol. 341, pp. 373-386, 2017, doi 10.1016/j.jpowsour.2016.12.011.

[5] A. M. Colclasure and R. J. Kee, “Thermodynamically consistent modeling of elementary electrochemistry in lithium-ion batteries”, *Electrochimica Acta*, vol. 55, no. 28, pp. 8960-8973, 2010, doi 10.1016/j.electacta.2010.08.018.

[6] G. M. Goldin, A. M. Colclasure, A. H. Wiedemann, and R. J. Kee, “Three-dimensional particle-resolved models of Li-ion batteries to assist the evaluation of empirical parameters in one-dimensional models”, *Electrochimica Acta*, vol. 64, pp. 118-129, 2012, doi 10.1016/j.electacta.2011.12.119.

[7] R. Xu, Y. Yang, F. Yin, P. Liu, P. Cloetens, Y. Liu, F. Lin, and K. Zhao, “Heterogeneous damage in Li-ion batteries: Experimental analysis and theoretical modeling”, *Journal of the Mechanics and Physics of Solids*, vol. 129, pp. 160-183, 2019, doi 10.1016/j.jmps.2019.05.003.

[8] L. Tang, J. Zhang, P. Cheng, and X. Hu, “Homogenized modeling methodology for 18650 lithium-ion battery module under large deformation”, *PLOS ONE*, vol. 12, no. 7, Jul. 2017, doi 10.1371/journal.pone.0181882.

[9] C. M. Julien and A. Mauger, “NCA, NCM811, and the Route to Ni-Richer Lithium-Ion Batteries”, *Energies*, vol. 13, no. 23, 2020, doi 10.3390/en13236363.

[10] K. Uddin, S. Perera, W. Widanage, L. Somerville, and J. Marco, “Characterising Lithium-Ion Battery Degradation through the Identification and Tracking of Electrochemical Battery Model Parameters”, *Batteries*, vol. 2, no. 2, 2016, doi 10.3390/batteries202001.3

[11] P. Vyroubal and T. Kazda, “Equivalent circuit model parameters extraction for lithium ion batteries using electrochemical impedance spectroscopy”, *Journal of Energy Storage*, vol. 15, pp. 23-31, 2018, doi 10.1016/j.est.2017.10.019.