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Modelling the calendering process of lithium-ion battery electrodes using the Discrete Element Method (DEM)

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Abstract. The electrochemical performance of a lithium-ion battery is strongly influenced by the microstructure of its electrode. A dried electrode consists of the active material (AM) particles and carbon-binder domain (CBD) phase. One of the important steps in electrode manufacturing is 'calendering', where a dried electrode is compressed between heated rollers to obtain a mechanically stable and uniform structure. The effect of calendering pressure on the porosity, tortuosity, and coordination number of an electrode is studied well using DEM. However, a thorough mathematical understanding of the compaction behavior of Li-ion battery electrodes remains largely unexplored.

In the present work, we aim to understand the compression behaviour of an electrode using DEM simulations. The simulation domain consists of spherical particles of varying sizes, representing the AM particles. The initial positions, shapes and sizes of the AM are obtained experimentally via XCT [1]. The domain is periodic in the lateral direction, with a moving top wall and a fixed bottom wall. Particle interactions are modelled using Edinburg elasto-plastic adhesive (EEPA) and bond-model, where the bond model captures the mechanical response of CBD phase. Simulations are performed on Altair EDEM.

It is shown that porosity and tortuosity obtained from the simulation data are well within the range of experimental values. The pressure-compression behaviour of the simulated structure closely aligns with the powder compaction behaviour described by the Kawakita equation.

1 Introduction

Lithium-ion batteries (LiBs) are widely used in electric vehicles (EVs), portable electronics, military equipment and medical devices due to their higher energy density and low cost [2]. Despite its widespread usage, there is still potential to enhance energy density, charging speed, and lifespan while reducing production costs. In LiBs, the electrode is where the insertion and extraction of lithium ions occur while charging and discharging. The complex microstructure of the electrode significantly influences electronic and ionic conductivity, ultimately affecting the battery's overall performance [3].

The electrode's microstructure is primarily influenced by its manufacturing process. It consists of active material (AM) and carbon additives, which are bound together using a binder. The electrode fabrication process includes mixing, slurry formation, coating, drying, and calendering. Calendering, a crucial step, involves compressing the dried electrode between heated rollers to create a mechanically stable and uniform structure. While this process decreases porosity to improve conductivity, it also increases tortuosity, which reduces ionic conductivity. Therefore, selecting an optimal calendering pressure that balances the trade-off between porosity and tortuosity is required to achieve efficient electrochemical performance of LiBs.

Discrete element method (DEM) simulations are commonly used for particle-level analysis of electrodes during calendering, which is difficult to do experimentally. To date several authors have incorporated DEM to model the calendering process. The inter-particle contacts and connectivity of the active material during calendering are analysed using fabric tensors [4]. Authors have reported the discrepancy between the results obtained through simulation and experiments. This could potentially be attributed to the absence of interparticle cohesive forces, the initial configuration of particles being different from the experiments, or the failure to account for the plastic deformation of AM particles in the simulation. In another study, it was shown that the elastic contact model (Hertz model) under high compression overestimates the real behaviour of electrodes [5], that can be accurately modelled using elasto-plastic models. Later, Ge et. al [6] used the Edinburg elasto-plastic adhesive (EEPA) with the bond model to simulate the calendering process. Here, the initial arrangement of active material was obtained from the X-ray tomography data of a real electrode. Many other recent studies are based on the similar contact model, as it can closely predict the experimental response of an electrode during calendering. A recent experimental study examined

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the deformation behaviour of electrodes during calendering [7]. Their findings suggest that the calendering process can be analysed as a powder compaction process. They also investigate the influence of roller temperature and diameter on the compression and elongation. However, this study did not specifically examine the in-situ behavior of the electrode during calendering.

In this study, we specifically employed the Discrete Element Method (DEM) to model the electrode calendering process. Our aim is to investigate the compaction behaviour of the electrode by analysing particle-level data obtained from the DEM simulations. In particular, we aim to develop a mathematical equation that can accurately predict the pressure–compression behaviour of electrodes during calendering and hence explain the particle behaviour.

2 Methodology

2.1 Simulation domain

The volume and position of the active material extracted from the XCT images of an uncalendered electrode are used to generate initial particle configuration for the simulation. The active material (AM) particles used in experiments are nearly spherical; therefore, for computational simplicity, they are approximated as spheres of the same volume in the simulations. The particle size distribution (PSD) of the AM particles used in the simulation is shown to be in good agreement with the experimental distribution (Fig. 1).

Simulation domain comprises of periodic box in the lateral direction (X and Y axes), with a top moving and fixed bottom wall (Fig. 2). The top wall, made of steel, mimics the calendering roller and the bottom wall, made of aluminium, models the current collector. The top wall is compressed at the fixed velocity of -0.1 m/s for 0.25×10^{-3} s in the Z-direction.

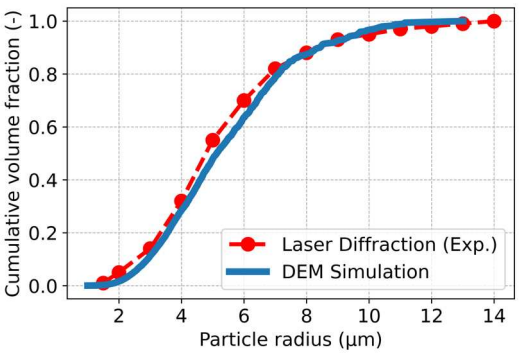

Figure 1. Comparison of the size distribution of AM particles obtained from experiment with the DEM simulation.

2.2 Discrete Element Method

Translational and rotational velocities of AM particles within the electrode are obtained by the time-integration of the following equations:

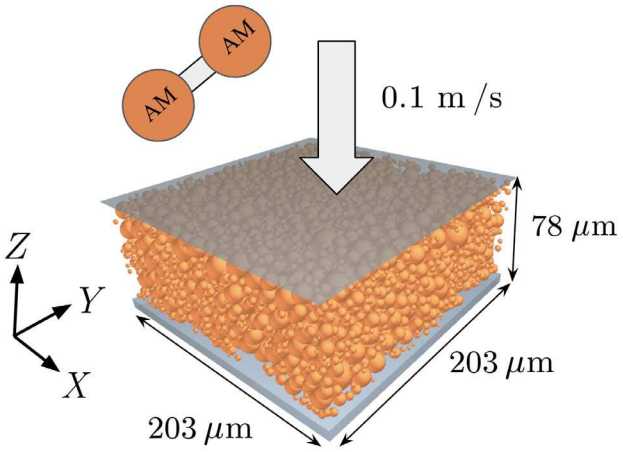

Figure 2. Schematic of the simulation domain used for the DEM simulation.

$$m\frac{d\vec{v}}{dt} = \vec{F}_c + \vec{F}_b \tag{1}$$

$$I\frac{d\vec{\omega}}{dt} = \vec{M}_c + \vec{M}_b \tag{2}$$

where m, I, $\vec{v}$ and $\vec{\omega}$ are the mass, moment of inertia, linear and angular velocity of any particle, respectively. $\vec{F}_c$ and $\vec{M}_c$ are the contact force and contact torque on a particle while it is in contact with any other particle or wall. Where as, $\vec{F}_b$ and $\vec{M}_b$ are the force and torque on a particle due to the existence of interparicle rigid bond. The interparticle and wall-particle contact force is determined using the EEPA model, as discussed in [6]. Interparticle rigid bonds were generated between AM, when interparticle distance, $\|\vec{r}_{ij}\| \leq 1.67(R_i + R_j)$. R_A and R_B are the radius of the interacting particles. Material properties and other simulation parameters taken from our previous work are listed in Table 1.

Table 1. Material properties of the AM particle used in the DEM simulation.

Particle Property	Value
Young's Modulus, Pa	1.4×10^{11}
Poisson's Ratio	0.25
Coefficient of Restitution	0.5
Static Friction	0.25
Rolling Friction	0.01
Density, kg/m^3	4750

2.3 Morphological analysis

Porosity and tortuosity of the calendered and uncalendered structure from the DEM simulation are compared with their experimental values. The estimation of electrode porosity is non-trivial due to presence of particle overlap under high compression. Porosity of the electrode is determined using the Monto-Carlo method, where random

points are generated within the domain to count the number of points that fall on the particles. Porosity is obtained such that $\epsilon = 1 - (N_{mc,particle}/N_{mc,total})$, where $N_{mc,total}$ is the total number of generated points and $N_{mc,particle}$ is the count of points falling on the particles.

Tortuosity (τ) is an important factor that determines the transport property across a porous structure. Geometrically it is the ratio of actual path to the straight path along the direction of flow in the porous media. For an electrode, tortuosity represents how easily a lithium ion can move through the electrolyte phase. In the performed simulation, void between the AM particles are meshed to solve the heat conduction equation. The ratio of total flux at the outlet to the theoretical straight flux (same porosity) is obtained using the battery analysis tool of Altair EDEM. Tortuosity and porosity are computed using the beta version of the battery analysis tool, which is planned for release in a future version of Altair EDEM. The results have been verified to be independent of mesh size. Other microstructural parameters, such as the coordination number and fabric tensors, can be derived from the particle data obtained through the DEM simulation; however, difficult to obtain experimentally. In this paper, since our objective is to demonstrate the validation, only porosity and tortuosity are calculated from the DEM simulations.

3 Results

3.1 Porosity and Tortuosity of Electrodes

To validate the simulation results against experimental data, the porosity obtained from the simulation is compared with its experimental values from [1]. Here, porosity represents the interstitial void volume fraction including pores and CBD phase between AM particles. The comparison is made for two different compression pressures: 0 Pa and 200 MPa. These pressures correspond to the force exerted on the top plate in the direction of compression. As shown in Fig. 3, porosity obtained from the DEM simulation is in good agreement with the experimental data for compressed structure. However, a slight difference in porosity is observed for the uncompressed electrode. This may be attributed to the approximation of AM particles as spheres in the DEM simulation. The simplification could lead to an underestimation of particle volume in the simulation results. Overall, results suggest that electrode porosity reduces by almost 20% on compressing electrode by 200 MPa.

Next, the tortuosity of both uncalendered and calendered electrode obtained from the DEM simulation is compared with experimental data (Fig. 4). Results show that the tortuosity of the calendered electrode is around 15% higher than that of the uncalendered electrode. A similar trend is observed in both simulation and experimental analysis. This behavior is expected, as compression reduces porosity and increases the coordination number, causing lithium ions to follow a longer transport path. The tortuosity values obtained from the simulation are lower compared to those from the experiments. This slight difference between the results could be because of the simplification of particle shapes in the simulation, as experimentally AM particles are generally non-uniform and non-spherical shapes. Additionally, the breakage of AM particles at higher pressure is not modeled within the existing DEM simulation.

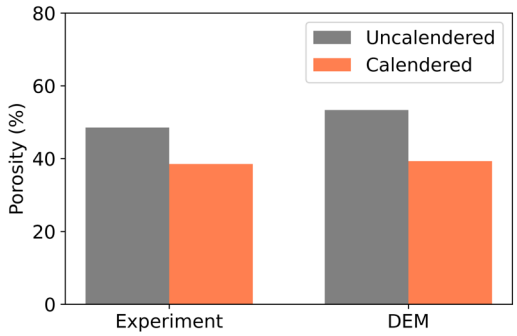

Figure 3. Comparison of the porosity obtained from the experiments and DEM simulation

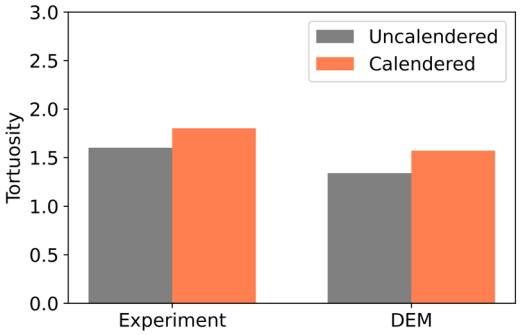

Figure 4. Comparison of the tortuosity obtained from the experiments and DEM simulation

3.2 Compaction behaviour of the electrode

Next, the compaction behaviour of the electrode is analysed by plotting the percentage change in electrode thickness against the applied pressure (Fig. 5). Electrode thickness is determined as the difference between the maximum and minimum z-coordinate of the particles. In DEM simulations, few particles may move beyond the simulation domain, and potentially leading to an overestimation of the electrode thickness. To address this, outlier particles that significantly deviate from the overall height distribution are excluded from the calculation. The percentage change is calculated relative to the thickness of the uncalendered electrode and referred to as the compression (C) of the electrode. The pressure (P) is determined as the force applied per unit area on the moving top wall.

Here, the compaction behaviour of electrode is modelled using the the Kawakita equation (Fig. 5), such that

$$C = \frac{aP}{b + P} \tag{3}$$

The Kawakita equation [8] is an empirical formula proposed for the compaction of pharmaceutical powders. Since, LiB electrode consists of granular particles (AM and carbon additives) and the calendering process closely resembles powder compaction, the Kawakita equation is expected to effectively model the electrode calendering. In Equation 3, constant a is the maximum compression at infinite pressure, where porosity of the electrode tends to zero and electrode becomes dense.

In Fig. 5 electrode compression is plotted against the pressure of the top wall, obtained from the DEM simulation. The compression plot is fitted with Kawakita equation and it is determined that fitting behaviour can be accurately represented using Kawakita equation with constants a = 29.07, b = 21.35 and R^2 = 0.99.

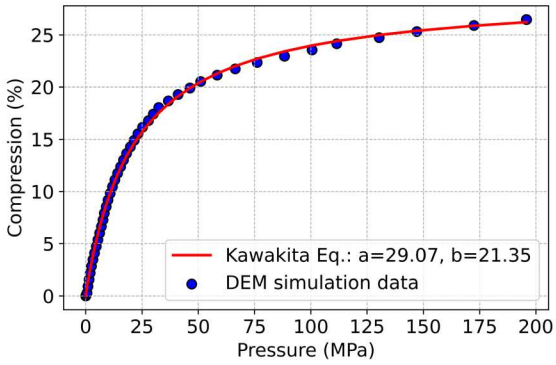

Figure 5. Pressure-compression plot obtained from the DEM simulation and its fitting with Equation 3

4 Conclusion and Future prospect

This paper investigates the calendering process of LiB electrodes through DEM simulation. The size and position of the active material (AM) are obtained from XCT data of an uncalendered electrode. The shape of AM particles in the simulation is approximated as a sphere. Interparticle and wall-particle interactions are modelled using the EEPA-Bond model. The porosity and tortuosity of both uncalendered and calendered electrode is calculated through simulations and compared with the experimental data. Following are the main conclusion of the presented paper -

1. Porosity of calendered structure is more than that of uncalendered structure. The values are well within the range of experimentally obtained porosity values.

2. The tortuosity of the electrode increases upon calendering; a similar behaviour has been reported in experiments.

3. The Pressure-compression behaviour of the simulated electrode is shown to resemble the powder compaction behaviour using the Kawakita equation.

Future work will examine the applicability of the powder compaction equation at higher compaction levels. Additionally, understanding the combined effects of roller temperature and compaction on the electrode microstructure, which will be accomplished through coupled thermal-DEM simulations.

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