Modelling the thermal and mechanical properties of electrodes under calendering process based on discrete element simulation

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ABSTRACT

In this study, we thoroughly investigated the impact of the calendering process on the microstructure of lithium-ion battery electrodes and explored the related fracture behavior and heat transfer characteristics. Through numerical simulations and experimental validation, we systematically analyzed the effects of different calendering parameters on the contact force and heat transfer efficiency between particles within the electrode. The results indicate that by appropriately adjusting the calendering parameters, we can optimize the electrode microstructure, increase the particle compactness, and thereby enhance the electrode's stability and mechanical integrity. These in-depth insights provide a substantial theoretical foundation for the manufacturing and design of lithium-ion batteries, potentially offering strong guidance for the development and improvement of future battery technologies.

Keywords: Discrete element method, lithium-ion battery electrodes, calendering

1. INTRODUCTION

With the increasing popularity of lithium-ion batteries, there is still room for improvement in their performance and durability. In the production of lithium-ion batteries, calender pressing is an essential step used to press the electrodes, Calender pressing can make the contact between particles within the electrode more uniform and enhance the contact force between the electrode and the current collector. It also helps to reduce the electrode thickness under the same active material loading, thereby improving the volumetric energy density.

Due to the significant expenditure on resources and raw materials required for experimental research, numerical simulation of battery electrodes using the Discrete Element Method (DEM) is highly regarded. The DEM method, first developed by Cundall and Strack, is based on a discrete numerical system that simulates the motion of particles by individually calendering the interactions and contacts between them[1]. Giménez proposed a simulation program and an appropriate DEM contact model to reproduce the microstructure of the electrode, and conducted a simple mechanical analysis of the anode using nanoindentation experiments[2]. Sangrós used the DEM method to model electrode particles, studying the impact of changes in microstructural parameters such as porosity, particle size, and electrode thickness on thermal conductivity[3]. Junpeng Zhang revealed the impact of anisotropy in electrode particle contact forces on the anisotropy of heat transfer within porous electrodes. He predicted the thermal conductivity of the electrode using porosity and quantitatively analyzed the heat transfer mechanisms during the electrode calendering process[4]. This paper aims to elucidate the effects of the calendering process on the mechanical Properties and heat transfer characteristics of electrode particles.

2. NUMERICAL SIMULATION

2.1 Discrete Element Method

The particles in the electrode are interconnected through a bonder, and they undergo elastic-plastic deformation during the calendering process. The Discrete Element Method (DEM), developed by Cundall and Strack, represents the interactions between discrete points in space using prescribed contact formulas[1].

In DEM simulations, two different contact relationships of particles should be modeled: particle-particle contact, referred to as particle contact, and particle-bonder contact, termed bonder contact. The contact model in DEM shows that when two particles or agglomerates are pressed together, they undergo elastic and plastic deformation, and the binder force increases with the increase in plastic contact area[5].

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The resultant force and moment acting at each bonder contact are composed of forces and moments generated from particle-particle overlapping and carried by the bonder. The formula is as follows:

$$\mathbf{F}_c = \mathbf{F}^l + \mathbf{F}^d + \overline{\mathbf{F}} \tag{1}$$

$$M_c = \overline{M} \tag{2}$$

In the equation, $\mathbf{F}^l$ represents linear force, $\mathbf{F}^d$ denotes damping force, $\overline{\mathbf{F}}$ stands for binder force, and $\overline{M}$ represents binder moment.

2.2 The generation of electrode particles

An electrode can be considered a porous, multilayer composite material. Mechanical actions and thermal conduction occur within the coating and between the coating and the current collector. The established industrial method to increase the energy density of lithium-ion batteries is calendering the electrode.

When creating the microstructure of the battery, spherical particles are first randomly filled in the rectangular area, and the size distribution and properties of the particles are set according to the parameters in Table 1[2] (the microstructure is as shown in Figure 1).Due to computational limitations, it is currently too costly to simulate a complete electrode at the microscopic scale. Therefore, the boundaries in the X and Y directions are periodic to reduce the size of the simulation domain. The representative volume element (RVE) of 150 × 150 × hA μm³ is sufficient to ensure effective and representative results while maintaining a reasonable computation time.

Table 1. Physical Properties of Spherical Particles in the Model[2].

Parameter	Value
Density, ρ	2.2 g cm−3
Young's modulus,E	0.45 GPa
Poisson ratio, ν	0.3
Area-related bond stiffness,Sb	13.5 · 1010 N m−3
Damping factor, α	0.95
Anode thickness,he	76.50 μm
Active material particle size	x10,3 = 4.03 μm
	x50,3 = 5.99 μm
	x90,3 = 8.94 μm

Under different calendering levels, the contact angle is less than 1°. Therefore, any area in the deformation zone is considered to be compressed in the vertical direction.

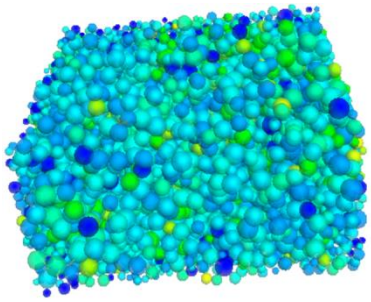

Figure 1. Schematic giagram of electrode microstructure.

2.3 Model Verification

The simulated model is validated using the experimental nano-indentation curve. The simulation verification process starts from the indenter applying pressure to the electrode until the maximum displacement of 7.65 μm, then unloading the indenter and returning it to its initial position. During the loading and unloading process, compression is performed by contcalendering the maximum indentation displacement at a constant speed of 0.15 m/s. The plastic (Wpl), elastic (Wel), and total (Wtot) deformation energy can be calculated based on the force-displacement curve as follows:

$$W_{tot} = \int_0^{h_{\max}} F_{load}(h)\,dh \tag{3}$$

$$W_{el} = \int_{h_f}^{h_{\max}} \mathrm{E}_{\mathrm{unload?}}(h)\,dh \tag{4}$$

$$W_{pl} = W_{tot} - W_{el} \tag{5}$$

Where hmax is the maximum displacement during indentation, h is the residual indentation depth, and Fload and Funload are the indentation forces during loading and unloading, respectively.

The verification process of this study is based on the research of Sangrós Giménez[6].The displacement and force situation of the nano-indentation head is detected and the curve is drawn to compare with the actual experimental measurement in as shown in Figure 2. The two curves are relatively consistent. Therefore, this model can correct.ly simulate the macroscopic elastic-plastic behavior of the material.

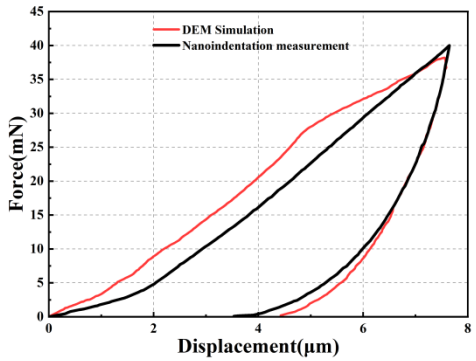

Figure 2. Comparison of nano-indentation force displacement between experiment and simulation.

3. RESULTS AND DISCUSSION

3.1 Analysis of Electrode Fracture Cracks

The electrode particles of lithium batteries will also leave cracks during calendering, and the development and penetration of these cracks have a significant impact on their mechanical properties. In the past decade, the use of the discrete element method to study crack propagation has matured, mostly analyzing at the two-dimensional level, because the visualization effect of three-dimensional crack development is not good. This section mainly analyzes the crack propagation situation of the two-dimensional prefabricated cracks. In PFC, in order to make the two-dimensional model in the two-dimensional numerical test equivalent to the mechanical properties of the three-dimensional entity, it is necessary to realize the porosity of the two-dimensional model equivalent to the three-dimensional entity. It is known that the porosity of the model built in three dimensions is 0.45. According to $y = 1.4618x + 0.08794$, where is the three-dimensional porosity and is the two-dimensional porosity. From this, the two-dimensional porosity of this model can be obtained as 0.25.

In the two-dimensional model, with the anode of 400 × 150 μm³ as the object, the model is established as shown in Figure 3a.A total of 1833 particles are generated in two dimensions. The mechanical integrity of the electrode material is directly related to the mechanical performance of the binder. Since all the particles simulated in this article are rigid bodies, the particles themselves will not deform or break, so the cracks studied in this article are all cracks in the binder. During the electrode calendering process, it can be observed that when the electrode calendering is about 11%, the internal bonding between the particles begins to break, as shown in Figure 3b. As shown in Figure 3c, when the calendering is about 15%,

the internal bonding between the particles in the electrode can be clearly found to be broken, but at this time, the electrode still has mechanical integrity. As shown in Figure 3d, when the calendering is 17%, micro-cracks can be observed inside the electrode, and the mechanical integrity of the battery electrode is damaged at this time. The calendering process strongly affects the performance of the battery, If cracks occur inside the electrode, it will lead to a reduction in the number of battery cycles and may even cause a short circuit inside the battery and cause a fire.

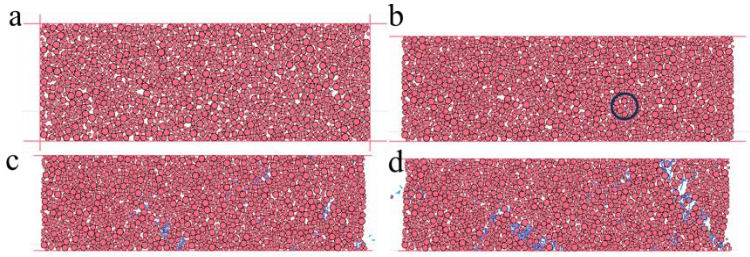

Figure 3. Distribution of intergranular cracks under different degrees of calendering: (a) 0% calendering; (b) 11% calendering (micro-cracks are circled in black); (c) 15% calendering; (d) 17% calendering.

3.2 Analysis of the impact of calendering thickness on heat transfer

In industrial production, hot oil is injected into the calenderer for calendering to cope with the strong compressive reality of the electrode, so that the consistency of thickness can be well contcalendered[4].

In this paper, the effects of strain in the calendering thickness direction of 0%, 0.5%, and 1% on the heat transfer of the electrode are studied. First, the wall on the left side of the two-dimensional model and the particles close to the wall are set to 380k, and the rest are all set to 300k. The specific heat capacity of the particles is 710 J/ (Kg · K)$^{-1}$.

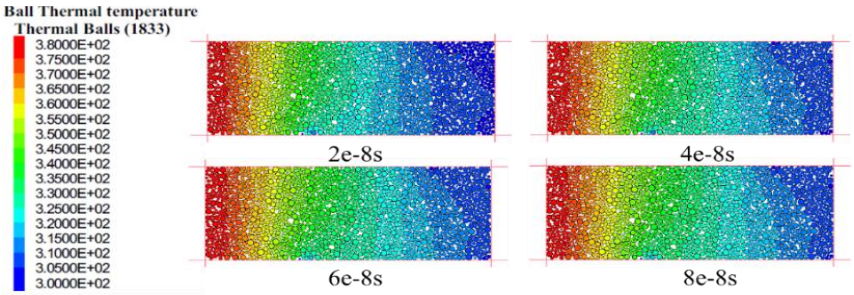

Figure 4. Heat transfer effect simulation diagram when the compaction degree is 0%.

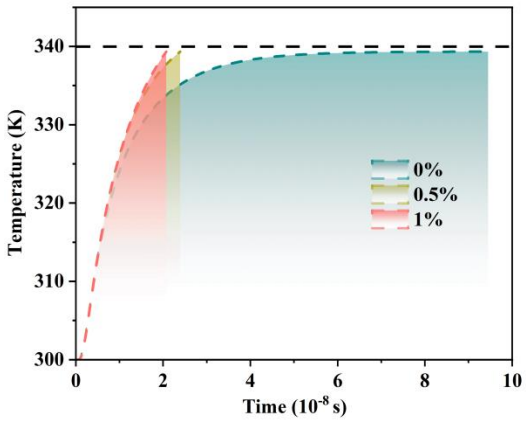

Figure 5. Diagram of particle temperature changes over time with different calendering amounts.

A simulation of the heat transfer effect at 0% compaction is given exemplarily in Figure 4. It is found that as the calendering strengthens, the heat transfer speed becomes faster (Figure 5). During the calendering process, as the porosity and thickness

decrease, leading to a linear increase in the coordination number. The larger the coordination number, the more contacts each particle has, leading to an increase in the possible number of heat transfer paths.

Considering that the calendering process determines the pore structure of the electrode and affects its heat transfer behavior, by establishing a DEM simulation model of the calendering and heat transfer process at the microscopic scale, the thermal behavior inside the electrode can be observed more intuitively, providing certain guidance for the production of batteries and subsequent safe use.

4. CONCLUSION

This paper uses the discrete element to numerically simulate the anode of lithium batteries, and deeply studies the impact of the calendering process on the microstructure, fracture behavior, and heat transfer characteristics of the electrode. The model is calibrated and validated using nano-indentation. The contact force and heat transfer characteristics of electrode particles during the calendering process were analyzed. It was found that as the calendering depth increases, the heat transfer rate will accelerate, and the temperature distribution will show more gradient characteristics. This paper also deeply analyzes the possible rupture and crack problems of the bonder in the electrode during the calendering process, and quantitatively analyzes the maximum compaction degree that the anode of the lithium-ion battery can withstand during the calendering process.

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