Review for mechanical integrity of lithium-ion battery

Characterization and Modeling of the Mechanical Properties

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Abstract

Li-ion batteries have been a dominant power source in commercial electronics and transportation, which is also the main power provider for newly-developing technologies such as self-driving cars. However,the safety concern is the main obstacle that hinders the wider applications of lithium ion batteries in these mobile use, in which batteries are confronted with various mechanical loads[1]. And in many cases, these can cause damage to the mechanical integrity of the batteries and then will easily develop into the thermal runaway[2]. As the beginning work of the final project, this report will review the current status of the mechanical integrity study on electric vehicle lithium-ion batteries, mainly focusing on the studies of different aspects, research methods and the unsolved problems. And main aspects of each study will be discussed in mechanical testing, component characterization and FE modeling.

Keywords: lithium-ion battery, mechanical abuse, internal short circuit

1. Introduction

The energy crisis and environmental pollution have been been chanllenges for human society in recent years[3], and thus an advanced technology which can store sufficient energy and easy to convert other types of energy into electric energy is in urgent demand[2].The coming era of electric energy and some other ripidly-developing technologies such as the self-driving cars are changing the energy storage system of vehicle from fossil to electrochemical energy storage systems[4, 5, 6]. And in this change of energy storage and propulsion system, lithium-ion batteries,have gained a variety of applications because of their high-energy density,durability and eco-friendliness[7, 8]. Considering the quantities of vehicles sold each year, it is reasonable to predict that vehicles may inevitably suffer from crash accidents and experience many other frequent intrusion or impact during working[9]. And actually, a number of fire and explosion incidents caused by LIB failure have been reported in recent years[10, 11],which caused the increasing concerns on the safety of the batteries.Among all the factors which can cause the failure of the batteries, such as manufacturing defects, overcharge or over discharge and mechanical abuse[1], all the other factors except the mechanical abuse have been studied detailed in the past several years and many methods have been adopted for prevention.

However, the mechanical integrity of LIBs package has only attracted considerable interest with respect to safety in recent years. Failures of batteries are also owing to the thermal runaway, and thermal runaway can be due to external or internal short circuit, which is the usual result of mechanical abuse. And the external short circuit happens when the exterior protection of battery tabs fail and two electrodes get in touch with each other[1]. And here the separators which separate the anode and cathode from each other can play an important role in the failure process. An indentation to a battery during manufacturing or usage in the field can cause deformation in its components, which in turn can lead to a failure in the separator. Negative and positive electrodes come into contact after separator rupture which leads to internal short circuit in the cell. Short circuit may trigger electrochemical reactions and result in thermal runaway in the LIB cell which can itself turn into a catastrophic event.To prevent that, it is of vital importance to understand teh deformation response from the macro scale to micro scale and then multi-scale.

To characterize the failure mechanism of LIBs, it should be noted that the whole process is quite non-linear,comlex,and thus cannot be described by one set of parameters in one scale level, so it is a must to consider the mechanism from components, cell and the batteries. In the following sections, the recent main develoments of mechanical characterization and modeling of Li-ion batteries under mechanical abuse conditions are reviewed.

The mechanical properties of LIBs can be studied at various length scales[1]. The smallest scale to study is the level of active material particles in the coatings of anodes and cathodes. Several studies have concentrated on the change of volume, deformation and crack formation in the active particles during the intercalation process in charging and discharging, evolution of degradation phenomena under externally induced mechanical loads and internally induced stress-diffusion coupling, damage evolution, and solid-electrolyte interphase (SEI) formation[12, 13, 14, 15]. These are the main causes of degradation in electrochemical properties of the cells as they go through a large number of cycles. The second scale is the component level, which means looking at properties of anodes, cathodes, current collectors, or separators at a single layer but in macro scale[16, 17, 18, 19]. The next stage in battery studies is investigating the properties at the single battery level (referred to as cell level in this manuscript). This can be done by considering cells as a homogenous material and ignoring their interior layered structure, or by more detailed analysis of evaluating the interactions of the layers that create a deformation at the cell level.[20, 21, 22, 23, 24] The last step will be considering the problem in hand at the system level, which includes stacks of cells, modules of batteries, and finally battery packs inside a vehicle[25, 26, 27, 28].

In this review, we will summarize the advances in mechanical test on micro particles, cell components, full cells(we will discuss two types of full cells models) under various loading conditions, and we will omit the discussion on modules and batteries packs. And the tests will include compression between flat plates, indentation (using hemispherical, cylindrical, conical punches, rigid rod), uniaxial or biaxial tension, and three-point bending. And tests at quasi-static and dynamic loadings, different SOCs, even the temperature(multi-physics) will also be discussed. And before we move onto the detailed discussion , it be noted that the time scale of the mechanical abuse events are often much samller than thermal or electrochemical loadings[1], and conseqently it is appropriate to do mechanical characterizations indepent from electrochemical state of cells also the components.

Apart from the experimental attempts on mechanical testing, it is also important to extract constitutive material behavior of battery cells or components, and several failure criteria with different levels of complexity have been proposed. Based on these, FE models are rapidly developed and modified, and all the above will be discussed or mentioned in the following sections.

2. Mechanical testing

2.1. The component

2.1.1. Electrodes

The mechanical testing of components of LIBs is the base of the further study, which consisits of two types of research:

1. Test the mechanical response of several components(anode and cathode, separators) and their failure mechanism.
2. Study the micro substances(the particles in both electrodes and the SEI interface) and give the fundamental explanation.

Actually for the first part, the base material of two electrodes is Aluminum or Copper whose mechanical responses are quite clear, so studies mainly focus on the mechanical response of the separators, about which we will detailedly discuss later. For the second part, many studies starting from chemical views focus on the stress on particles adhere to the electrodes during the charge and discharge.

We first discuss the second part, the studies can be divided into following three types:

1. The expansion of particles during charge and discharge circling causes the cracks and failures of the particles[29, 30], resulting in decrease of capacity and short life cycle.
2. The volume change of electrodes changes the stress state of separators[31] and causes the failure of the coating adhesion[32, 33] which will also result in decrease of capacity and short life cycle.
3. The failure of SEI interface[34] during charge and discharge.

The anode and cathode are composed of the current collectors with chemical-active particles. Several studies have been conducted on electrodes in plane and out of plane. And studies in plane suggest that the main mechanical response of electrodes are decided mainly by current collectors while the particles actually can have effect on the mechanical response[35, 36].And also studies have implied that the compression response is independent of SOC[37].

Following the mechanical tests of electrodes, several analytic models are also developed.And because of the anisotropy, the in-plane and out-plane properties are described separately. Models such as ideal elastoplastic model[38], bilinear elastoplastic model[39] and foam material model[40] are employed and also the effects of the particles are considered[35].

Aditionaly, the failure discipline of battery shell is proposed and well validated in quasi-static region[41], however the dynamic response is still in vacancy.

2.1.2. Separators

The component that prevents the electronic current occcuring between the electrodes, namely, prevents the direct contact between them, is the separator.The separator, as a consequence, must address the folowing requirements[19]:

- it should be porous to allow liquid electrolyte to carry lithium ions
- it should have sufficient strength to prevent contact between the electrodes.

The second requirment(mechanical strength) is the primary subject of the studies. Early studies[42] have employed two major measures to address the Young's modulus in machine direction(MD) and puncture strength[1].

While the above parameters may be sufficient to address integrity of separators during battery manufacturing and transport, they do not provide a full understanding of mechanical behavior of these membranes. Better understanding of mechanical properties of separators is especially relevant when the behavior of the battery under external mechanical loading (such as in the event of crush or drop) is considered[19]. Constitutive models describing mechanical behavior of separator can be used in numerical models for better predictions of battery response and improvements in safety[20, 43]. In addition, during

[1]Machine direction is chosen for dry processed separators since it is the direction of windings during jellyroll processing and thus the material should withstand tension from winding machine.

battery charging and discharging, separator can deform due to swelling of electrodes and thus change the effective properties in terms of ionic conductivity[44].
Recent studies on separators start to consider the complex situations such as different SOCs and in electrolyte solutions because the crashed usually occur while driving. A elastic response model of a commercial separator immersed in fluid to compression at different strain rates was established[45], which shows that the response of the separator is determined by combination of viscoelastic behavior of the polymer skeleton and poroelastic behavior, due to the flow of the fluid in the pores and poroelastic behavior causes effective stiffening of the separator, which increases with the strain rate. The quantitive model for the compressive behavior of the separator based on viscoelasticity and poroelasticity using two characteristic mechanical properties of the separator: effective Young's modulus determined in the region of small deformation and flow stress sucessfully describes the difference of these properties for the dry material and immersed in fluid. And the strong dependence of strain rate is also determined.
Although these tests of a fliud-immersed separator provides many insights on the separator behavior in the battery than the dry tests, they still do not represent the realistic loading conditions[45]Actually,in the battery the loads on a separator are compressive, both due to electrode swelling upon lithium intercalation and stacking loads[46]. Since the separators have complex anisotropic structure, and mechanical properties in lateral ('machine')direction and transverse direction are noticeably different[16],it is not viable to predict the behavior in compression based on the properties obtained from these tests. More recently, an experimental study of the separators under mechanical loading was conducted[18], in which researchers discovered two distinct deformation and failure mechanisms and based on that a criterion for predicting onset of soft shorts.

2.2. *Battery cell*

At the scale of battery cells, safety studies were more focused on thermal and electrochemical aspects[47, 48, 49]. For the mechanical peroperties and response, over the past five years a series studies have investigated the mechanical deformation and onset of short circuit in lithium-ion battery cells. Punch loading has been used as a method of studying internal short circuits in batteries by focusing on thermal response of the cells[50, 51]. And then first measurements of force-displacements in mechanically induced short circuit tests of small pouch cells and cylindrical cells was reported[20, 22, 21]. Following that, in-plane compressive properties of battery cells and modules of large pouch cells were studied[43, 52], while structural analysis of large cylindrical cells was later reported[53].

2.2.1. *Coupled strain rate and SOC dependencies*

Based on the previous pioneering work which focused on the individual components properties including electrodes and separator and the general response of the LIB subject to various mechanical loadings, such as radial crushing, bending and indentation. In the battery cell level, bending, indentation, and radial crushing [21, 22] tests were proposed to study the overall mechanical response subject to loadings, where jellyroll is regarded as a macroscopic homogeneous model. In a recent work, an anisotropic homogeneous model describing the jellyroll and the battery shell is established and validated through compression, indentation, and bending tests at quasi-static loadings[9]. In this model, state-of-charge (SOC) dependency of the LIB is further included through an analogy with the strain-rate effect. Moreover, with consideration of the inertia and strainrate effects, the anisotropic homogeneous model is extended into the dynamic regime and proven capable of predicting the dynamic response of the LIB using the drop-weight test.
Additionally, another study investigates the electrochemical failure behaviors of LIBs with various SOCs under both compression and bending loadings, underpinned by the short circuit phenomenon[37]. Mechanical behaviors of the whole LIB body, which is regarded as an intact structure, were analyzed in terms of structure stiness. Results showed that the mechanical behaviors of LIBs depend highly on SOC. Experimental verification on the cathode and anode sheet compression tests show that higher SOC with more lithium inserted in the anode leads to higher structure stiness. In the bending tests, failure strain upon occurrence of short circuit has an inverse linear relationship with the SOC value.

2.2.2. *Dynamic loading*

Most of the studies and continued research on mechanical characterization of lithium-ion cells and their components were focused on quasi-static testing[24], however, dynamic loading,in essence is the bottleneck in studying the LIB mechanical integrity at crash scenarios[9].
One difficulty when doing dynamic testing arises from the combination of high punch velocities in conjunction with the small cell thicknesses. And in a newly-published work, dynamic local indentation tests have been performed in the velocity range between $0.01mm/s$ and $5m/s$[24]. With two different types of pouch cells, it was demonstrated that the critical force necessary to generate an internal short, defined as first force maximum in the load displacement curves, drops roughly in the test velocity range. The apparent strain rate effect of decrease in critical load is an interesting feature of the dynamic response of pouch cells. It is both counter intuitive and difficult to explain. Also it was found with the wet cells that the voltage drop often starts before the force maximum was reached. Different failure modes were found which are consistent with two failure modes observed at dry processed separators.

3. Modeling and Simulation

In the past years, many pioneering efforts were made to elucidate the mechanical safety behavior of LIBs on multiple length scales, ranging from component material scale to battery pack scale. On the component level, the mechanical properties of the case[22], separator[54], anode, and cathode foils[43] of the cell have been investigated. These results will help to understand the failure mechanism of internal short circuit of lithium-ion battery, but can not characterize the global mechanical behavior of the battery. The finite element (FE) models were used to understand the mechanical properties and predict extreme cases. Since coating of active material coating and separator are highly porous and soaked in the electrolyte, the detailed modeling including each component and interaction among them is very complicated. Therefore, it is reasonable and acceptable to take jellyroll as a homogeneous material[21, 55]. Furthermore, the dynamic behavior[9] and SOC effect[37] of the battery have been studied and the results suggested that higher SOC leads to higher structure stiffness.

3.1. Homogenized modeling methodology

Effective lithium-ion battery module modeling has become a bottleneck for full-size electric vehicle crash safety numerical simulation. Modeling every single cell in detail would be costly. However, computational accuracy could be lost if the module is modeled by using a simple bulk material or rigid body. To solve this critical engineering problem, a general method to establish a computational homogenized model for the cylindrical battery module is proposed[56].

The homogenized method is actually treating the whole cell as one material and then establish the mechanical model to describe the mechanical response. And the descriptions adopted in former researches mainly have two types:

- The method raised by Greve and Feherenbach[21]:

$$\sigma = A\epsilon^n + B \tag{1}$$

- The method raised by Wierzbicki and Sahraei[55]:

$$\sigma = A\epsilon^n \tag{2}$$

The two models have been widely used in the FEM modeling of batteries and have gained good validation. However,the model can not describe the three point bending stress state and also fail to decribe the dynamic response. So the new homogenized model should be established to improve the remainning results. Besides, detailed model was also developed to describe the every single layer's mechanical properties[52]. And there are also studies on the deforming and failure mechanism of each component using this model.[38, 41, 57]. But this model can only simulates the single stress condition becasue of the lacking information of the mechanism of components considering the anisotropy. Also several attempts to combine the detailed model with the homogenized model[40, 21] have gained good effects and are still in the development.

3.2. Dynamic mechanical integrity modeling

Previous investigations for LIB mechanical behavior started from the elastic regime under quasi-static loadings[58] where a multi-scale numerical simulation model was suggested to describe the surface kinetics and electrochemicalmechanical phenomenon in a simplified LIB model. Considering the fact that mechanical, electrochemical and thermal properties of LIB were closely and complicatedly intertwined, it was difficult to analyze and predict LIB failure in terms of the mechanical integrity. It is not surprising that previous studies of LIB mechanical integrity and safety focusing on the quasi-static rather than dynamic behaviors due to the safety concerns e.g. possible fire or explosions in dynamic experiments. However, LIB usually experiences severe damage or fail mechanically and electrically at extreme events such as impact and dynamic crushing from time to time where dynamic mechanical loading is involved. In a new work[59], by considering the strain rate and inertia effect of the battery structural and material, the dynamic mechanical behavior of lithium-ion battery is investigated. Different mechanical failure behaviors are obtained through the combination of numerical simulation and the suggested battery mechanical integrity criteria.

3.3. Short circuit and mechanical failure criterion for simulations

When it comes to the failure criterion for battery failure, early studies focused on the two direct parameters: voltage and temperature, which are mostly used for deciding the failure of the batteries.Generally, when the short circuit occurs, the voltage begins to decline and the temperature becomes higher. However, these methods are not always effective in predicting the failure, so considering the mechanical response of batteries can be another way for the failure criterion.

In this aspect, both experiments and simulations have been carried out.In indentation experiments, short circuits are found when the loading suddenly reduce[21, 20], and in three point bending experiments, shorts circuits often take place after the sudden cut-down[37, 22].Moreover, researchers are also interested in the prediction and determination of the short circuit and mechanical failure by simulation ways.For the homogenized method, stress-based criterions are usually adopted. For example, maximum principal stress law was used as the short circuit criterion[22]. And $More - Coulomb$ rule was also used[21], which considers the maximum principal stress σ_1 and minimum principal stress σ_3 and goes as follws:

$$\sigma_1 = p\sigma_3 + 2c \tag{3}$$

where p and c are fitting parameters.

Furthermore, the double shear strength theory which includes maximum principal stress, minimum principal stress and intermediate principal stress is more accurate in predicting[59], and its equivalent stress takes the form:

$$\sigma_{eq} = \begin{cases} \sigma_1 - \dfrac{\alpha}{1+b}(b\sigma_2 + \sigma_3) & \sigma_2 \leq \dfrac{\sigma_1 + \alpha\sigma_3}{1+\alpha} \\ \dfrac{1}{1+b}(\sigma_1 + b\sigma_2) - \alpha\sigma_3 & \sigma_2 \geq \dfrac{\sigma_1 + \alpha\sigma_3}{1+\alpha} \end{cases} \tag{4}$$

And these criterions are quite effective in predicting the mechanical failure in homogenized model.

For detailed model, FEM has been a powerful tool for the modeling and helps to predict the failure od the whole battery based on the deformation and failure behavior of each component[36]. Most studies suggest that the stress of the separators can also help predict the failure[38, 60] and the failure of separators can be a major cause of the short circuit[61].

4. Conclusion and our further research

4.1. Summary and Remarks

In the above sections the various approaches of mechanical testing, material characterization, finite element modeling, and validation procedures used in investigating the mechanical integrity of Li-ion batteries at the cell level have been discussed. However, there remains much work on the mechanical test, material characterization and FE modeling[1].

An anisotropic model which can produce more precise prediction especially in terms of failure location and direction is in urgent demand for the multi-physics modeling, although simple isotropic foam models still produce accurate results in terms of global response of the cell. It is known that properties are significantly different in tension and compression, and the models that consider this variation and produce more predictive responses should be based on more deeper understanding of the mechanical response of batteries in multi-scale.

As for the FE model, finite element analysis provides a cost effective tool which can be used to optimize the design parameters and improve the safety of the cells without the need to conduct more experiments. Depending on application of the model, different levels of detail have to be included. A model that is developed to simulate a cell in a battery pack for vehicle level crash has different requirements than a model to design layers of electrode and separators for improved mechanical integrity. The former requires the largest possible element size to reduce computational time and has to only predict global deformations and failures, while the latter has to include as many details as possible from electrode separator assembly to clarify effects of interactions between layers. All of the above requires a more deep and clear characterization of batteriy and its components in different scales and under varied loading conditions.

4.2. Our further research: an introduction

4.2.1. Research focus and current status

In our reviewed sections, research work on lithium-ion batteries and components as well as their physical and electrochemical properties have been detailedly stated. However, one important property of the batteries which can greatly affect the performance of batteries have not been fully studied, that is, the interfaces of several components preperties in batteries, such as the coating between the electrodes and the active particles, the separators between two electrodes.

For the coating towards the electrode(particle-binder-substrate interface), its adhension strength is of vital importance and it must resist further cell processing steps, such as electrode cutting, winding and electrolyte wetting[62]. More specifically, the coating has to withstand the mechanical demands during cell operation. This includes the mechanical stress during lithium intercalation and de-intercalation due to the expansion and shrinkage of the electrochemically active material particles[63, 64, 65]. This swelling especially induces mechanical tension at the particle-binder-substrate interface. Hence, mechanical properties strongly influence the cycle performance and life of the cell[66, 67, 68, 69]. A local delamination of the coating from the current collector could impair the electrical contact[70] and leads to inhomogeneities in current density in the electrode volume. This in turn results in local stress-dissimilarities and, as a consequence, to a smaller life span. Furthermore, electrical decoupling affects the content of electrochemically accessible active material and, therewith, the capacity based electrode balancing of the cell. In this case the state of safety of a lithium-ion battery can be influenced if the anode capacity drops and deposition of metallic lithium occurs. Also, the friction properties of the inerface between the active component and the separates can have a profond effect on the performance of batteries under mechanical loading.

The coating adhesion strength of lithium-ion battery electrodes is a very important mechanical property, affecting the electrochemical life time of battery cells and electrochemical handling during cell manufacturing[62]. What's more, the debonding between the coating material and the current collector is also one of the most common failure scenarios. The characterization and full understanding of the mechanical response and the failure mechanism of the coating as well as other interfaces will not only directly provide the more detailed and accurate modeling of the internal structures of batteries which can, for example, help improve the FE models, but also can help understand and solve several common failure modes of the batteries.

4.3. Research goals and plan

Our research is aimed at establishing a complete and accurate model of the coating between electrodes and active particels and that between separators and active particles which includes the comprehensive understanding of that issue. Considering that past researches mainly focus on the qusai-static loading conditions, we will further consider the dynamic loading, with or without electrolyte and also complex loadings to get their overall mechanical response. Studies will be carried out according the follwing plan:

1. Design and set up the measurement establishment and conduct the quasi-static tests.
2. Conduct the tests under conditions, such as with or without electrolyte and different temperatures.
3. Perform the dynamic tests under different loading rates.
4. Do the characterization of the mechanical response of the interfaces and establish the models based on the testing results.

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