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Failure in lithium-ion batteries under transverse indentation loading

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HIGHLIGHTS

- Unique failure patterns under transverse punch loadings are observed.
- Closed-form solution is derived for traverse loadings on Li-ion cells.
- Proposed Mohr-Coulomb fracture criterion predicts well failure of lithium-ion cells.

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ABSTRACT

Deformation and failure of constrained cells and modules in the battery pack under transverse loading is one of the most common conditions in batteries subjected to mechanical impacts. A combined experimental, numerical and analytical approach was undertaken to reveal the underlying mechanism and develop a new cell failure model. When large format pouch cells were subjected to local indentation all the way to failure, the post-mortem examination of the failure zones beneath the punches indicates a consistent slant fracture surface angle to the battery plane. This type of behavior can be described by the critical fracture plane theory in which fracture is caused by the shear stress modified by the normal stress. The Mohr-Coulomb fracture criterion is then postulated and it is shown how the two material constants can be determined from just one indentation test. The orientation of the fracture plane is invariant with respect to the type of loading and can be considered as a property of the cell stack. In addition, closed-form solutions are derived for the load-displacement relation for both plane-strain and axisymmetric cases. The results are in good agreement with the numerical simulation of the homogenized model and experimentally measured responses.

1. Introduction

Development of reliable deformation and failure models of lithium-ion cells relies heavily on a good set of experimental data. At a level of individual cells, a choice of tests is limited to compression, indentation, and bending. Tensile and shear tests cannot be performed because cells do not have rigid shoulders to allow for firm mounting in the mechanical, hydraulic or pneumatic grips. Sahraei et al. [1–4] in a series of papers established a set of experiments needed to calibrate a homogenized model of batteries subjected to out-of-plane and in-plane compression. Also, a procedure of predicting failure of cells, understood as an onset of an electric short circuit, was developed by means of an inverse experimental/computational approach. Bending tests were performed by Sahreai et al. on small flat cells with and without pouch cover [2]. It was found that the pouch cover, that contributed little to the resistance to indentation, has a major effect on the bending strength. A simple model of the multilayered beam was able to explain the source of these differences. Greve et al. [5] performed a similar type of tests to establish failure limit by means of a coupled experimental/computational method. Zhang et al. [6,7] developed a lumped mechanical model for a coupled modeling methodology encompassing the mechanical, electrical and thermal response. Wang et al. [8] studied in-plane confined compression of pouch cells and explained a regular buckling pattern through the development of shear localization. Mason [9] constructed an apparatus to perform in-plane compression tests with controllable through thickness confinement force. Zhu et al. [10] subjected 18650 cylindrical cells to axial compression and developed a detailed computational model. The prediction of this model agrees

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closely with post-mortem tomography of damaged cells. Kisters et al. [11] recently reported dynamic impact tests on the pouch and elliptic cells with axisymmetric punches.

The question that must be posed is whether all these types of tests correspond to typical accidental loading pattern of various cells in everyday situations. The answer is no. For example, while the indentation of cells by an axisymmetric punch with different tip radius is widely used to calibrate battery models, it is also only one type of load application. Equally important is a plane-strain loading where a pouch or prismatic cell is loaded transversely by a long cylinder. Actually, in a number of tests proposed by various standards [12,13], loading is applied through rigid cylinders. Recent work on fracture of metallic material has proven that the plain strain is the most constrained case among all possible stress states and the corresponding equivalent failure strain is the lowest. Understanding and quantification of plain-strain failure of batteries will always provide the lower bound or worst case scenario. Still, a rigorous analysis of deformation and failure of cells under such loading is missing.

The objective of the present paper is to close the gap in our current experimental practice by characterizing the failure patterns under the various loading conditions and developing the failure criterion at the cell level. The results obtained have proven that the Mohr-Coulomb (MC) failure criterion [14], used in many branches of solid mechanics is also applicable for predicting failure of lithium-ion cells. A simple set of tests is then proposed to calibrate the two-parameters of the MC model. The present model is physically based and could replace the inverse experimental/numerical methodology that has dominated the current literature on cells failure.

Another major new result reported here is the derivation of the closed-form solution for the load-displacement relationship of cells subjected to lateral indentation by hemispherical and cylindrical punches. Both solutions compare well with the experimental results and finite element (FE) simulation. So, it can be used instead of a numerical method with no compromise in accuracy. It is believed that the present paper is putting the development of mechanical models of batteries on a firmer theoretical foundation and is providing new analytical and numerical tools for more reliable prediction of strength and onset of an electric short circuit for large format lithium-ion pouch cells.

2. Cell-level test and modeling

The lithium ion cell studied here is a 26.3 Ah large pouch-type cell with $LiMn_2O_4/LiNi_{1/3}Co_{1/3}Mn_{1/3}O_2$ - graphite chemistry and carbonate-based liquid electrolyte. The cell dimensions are 232 mm (L) x 153 mm (W) x 7.5 mm (T). The architecture of the cell consists of alternating layers of 20 anodes, 19 cathodes and separators in between them. All the cathodes and anodes are double-side-coated and the thicknesses of electrodes are 171 µm for cathode and 143 µm for anode. The current collectors are 20 µm aluminum foil for cathode and 10 µm copper foil for anode. The separator (27 µm) is dry-processed polypropylene (PP) with ceramic coatings on both sides. The exterior of the cell is an aluminum laminated film with 150 µm thickness. Except the materials mentioned here, there is no other material layer affects the mechanical strength of the lithium ion cell.

Cells were tested to evaluate load-displacement response and short circuit initiation under transverse punch loadings. All tests were performed at room temperature. Flat, hemispherical and cylindrical punches were used for tests and the list of punches are as follows:

- Flat punch (ø44.5 mm)
- Hemispherical punch (ø12.6 mm/ø28.6 mm/ø44.5 mm/ø90 mm)
- Cylindrical punch (ø15 mm/ø28.6 mm)

The strain state under flat and hemispherical punches are axisymmetric and that of lithium-ion cylindrical punch is predominantly plane-strain. The cylinders were aligned with the machine direction (MD) of the electrodes and the separator, so the plain strain cross section became transverse direction (TD). The experimental results were also compared to the results from a global homogenized computational model.

2.1. Experimental results

All cells were tested at 0% state of charge with open circuit voltage of about 3 V. The indentation rate of the punch was 1 mm/min (quasi-static) using a 200 kN loading frame (MTS Systems Corporation). The cell voltages were monitored during the tests to identify an internal short circuit where the voltage starts to drop from the initial value. The tests were stopped after observing peak load and voltage drop. Should the tests be continued, there will be a progressive compression of the coated material until full densification. In the final phase, the load will rapidly increase. Details of experimental method can also be found in the study by Sahraei et al. [4], which reported the characterization of lithium-ion pouch cells using the same method.

Load-displacement curves and voltage changes are shown in Fig. 1. Comparisons between the load-displacement curves and the voltage curves show that the force drops match with where the voltage drops, implying that initiation of inner mechanical failure triggers a short circuit in all test cases. For ø44.5 mm and ø90 mm hemispherical punch tests, only load-displacement responses were measured without voltage profile because, due to the shortage of cells available to test, we used already short-circuited cells by other indentation tests. (The locations of indentations were far enough not influencing each other.)

2.2. Finite element simulations

Finite element models were developed using LS-DYNA with a similar procedure developed by Sahraei et al. [3,4]. There were 16 elements through the cell thickness in all FE simulations giving the mesh size 0.469 mm. The cell stack was described by the compressible foam model (MAT 63) from the library of LS-DYNA materials and the compressive curve calculated from 44.5 mm flat punch is used for the input. The properties in tension are assumed to be linear up to a tensile cut-off value of $\sigma_f = 21\text{MPa}$. This value was obtained from a composite tensile curve calculated from the uniaxial tensile test results of cathode, anode, and separator. The compression curve was obtained from the 44.5 mm flat punch test (Fig. 1 (a)) with the assumption that there was a negligible edge effect. The edge effect of a flat punch is previously calculated by Sahraei et al. [4] and was shown to be less than 5% for a 25.3 mm diameter flat punch, and smaller for larger punch sizes. The punches were treated as rigid bodies because the strength of steel punches is more than an order of magnitude larger than that of the battery. Two different failure criteria were used in the numerical simulations. The initiation of failure in this section is assumed to be controlled by maximum principal strain ε_f and this value is calibrated by comparing experimental and modeling results. The application of the MC criterion is covered in Section 4. Also, it should be noted that a model is expected to be realistic until one of its elements reaches to the failure criterion. The physics after failure is not considered in this model, which is aimed at predicting load-displacement and failure (the first peak).

Modeling results are plotted in Fig. 1 in addition to the experimental data. Their force-displacement curves match generally well with the experimental results. However, the magnitude of the maximum principal strains ε_f was found to be different for the axisymmetric and the plane-strain conditions to match the failure points. For the axisymmetric tests (flat and hemispherical punches), the model was calibrated with the maximum principal strain $\varepsilon_f = 0.1$ and, for the plane-strain tests, the maximum principal strain was lowered to $\varepsilon_f = 0.043$ in order