Material Characterization of Li-ion Battery Segments Subjected to Lateral Compression and an In-Plane Tension Loads

by

Sagy Hakoon

B.Sc., Tel-Aviv University (2005)

Submitted to the Department of Mechanical Engineering in Partial Fulfillment of the Requirements for the Degrees of

Master of Science in Naval Architecture and Marine Engineering
and
Master of Science in Mechanical Engineering at the
Massachusetts Institute of Technology

February 2017

© 2017 Massachusetts Institute of Technology. All rights reserved.

Signature of Author: _________________

Department of Mechanical Engineering
January 18, 2017

Signature of Author: _________________

Certified by: __________________________
Tomasz Wierzbicki
Professor of Mechanical Engineering
Thesis Supervisor

Elham Sahraei
Mechanical Engineering
Thesis Advisor

Accepted by: __________________________

Rohan Abeyaratne
Professor of Mechanical Engineering
Chairman, Department Committee on Graduate Students
Material Characterization of Li-ion Battery Segments Subjected to Lateral Compression and an In-Plane Tension Loads

by

Sagy Hakoon
B.Sc., Tel-Aviv University (2005)

Submitted to the Department of Mechanical Engineering on January 18, 2017, in partial fulfillment of the requirements for the degrees of Master of Science in Naval Architecture and Marine Engineering and Master of Science in Mechanical Engineering

Abstract

In the last two decades, Lithium-ion (Li-ion) batteries have become an inherent part of day-to-day life thanks to their widespread use in many consumer products and electric vehicles. While these batteries possess great advantages, they also carry an inherent safety liability: In case of a crash event, short-circuit failure of the battery may develop, leading to thermal runaway, fires and even explosions. Hence, a comprehensive study is required, aimed to modulate these batteries and optimize their testing standard.

The objective of this research was to characterize the effect of lateral compression on the in-plane tensile failure load of Li-ion battery segments. A new experimental system was developed, which allows fine control of the compression load, and decouples the out-of-plain compression load and the in-plain tension load. Then, measurements were conducted with single-layer, 4-layers and 11-layers specimens, producing characterizing graphs of the tensile load versus displacement.

For all types of specimens, results show an observable decrease in the failure load for increasing pre-compression load, as expected. Furthermore, measurements confirmed that the relation between the tensile load and the displacement does not change for different compression loads.

For the multi-layer specimens (4 layers), the failure sequence was studied. It was found that the sequence may alter for different pre-compression loads. Nevertheless, on all cases, the cathode failed first, and the anode failed second.

Throughout all experiments, failures were located on the edge of the compression area of the specimen. Several methods were used to encourage emergence of failure at the center, but with no success. A hypothesis to explain the development of this mode of failure is suggested at the end of this work.
Acknowledgement

The research presented in this document was carried out at the Impact and Crashworthiness Lab at the Department of mechanical engineering at MIT. It was completed with the support of several individuals, whom I would like to mention:

First, I would like to thank Professor Tomasz Wierzbicki, founder and chief of the Impact and Crashworthiness Lab, for embracing me to the unique and prestige Li-ion battery research group. His profound knowledge and wide experience guided me towards a successful completion of this work.

Also, I would like to express my deepest thanks to my brilliant advisor, Dr. Elham Sahraei, for her excellent guidance, high availability and endless patient and support. Thanks to her enlightening, knowledgeable, comments during our discussions, complicated issues were solved and new solutions were developed.

Next, I would like to express my high appreciation and gratitude to the senior doctoral researcher in the Li-ion battery group, Mr. Xiaowei Zhang, for his thorough and comprehensive guidance at the lab and for his contribution along the design process. I would also like to thank Mr. Zhang for his encouragement and friendship.

Finally, I would like to thank my precious family: My wife, Oshrit, for being so much more than a supportive spouse, best friend and a wonderful mother, and my beloved, beautiful children, Noam and Evyatar, who always keep my mind focused on what is truly important.
# Table of Contents

Abstract ........................................................................................................................................ 1  
Acknowledgement ....................................................................................................................... 2  
List of Figures ............................................................................................................................ 4  
1. Introduction .......................................................................................................................... 6  
2. Experimental apparatus ........................................................................................................ 9  
   2.1 Scheme of the experimental apparatus ............................................................................. 9  
   2.2 Zero and Non-Zero Pre-Compression Relation .............................................................. 10  
   2.3 Tension Compression System (TCS) ............................................................................ 10  
3. Design Validation Experiment ............................................................................................. 13  
   3.1 Phase I of validation experiments: Using existing punchers .......................................... 15  
   3.2 Phase II of validation experiments: Using costume-made punchers ............................... 18  
   3.3 Conclusion ...................................................................................................................... 19  
4. Results .................................................................................................................................. 21  
   4.1 Zero load on anisotropic separator .................................................................................. 21  
   4.2 Zero load on isotropic separator .................................................................................... 23  
   4.3 Pre-compression on isotropic separator ......................................................................... 24  
   4.4 Multi-Layer Experiments .............................................................................................. 26  
   4.4.1 Zero load on multi-layer specimen .......................................................................... 26  
   4.4.2 Pre-Compression on multi-layer specimen ............................................................. 28  
   4.4.3 Failure Sequence of Multi-Layer set ......................................................................... 29  
   4.4.4 11 layers test .............................................................................................................. 32  
   4.5 Using Foam in Compression Area .................................................................................. 34  
5. Summary and Conclusions .................................................................................................... 37  
6. Future Work .......................................................................................................................... 39  
7. References ............................................................................................................................. 40  
Appendix I – Drawings of Tension Compression System ......................................................... 41  
Appendix II – Design Improvements for the TCS system ......................................................... 56  
Appendix III – Plate Strength Calculation .............................................................................. 59  
Appendix IV – New Contact Interface Suggestion .................................................................. 61
List of Figures

Figure 1: Li-ion battery cell construction. The cell is soaked in Lithium salt solution (the electrolyte) ...................................................................................................................................... 7
Figure 2: Desired load case on battery cell. Blue load: Constant out of plane pre-compression. Green load: Increasing in-plane tension ..................................................................................................................... 8
Figure 3: Schematic drawing of the experimental apparatus ............................................................................................................................... 9
Figure 4: qualitative illustration of the effect of compression load on maximum tensile load .... 10
Figure 5: 1st concept for Tension Compression System (TCS) ............................................................................................................................. 11
Figure 6: Second concept: left is the compression system using a compression spring. The segment is located between the two rods. Right is the moving part which is connected to the INSTRON and mounting the segment............................................................................................................................. 12
Figure 7: Second design concept: left (a): design drawings, right (b): full set up as built in the ICL. Upper part is connected to INSTRON load cell, bottom part is connected to INSTRON table... 12
Figure 8: Separators after failure exposed to lateral compression and in-plane tension. At the left specimen where the failure is inside the compressed area combined with outside failure, at the right specimen where the failure occurred outside the punched area .............................................................................. 13
Figure 9: Experimental setup. After reaching to required compression load, the tensile load is applied by hand................................................................................................................................................. 14
Figure 10: Experimental setup for 1.5mm flat puncher with sharp edges and 2 thin plates attached for specimen mounting. On the right is enlarged view on the puncher (qualitative geometrical relations) ......................................................................................................................................................... 15
Figure 11: Experiment #3: mounting with rounded protrusion ........................................................................................................................................... 16
Figure 12: failure mode of using 11mm cylindrical puncher, Compression load 1500N, isotropic specimen. The failure is on puncher’s edge........................................................................................................................... 17
Figure 13: failure mode of using 11mm cylindrical puncher, Compression load 200N, anisotropic specimen. The failure is in the compressed area.................................................................................................................. 17
Figure 14: hemispheric puncher with a flattened tip: 3 and 6 mm diameter ............................................................................................................................... 18
Figure 15: phase II of validation experiments. Anisotropic separators after failure using 6mm puncher. The red color was used to view more clearly the failure area ........................................................................................................................................... 19
Figure 16: phase II of validation experiments. Anisotropic separators after failure using 3mm puncher. For 650N the failure mode was already outside of the compression area ........................................................................................................................................... 20
Figure 17: Experiment #7: single layer anisotropic separator right before failure, 12.7mm puncher.......................................................................................................................................................................................................................... 22
Figure 18: Experiment #7: single layer anisotropic separator for zero compression load, 20mm diameter puncher ........................................................................................................................................................................................................................................ 22
Figure 19: Experiment #7: Off center Failure. Left: 20mm hemisphere, Right: 6.35mm hemisphere. ........................................................................................................................................................................................................................................... 22
Figure 20: isotropic separator – zero compression load ........................................................................................................................................................................................................................................ 24
Figure 21: Zooming in on the pre-compression ........................................................................................................................................................................................................................................ 24
Figure 22: Force vs. Displacement for various pre-compression stress on isotropic separator specimen ................................................................. 25
Figure 23: layers order in the multi-layer specimen ................................................................. 26
Figure 24: Force-Displacement curve multi-layer without pre-compression. The picture on the left is a typical type of failure of the four layers. The order (bottom-up) is in the same order as in the fixture ...................................................................................................................................... 27
Figure 25: A typical look of the multi-layer specimen after multi-layer failure. From left to right: Anode (bottom), intermediate separator, cathode, separator (top) ................................................................. 27
Figure 26: Force-Displacement curve multi-layer without pre-compression .......................... 28
Figure 27: Force-Displacement curve multi-layer under various pre-compression ............... 29
Figure 28: Determining the failure sequence for zero pre-compression ................................. 31
Figure 29: Determining the failure sequence for 10Mpa pre-compression ......................... 32
Figure 30: 11 layers zero load test. The order of layers shown on the left is in the same order as in the fixture .................................................................................................................................. 33
Figure 31: compression area - new setup ............................................................................... 34
Figure 32: left – foam and hemisphere before compression. Right - Foam is compressed and squeezed – highest and stress at the center to encourage failure strain ........................................... 34
Figure 33: hemisphere Teflon puncher – Comparison between zero load and 70N load using medium hardness foam ................................................................................................................. 35
Figure 34: hemisphere Teflon puncher – Comparison between two different foams under the same pre-compression .......................................................................................................................... 36
1. Introduction

Rechargeable Lithium-ion (Li-ion) batteries are widely used in many day-to-day life electrical devices. Thanks to their high energy density and the compact, light packaging, this type of battery has no competitors in the industry of portable devices such as smartphones, laptops, and digital cameras. It is also used in power tools such as drills, sanders, and saws. Research and development of new applications of Li-ion batteries is continuously evolving. In the field of underwater vessels, for example, the applicability of using them as the main power source is being thoroughly studied [1], [2]. Another, more innovative example is using Li-ion batteries in electric vehicles [3].

There are several other types of rechargeable batteries, among which we can find Lead-acid, Aluminum-ion, and Nickel metal hydride batteries. However, Li-ion batteries offer many advantages compared to these: They have higher power density than Lead-acid batteries, they offer longer shelf-life than Aluminum-ion batteries, and their self-discharge is much lower than that of Nickel-based batteries. These advantages keep the Li-ion batteries on top, far above the others.

Nevertheless, with all the remarkable advantages mentioned, the Li-ion batteries contain an inherent safety liability since their electrolyte, Lithium salt, is flammable; when a short circuit failure occurs in the battery (contact between the anode and cathode), a process of thermal runaway may develop causing fires and even explosions [4]. Several Incidents like explosion of a smartphone or laptop, and car fires due to battery failure have occurred in recent years, leaving catastrophic outcomes in some cases. In an attempt to diminish the odds for such a failure, the testing standard for Li-ion battery cell is being continuously examined and expanded by both academy and industry.

Figure 1 depicts a typical Li-ion battery cell, assembled of the following segments:

i. Anode - the negative electrode, made of graphite.
ii. Cathode - positive electrode, pure Lithium metal oxide.
iii. Conductive surfaces – Aluminum, attached to the cathode and Copper, attached to the anode.
iv. Electrolyte – the liquid medium made of Lithium salt, which allows the Lithium ions move freely from the cathode to the anode (charging) or from the anode to the cathode (discharging).

v. Separator – mechanical partition between the anode and cathode preventing short circuit. The separator allows the Lithium ions to move freely (micro-porosity property).

![Li-ion battery cell construction](Image)

Figure 1: Li-ion battery cell construction. The cell is soaked in Lithium salt solution (the electrolyte)

Short circuit in the battery cell occurs when the separator is pierced or collapses. Piercing might happen, for example, when Copper or Aluminum metal separation is developed; the shreds pinch the separator and create damage. Today, thanks to adequate design and manufacturing, the odds for this scenario has been reduced significantly. The more likely failure of the battery cell is collapse of the separator due to mechanical overload, weather it is a static load, dynamic load or both. This scenario might develop in case of a crash, and its exact buildup and consequences are hard to predict. Hence, a study on the mechanical properties of the separator and other segments of the batteries is essential in order to characterize the failure for a given load and develop better battery packs.

The Impact and Crashworthiness Lab (ICL) at MIT has begun its study on Li-ion batteries in 2010, conducting simulations and experiments on different types of battery cells and their interior components. The first experiments were uniaxial load tests for batteries type “A” [5] and batteries type “B” [6]. The latter work also included biaxial punch tests using 25mm hemisphere punchers on both types of batteries, single layer tests, in which only one of the battery components is subjected to load and multi-layer tests. Later, uniaxial and biaxial load experiments were conducted on prismatic, elliptic, and pouch battery cells [7]. The experiments results, displacement versus load curves, were then used as an input values for numerical simulations of large scale of an elliptic battery cell in which the maximum stress and strain before failure is evaluated.
In the current work, another load case which the battery components might be exposed to is being investigated using a unique experimental system designed for this purpose. Figure 2 describes this load case: Applying increasing in-plane tensile load on segments while they are subjected to out-of-plane pre-compression. The motivation to study this scenario can be traced in previous work [8] in which a micro mechanical model was developed and subjected to various loading scenarios in order to better understand the sequence of failure in Li-ion components.

Figure 2: Desired load case on battery cell. Blue load: Constant out of plane pre-compression. Green load: Increasing in-plane tension

The objective of this research is to characterize and anticipate the effect of the lateral compression on the failure load of Li-ion battery cells. Whereas in previous works the specimens were exposed to biaxial stresses using perfect hemisphere punchers, the new system developed here enables full control of the compression load, and decouples the out-of-plain compression load and the in-plain tension load. Experiments were conducted on both single and multi-layer battery components.

Chapter 2 of this work describes the experimental apparatus, including the Tension-Compression System (TCS) that was designed. Considerations in design and manufacturing are presented, as well as alternatives.

Chapter 3 describes validation experiments that were conducted with a simplified TCS system, in order to endorse the selected design.

Chapter 4 describes the experiments that were conducted, specifies the results, and discusses them.

Chapter 5 summarizes this work and specifies the conclusions and chapter 6 is a reference list.
2. Experimental apparatus

2.1 Scheme of the experimental apparatus

Figure 3 depicts the experimental set up that was held in the ICL. The new Tension-Compression System (will be called TCS from now on) that was designed was integrated into INSTRON machine (Model 5944). The experimental procedure was defined as follows:

First, the TCS was installed in between the INSTRON testing table, and the INSTRON load cell (one time step). Section 2.2 will explain this in more detail. Second, a specimen was placed in the TCS, and a pre-compression load was applied on it. Third, the INSTRON machine was activated for vertical movement to incrementally increase the measured tension load vs. displacement until failure.

For each set of components and pre-compression load, the above procedure was repeated several times in order to validate coherent results. Then, the process was repeated for different pre-compression loads, and results were compared.

*Figure 3: Schematic drawing of the experimental apparatus*
2.2 Zero and Non-Zero Pre-Compression Relation

One hypothesis of this research is that the relation between the in-plane tensile load and the displacement is preserved for different out-of-plane pre-compression loads. We assume that different pre-compression loads will only result in different maximum tensile loads before failure. Hence, we expect that the curves for different compressions will follow the same trend, as illustrated by the qualitative graph in Figure 4.

![Figure 4: qualitative illustration of the effect of compression load on maximum tensile load](image)

2.3 Tension Compression System (TCS)

As mentioned above, in most previous studies, researchers used hemisphere punchers. This technique forces coupling between the tensile and compression loads, since one load magnitude effects on the other. The main consideration that was taken into account in the planning and design process of the new TCS was to allow decoupling between in-plane and out of plane loads.

Two design concepts which meet this principal were evaluated: The first concept was to use a pulley system for tension, and weights for uniform pre-compression as shown in Figure 5. This system is simple and easy to control and manufacture, but coerces two significant liabilities: First, the friction loses of the pulleys and the weight-segment interface might reduce the measurement accuracy of the maximum tensile load. Second, the loads
needed to be applied in order to initiate compression stress were very large: For example, pre-compression stress of 10 MPa, which is relatively low, will require approximately 100kg weight for a 10X10 mm segment.

<table>
<thead>
<tr>
<th>Item #</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Weight</td>
</tr>
<tr>
<td>2</td>
<td>Segment</td>
</tr>
<tr>
<td>3</td>
<td>Segment holder</td>
</tr>
<tr>
<td>4</td>
<td>Slider</td>
</tr>
<tr>
<td>5</td>
<td>Rail</td>
</tr>
<tr>
<td>6</td>
<td>Pulley</td>
</tr>
<tr>
<td>7</td>
<td>Pulley</td>
</tr>
<tr>
<td>8</td>
<td>Mounting plate</td>
</tr>
</tbody>
</table>

*Figure 5: 1st concept for Tension Compression System (TCS)*

The second concept was to create a system assembled of two mechanisms: one fixture (static) that applies the pre-compression load using a compression spring and a second fixture (dynamic) that mounts the specimen. The latter is connected to the INSTRON load cell and moves with it. This way, while the TCS controls the static, vertical compression load, the INSTRON machine applies the in-plane tensile load. Figure 6 presents the compression (left) and tension (right) mechanisms, which share the same mounting circular plate. Figure 7 depicts the detailed design of the TCS (left) and the set up as was finally assembled (right).
Figure 6: Second concept: left is the compression system using a compression spring. The segment is located between the two rods. Right is the moving part which is connected to the INSTRON and mounting the segment.

Figure 7: Second design concept: left (a) design drawings, right (b) full set up as built in the ICL. Upper part is connected to INSTRON load cell, bottom part is connected to INSTRON table.

The pre-compression load is applied by pressing two parts against each other: the top part with conical geometry (part #005 in Figure 7a) and the bottom part, the puncher (part #012), which is replaceable. The pre-compression is applied by rotating the two M6 nuts that compress the spring. The pre-compression load is measured using a 500N load cell located
below part #010. Shafts (part #008) guided by linear bearings were added to ensure alignment in the vertical direction. The complete, detailed drawings of the TCS are presented in Appendix I. This second design successfully overcomes the friction loses that the first design suffers from, and thanks to the addition of the load cell enables a more accurate measurement of the compression load, up to the level of the load cell error. In addition, the presence of a spring in the system allows a uniform distribution of the load along the specimen- another advantage over the first option.

In order to ratify the selected design concept, a preliminary experiment was conducted. The details of this experiment as well as its successful results are presented in the next chapter (chapter 3) of this work. Finally, the designed TCS was manufactured at MIT Machine Shop and was successfully implemented at the lab. We will further discuss the attributes of the designed system on the next chapters of this work.

3. Design Validation Experiment

In order to validate the design concept before heading to manufacturing, a simplified, preliminary experiment was conducted. The purpose of this experiment was to insure that the selected design allows us to investigate the particular, required load case. This will be the case if the failure occurs at the center of the specimen, inside the punched area- a thing which allows us to assume that the failure is affected by the compression load.

A second purpose of the validating experiment was to grossly evaluate the maximum pre-compression stress for which the failure would still occur in the compressed area of the specimen as oppose to a failure located at the boundary or outside of the boundary of the compressed area. In other words, we wanted to assess the maximum compression stress for which the design concept would still be valid.

Figure 8 demonstrates failure in and out of the punched area of a tri-layer separator (PE/PP/PE): On the left there is a combination of inside and outside failure whereas on the right the failure occurred outside only.
Experimental procedure was as follows: The pre-compression load was applied by lowering the puncher using the ‘Down’ button on INSTRON the control panel until it reaches contact with the tested specimen, which is placed on a flat surface. After reaching contact, the toggle on the control panel was rotated down to increase very slowly the load until the desired pre-compression is reached. The tensile load was grossly applied by hand moving of the segment mounting upward as shown in Figure 9. The experiment was conducted in two phases: In the first phase, existing punchers available at the ICL were used in order to apply the compression load. In the second phase, the same process was repeated, this time by using especially made punchers, which were later used in the designed TCS. Both parts were conducted with a 2KN load cell.

Figure 8: Separators after failure exposed to lateral compression and in-plane tension. At the left specimen where the failure is inside the compressed area combined with outside failure, at the right specimen where the failure occurred outside the punched area.

Figure 9: Experimental setup. After reaching to required compression load, the tensile load is applied by hand.
In the first phase, two types of separator were used: Isotropic (ceramic) and anisotropic tri-layer (PE/PP/PE). Later on, throughout the second phase, only the anisotropic type was used. All specimens were 44.45mm in diameter (1¾”) and were cut using a round knife of this geometry. Additionally, in all experiments, a Teflon film was placed between the specimen and the puncher to reduce friction effects.

3.1 Phase I of validation experiments: Using existing punchers

Experiment #1: A 1.5mm diameter puncher (cone shape) with sharp edges was used, and the separator was mounted between two flat plates attached to each other by four screws as shown in Figure 10. The specimen was an isotropic (ceramic) separator. The first step was to apply pre-compression load of 500N. The result was failure before applying the tensile force. We concluded that the sharp edges of the puncher combined with a relatively small diameter are not suitable for this kind of load case.

![Experimental setup for 1.5mm flat puncher with sharp edges and 2 thin plates attached for specimen mounting. On the right is enlarged view on the puncher (qualitative geometrical relations)](image)

Experiment #2: The same separator and flat mounting technique as in experiment #1 were used, this time with a flat 11mm diameter cylindrical puncher. The first load was of 1500N. This time there were no visual defects before applying tension, and we began increasing the in-plane load manually. The failure of the specimen occurred on the mounting boundary, which is, of course, not the desired failure mode. Reducing the load to 1000N and
500N did not change the failure mode. We assumed that the failure occurred on the mounting boundary because the mounting plates had created fixed boundary conditions, which compel stress concentrations at that point.

Experiment #3: we changed the mounting fixture to the one shown in Figure 11. This kind of mounting was already used before in [6] for testing specimens with perfectly hemispheric punchers (as oppose to flattened hemisphere punchers which were used so far in the current research). The rounded protrusion close to the inner diameter is designed to eliminate the fixed boundary condition problem which we encountered in experiment #2.

![Figure 11: Experiment #3: mounting with rounded protrusion](image)

The first pre-compression load in this experiment was 1500N: As expected, the failure did not occur at the mounting boundary, but on the cylindrical puncher edge as shown in Figure 12. Nonetheless, this failure was still not the kind we were aiming for since it is still outside of the compressed area. The compression load was reduced to 1000N, 500N, and finally zero, but the failure mode did not change. We assumed that this behavior is due to the rough, manually operation and will be solved by the fine control that the TCS will allow.
Experiment #4: Same setup as in Experiment #3, except for the specimen, which was replaced from isotropic (ceramic) to anisotropic (PE/PP/PE). At first, 500N pre-compression load was applied. The result was a combination of out of center failure due to boundary effects and around center material flow which suggests that the failure is due to the compression. Reducing the load to 200N and later to zero resulted in center-located failure mode with almost no influence from the boundary effect. Figure 13 depicts the specimen after the 200N failure. Indeed, the development of the desired failure mode confirmed that the tension-compression load case is feasible at the lab and can be investigated by the designed apparatus.
It is important to mention at this point that since the tensile load was applied manually, the errors are relatively high. Hence, no quantitative discussion is conducted yet.

3.2 Phase II of validation experiments: Using costume-made punchers

The next step was to conduct experiments using costume-made punchers. The phase II experimental set-up was similar to that of phase I, with the mounting method as shown in Figure 11. The only exception was the puncher type, which was replaced with the one presented in Figure 14. In order to reach a wide range of stresses (at least tens of MPa), two punchers were manufactured with different flattened area: 6mm and 3mm diameter. For this validation phase, only the anisotropic (PE/PP/PE) separator was used.

![Figure 14: hemispheric puncher with a flattened tip: 3 and 6 mm diameter](image)

Experiment #5: Here, the 6 mm puncher was used. This puncher allows enlarging the pre-compression load by smaller increments compared to the 3mm puncher. First, the pre-compression load was raised from 15N to 125N, in 10N increments. On all 12 cases the specimen failed, and showed a combination of both in- and out-of-compression-area rupture. Next, the compression load was raised to 250N and to 350N and the result did not change. Figure 15 depicts 7 specimens out of the 14. The last experiment (350N) is equivalent to 12.4MPa, which is relatively low.

Experiment #6: The puncher was replaced by the one with 3mm diameter. Similarly to the previous case, pre-compression load was raised in very small increments, with no significant change in results. Transition developed after 525N compression load, which is equivalent to 74.3MPa. At the next load, which was 650N (~92MPa), the failure mode
changed, and the specimen rupture was observed to be outside of the compression area. Figure 16 depicts tested specimens after failure.

3.3 Conclusion

In conclusion, the design validation experiment achieved its purpose. It demonstrated both the feasibility of the experimental method and its suitability for the required load case investigation. It also allowed us to get a sense of the compression load for which our method will not suit anymore. In terms of the experimental method, the validation experiment emphasized the fact that a more delicate, fine control on the tensile stress was required, and therefore paved the way for manufacturing our designed TCS.

Figure 15: phase II of validation experiments. Anisotropic separators after failure using 6mm puncher. The red color was used to view more clearly the failure area.
Figure 16: phase II of validation experiments. Anisotropic separators after failure using 3mm puncher. For 650N the failure mode was already outside of the compression area.
4. Results

4.1 Zero load on anisotropic separator

After completing the validation experiment, the TCS was manufactured by the MIT Machine Shop and was fully integrated with the INSTRON machine. For the first set of experiments the anisotropic separator was used. The execution of this set of experiments was also a trial-and-error process which was used to study the advantages of the new TCS.

As mentioned in the previous chapter, specimens failed both at their center and off-center for anisotropic separator. The next series of experiments was aimed to better understand the two phenomena and decouple them. It is the major part of the experimental work that was done in this research. Sufficient database was collected by using several types of punchers in several different component combinations. For convenience, experiments were designated by serial numbers, and results are presented graphically, with puncher drawing next to the graph.

Experiment #7: Beginning with the widest hemisphere puncher (20mm diameter) combined with zero load we found that the failure occurs outside of the flat area, i.e. not in the future to be compressed region. As seen in previous works [9], the direction of the failure was perpendicular to the machine direction of the specimen. More experiments were conducted with narrower punchers (12.7mm and 6.35mm) trying to reduce the contact between the uncompressed area of the specimen and the puncher, but the failure always appeared off the flat area.

Figure 17 shows a specimen moments before failure. The transparent areas are those that exhibit the largest stresses. Figure 18 depicts the results for two identical samples: The blue curve demonstrates one-phase failure, in which fracture was developed in a single location on the specimen, whereas the red curve demonstrates a two-phase failure, where the specimen developed two fractions, one after the other. Specimens failed in both manners, with no special preference. Figure 19 depicts two of the specimens, where the off-center location of the failure is clearly seen.

The driving phenomenon which initiates this off-center failure is probably the friction on the curvature of the puncher. In an attempt to reduce this friction, a Teflon sheet was applied in between the separator and the steel punchers, but the result remained unchanged.
Figure 17: Experiment #7: single layer anisotropic separator right before failure, 12.7mm puncher

Figure 18: Experiment #7: single layer anisotropic separator for zero compression load, 20mm diameter puncher

Figure 19: Experiment #7: Off center Failure. Left: 20mm hemisphere, Right: 6.35mm hemisphere.
4.2 Zero load on isotropic separator

Unlike the anisotropic separator, the isotropic one is expected to have no failure sequence: The failure occurs at ones without any directional preference [9]. In addition, some preliminary trial-and-error attempts that we conducted at the lab convinced us that an isotropic specimen is more likely to develop the failure at the center.

As described in chapter 3, throughout the design validation experiment all isotropic specimens had demonstrated off-center failure. Nevertheless, we anticipated that by the presence of the TCS in the system, a significant amount of inaccuracies that existed in the validation experiment will be eliminated, and this may settle the failure in different location on the specimen.

Experiment #8: Same set-up as in Experiment #7 was used, this time with a 3mm flat surface diameter puncher (1/2" hemisphere). Figure 20 depicts three Force-displacement curves, for three identical specimens that were examined. Except for a slight change in the maximum load before failure, results are quite consistent.

On all three cases, failure occurred on the edge of the flat area, in the future-to-be compressed part. This implies that the TCS has indeed improved the accuracy, as the failure is now located closer to the center. However, the on-edge location suggests the existence of new conditions, which coerce lack of material flow at the center of the specimen and by that, prevent development of the desired failure mode. In order to further study this phenomena, we continued to the next experiment, with non-zero load on an isotropic specimen.
4.3 Pre-compression on isotropic separator

Experiment #9: Pre-compression load was added to the system, and the same 3mm puncher was used. The load was applied on the flat surface of the puncher against another flat surface (larger), with the specimen mounted between the two, as shown in Figure 21.
4 different loads were applied, equivalent to stresses of 10, 30, 50 and 60MPa as calculated in Table 1. For each load, the experimental procedure was repeated several times. Results were proved to be coherent, and hence every load in the force-displacement graph in Figure 22 is presented by a single, typical curve. As can be clearly seen in the figure, the maximum failure load was decreased as the pre-compression load was increased. For the 10MPa pre-compression, there is no significant change in failure load compared to the zero load. On most cases, a single-phase failure was observed, but there was also a small number of two-phase failures, such as the orange curve in Figure 22.

<table>
<thead>
<tr>
<th>Desired compression stress $\sigma_c$ [Mpa]</th>
<th>$F = \sigma_{comp}A_c$ [N]</th>
</tr>
</thead>
<tbody>
<tr>
<td>10</td>
<td>71</td>
</tr>
<tr>
<td>30</td>
<td>212</td>
</tr>
<tr>
<td>50</td>
<td>353</td>
</tr>
<tr>
<td>60</td>
<td>424</td>
</tr>
</tbody>
</table>

Table 1: Required load for desired pre-compression stress

Figure 22: Force vs. Displacement for various pre-compression stress on isotropic separator specimen
On all cases, and in consistency with Experiment #8, failure was located on the edge of the compression area and not at the center. Nonetheless, as well demonstrated by the graph in Figure 22 the separator was proved to be sensitive to changes in pre-compression and there was a definite connection between the in-plane failure load and the out-of-plane compression load. This indicates that the new boundary conditions created by the compression do not fully prevent material flow in the specimen.

We concluded this part of the work by suggesting that although the specimen doesn't develop the desired failure mode, the current setup enables the establishment of a relation between the compression load and the maximum failure stress.

4.4 Multi-Layer Experiments

4.4.1 Zero load on multi-layer specimen

So far, all experiments were conducted with a single layer separator. The current part of this work will focus on multi-layer specimens. Figure 23 depicts the set of layers constituting the specimen. This specific order of layers was selected as it corresponds with simulations developed in [8].

![Separator](image1)

![Separator](image2)

![Separator](image3)

Figure 23: layers order in the multi-layer specimen

Experiment #10: First, the multi-layer specimen was mounted in the same apparatus as in Experiment #9, combined with no compression load. Figure 24 presents force-displacement curves for 4 identical specimens. Results are coherent, as the curves follow similar trend.

Throughout the measurements, procedure was stopped several times, immediately after a failure was observed, and the layers were separated in order to better characterize the failure sequence. On all cases, the first layer to fail was the cathode, and this event is reflected by the first peak drop in Figure 24, at around 20N.
The final peak drop in the graph, around 60-70N, is a result of a separator-layer failure. Other peak drops were found to be related to either a single-layer failure or a secondary failure, which is an additional fracture developed in a layer that already encountered failure. Section 4.4.3 will discuss the failure sequence in more detail.

![Force-Displacement curve multi-layer without pre-compression](image)

*Figure 24: Force-Displacement curve multi-layer without pre-compression. The picture on the left is a typical type of failure of the four layers. The order (bottom-up) is in the same order as in the fixture*

Figure 25 depicts the 4 layers of a specimen after the experiment. As before, specimens failed at the flat area, on the edge of the soon-to-be compressed part. Our assumption regarding this observation will be discussed in chapter 4.4.2, where pre-compression load is added to the multi-layer measurements.

![Multi-layer specimen after failure](image)

*Figure 25: A typical look of the multi-layer specimen after multi-layer failure. From left to right: Anode (bottom), intermediate separator, cathode, separator (top)*
Experiment #11: Same experiment as #10 was conducted, only this time with a narrower puncher (same flat surface diameter, 1/4" hemisphere). The results for two identical specimens are presented by the red and green curves in Figure 26, and show great similarity. A third, blue curve which represents the results of Experiment #10 was added to the same graph for comparison. The curves of the two punchers display a significant difference, as the narrower one led to larger failure loads than the round puncher. This shows that the puncher geometry plays a great role in determining the exact nature of the failure and therefore a rigorous experimental procedure should comprise that into considerations.

![Force-Displacement curve multi-layer without pre-compression](image)

**Figure 26: Force-Displacement curve multi-layer without pre-compression**

4.4.2 Pre-Compression on multi-layer specimen

Experiment #12: In the same manner as in the single-layer case, the zero-compression multi-layer experiment was followed by non-zero compression measurements. The set-up was identical to the one in Experiment #11, with the narrow puncher comprised in it. Specimens were subjected to pre-compression loads of 10, 20 and 30 MPa and as before, some measurements were stopped immediately after a failure in order to better observe the layers.
Figure 27 depicts a representative series of measurements for 4 identical specimens, under 4 different loads. Similarly to the single-layer case, larger pre-compression loads led to smaller maximum failure load. The four curves show resemblance which coincides with our basic hypothesis that the trend of the curve does not change as a function of the pre-compression load.

### 4.4.3 Failure Sequence of Multi-Layer set

An important issue to be addressed is the failure sequence in the multi-layer set. In order to collect sufficient data for this part, we repeated experiments #11 and #12 many times, with identical isotropic specimens. As before, the process was stopped several times immediately after a load-drop and the specimen was spread into single layers for observation. Our aim was to characterize the failure sequence, and check if it is changed for zero and non-zero pre-compression loads, as was demonstrated in the simulations in [8].

On all measurements it was found that the first layer to fail was the cathode. This was also the case for other pre-compression loads, as mentioned in 4.4.1. The distinct peak-drop related to this failure is the first one seen in Figure 28. The cathode's
fracture was observed to be more brittle compared to others, and was followed by additional, secondary fractures on this layer.

The second layer that failed was the anode. The exact point for which this failure occurs was less easy to track, and therefore it is not as distinctive as the previous one. The suggested failure range is encircled in Figure 28. The 4 curves in this figure represent 4 measurements with 4 identical specimens. The process was stopped several times between the cathode failure and the circled area in an attempt to locate the emergence of fracture, but by the time the specimen was separated to layers, the fracture on the anode was already well developed. Therefore, our conclusion is that the failure of the anode starts immediately after the cathode’s failure.

An important observation is related to the failure of the separators. As described in section 4.4.1, the final peak drop seen in the graphs (Figure 27) is related to the failure of one of the separators. Aiming to better understand if there’s any preference in the sequence of failure, we compared the results for zero load (Figure 28) and 10 MPa (Figure 29) pre-compression loads. In the case of zero-load, the upper separator failed before the inner one and in the case of 10 Mpa, the inner one failed first. Repeated measurements confirmed this result, and this suggests that the failure sequence is changed with pre-compression load.

It is useful to analyze our results more carefully and to compare them to the simulation developed in [8]. We will start by discussing the zero-load case, where our observations had shown that the cathode failed first and the anode failed shortly after. This is in agreement with the simulative results, as can be learned from case ‘b’ on figure 6 of [8]. However, in the second half of the sequence, the simulation had shown that the inner separator failed first, and this result differ from the one observed in our experiment. It is possible that the difference in the case of zero load is due to imperfect resemblance in experimental conditions. By improving the similarity between experiment and simulation, the results may be more similar. Ways to create this improvements are suggested in chapter 6 of this work, which describe potential continuation of this research.

Moving forward to the non-zero load case, we can see that there is an agreement in failure sequence between the experimental results and case “e” on figure 6 in [8]:
first the cathode, then the anode, third is the inner separator and last is the upper separator. This resemblance was confirmed by repeated experiments.

In conclusion, we can assume that there is a difference in failure sequence between the zero and non-zero case.

![Figure 28: Determining the failure sequence for zero pre-compression](image)
4.4.4 11 layers test

So far, our specimens resembled a simple, basic, battery cell, with one anode and one cathode. Real life battery is composed of a large number of layers, and therefore there is an interest in studying a specimen with a repeated, identical, structure. Experiment #13: In the same set up as the last one, we used an 11-layer specimen (layers were identical to those that were used before) with no compression load. A representative curve of the experiments, and a spread of the layers after failure are given in Figure 30.
Figure 30: 11 layers zero load test. The order of layers shown on the left is in the same order as in the fixture.

One important observation is related to the location of the failure. We anticipated that in an 11-layer structure, the inner layers, which are located further from the mounting surfaces, might fail closer to the specimen's center. As can be seen on the left side of Figure 30, our results misalign with this hypothesis, since all the layers failed, as before, on the edge of the future to be compressed area. This implies that the conditions created by the puncher geometry are likely to affect the specimen through all layers. One possible way to reduce this effect is suggested in chapter 6.

The failure sequence was not studied for this set of layers. Nonetheless, it was easy to observe that like in the 4-layer case, the separators failed last, beginning at about 180N tension load for the first one to fail. Another interesting phenomenon was that whereas the 11 layers specimen contained 6 separators, the graph exhibits only 4 drops, which implies that some of the separators failed simultaneously.
4.5 Using Foam in Compression Area

In pursue of a more center-oriented failure, we continued with a new set up design, intended to induce the development of this failure mode. First, foam was added to the compression interface, between the specimen and the upper surface, as seen in Figure 31. The assumption was that by pressing the foam, its center will develop the largest stresses, and this will function as a catalyst to developing maximum stress at the center of the specimen. This way, there is a greater chance of failure at the center. In order to further encourage the development of this desired failure mode, the puncher was replaced by a perfect hemisphere puncher, made of Teflon. Figure 31 and Figure 32 present the new set up components. Figure 32 shows how the foam was squeezed during compression.

Figure 31: compression area - new setup

Figure 32: left - foam and hemisphere before compression. Right - Foam is compressed and squeezed - highest stress at the center to encourage failure strain.
Several types of foam were examined, which are mainly differed by the level of hardness. They are presented in Table 2.

<table>
<thead>
<tr>
<th>Level of hardness</th>
<th>Pressure range [psi]</th>
<th>Pressure to compress foam by 25% [psi]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Soft</td>
<td>9-14</td>
<td>12</td>
</tr>
<tr>
<td>Medium</td>
<td>15-30</td>
<td>18</td>
</tr>
<tr>
<td>Hard</td>
<td>30-60</td>
<td>45</td>
</tr>
</tbody>
</table>

*Table 2: hardness level of foam*

Experiment #14: In the new set-up, 5 identical specimens were tested. Specimens were single layered and isotropic, similar to those used in the first part of this work. 3 specimens were tested under zero load and 2 were tested under 70N load. On all 5 measurements, a medium hardness foam was used. Results are given in Figure 33.

*Figure 33: hemisphere Teflon puncher – Comparison between zero load and 70N load using medium hardness foam*
From this set of measurements we can deduce that increasing the compression does not necessarily decrease the failure load. This stands in disagreement with our basic hypothesis. Additionally, the failure location was varied from one measurement to the other. For example, in the case of zero load, one failure occurred far off center, whereas another occurred near center. The different curves, though, still exhibit the same trend, which implies that the stress distribution near the center and out of the center are similar.

Experiment #15: Additional set of measurements with Teflon hemisphere puncher was performed using the other two type of foams, soft (12psi) and hard (45psi). 4 identical, single layered isotropic specimens were tested, all under 40N load. Results are shown in Figure 34 and show that there is no significant difference between the two types of foam in terms of the maximum failure load. Here, like in Experiment #14, there was no consistency in failure location and yet, the curves kept similar trend.

![Figure 34: hemisphere Teflon puncher – Comparison between two different foams under the same pre-compression](image)

To conclude, using the new set up with the foam did not encourage development of the desired failure mode. Other solution will be offered in chapter 6.
5. Summary and Conclusions

- The study described in this document was aimed to characterize the effect of pre-compression on the failure load of Li-ion battery segments. Towards achieving this goal, few concepts were considered, and eventually a new system was designed and manufactured. The new system enables applying out-of-plane constant compression and in-plane increasing displacement simultaneously, with fine control, and with no coupling between the two, and by that allows us to study the effect of different compression loads on battery segments. The system was successfully integrated with the INSTRON machine at the ICL and was fruitfully used in a variety of experiments.

- Validation experiments, that were meant to validate the TCS design were probably over simplified and demonstrated the evolution of mixed failure modes- both in- and out-of-the compressed area. However, the author assures that achieving the desired failure mode with the existing TCS is only a matter of time, and by conducting a large number measurements, using different geometrical interfaces, this goal will be achieved.

- The second part of the experimental work was focused on single-layer specimens, combined with several types of punchers. Isotropic specimens were found to demonstrate more consistent measurements than anisotropic specimens, and therefore were used from this point forth. Measurements using flat surface steel punchers indicated an observable decrease in the tensile failure load for increasing pre-compression load. Same was found in the next part of our work, for multi-layer specimens.

- For the multi-layer specimens, the failure sequence was studied. It was found that the sequence may alter for different pre-compression loads. Nevertheless, in all experiments, the cathode failed first, and the anode failed second in gradual process, as demonstrated in figure 34 in [5].

- Throughout this work, different methods were used in an attempt to encourage development of the desired failure mode, at the center of the specimen. Several geometrical interfaces were examined and attempts of using foam on the specimen were made, but with minor or no success.

- Even though the desired failure mode did not develop, we could notice a distinct correlation between the compression load and the failure load. Hence, we feel certain to
conclude that the failure is a direct outcome of the compression. Yet, we suggest a hypothesis which might explain the development of an off-center failure: The artificial fixed edge of the specimen, which does not allow material flow between the compressed surface and the pure-tension area, create new boundary conditions. These may contribute a minor compression even under zero compression load, or can affect the specimen in a manner similar to compression. Additionally, it was seen that most failures were located on the interface between the compressed area and the pure tensioned area, probably because of high stress concentrations in this interface.

- As mentioned at the beginning of this paper, one basic hypothesis was laid at the base of this work: The relation between the tensile load and the in-plane displacement does not change for different out-of-plane compression loads. Our experiments confirmed that this hypothesis holds very well, both for single and multi-layer specimens.
6. Future Work

- At the end of this work, we would like to recommend several improvements, which we believe are likely to improve the results.

- Design improvements: Each measurement that involved pre-compression load required setting of the upper compression surface in the middle of the specimen. This was done manually and, as a result, experiments were repeated many times in order to get sufficient number of coherent results. To overcome this issue, we suggest adding a linear bearing in the compression system that will compel identical, precise location every time. Drawings are presented in Appendix II. The changes involve replacing two parts (#004 and #005) and adding the mentioned linear bearing.

- Compression interface: Another design for the compression interface is suggested in Appendix IV. The geometry of the new interface has a relatively large curvature (the largest we had so far) in order to create a gradual slope change that may prevent the emerging of an off center failure mode.

- Once the desired failure mode is achieved, the next step is to build a representative volume element model subjected to the specific geometric interface and run numerical simulations in order to validate the experimental results by using a different calculation method.
7. References


[9] Xiaowei Zhang1, Elham Sahraei1,2 & Kai Wang1, Li-ion Battery Separators, Mechanical Integrity and Failure Mechanisms Leading to Soft and Hard Internal Shorts, Cambridge: Scientific Reports, 2016.


Appendix I – Drawings of Tension Compression System
moving element
assembly
SECTION A-A
SCALE 1:1

SECTION B-B
SCALE 1:1

DETAIL C
SCALE 2:1

upper gripper

mild steel
shaft will be supplied

DETAIL A
SCALE 5:1

guide
SECTION A-A

SECTION B-B

editted dimension will be determined according to load cell diameter
Appendix II – Design Improvements for the TCS system
Part #004:
Part #005

DETAIL C
SCALE 2:1

SECTION A-A

DETAIL B
SCALE 5:1

SOLIDWORKS Student Edition
For Academic Use Only.

005_cone part for 0.5" rod

A4
Appendix III – Plate Strength Calculation

The main strength consideration in the design process is the bending and stress concentration of the plate that counteract the spring, i.e. drawing #004 in

Appendix I – Drawings of Tension Compression System

To simplify the calculation we will model the plate as a simple beam:

The spring load will be converted into point load, and the boundary conditions are simply supported:

The Maximum bending moment is at the center and equal to [10]:

\[ M_{\text{max}} = \frac{FL}{4} \]

And stress:

\[ \sigma_{\text{max}} = \max \left( \frac{M_y}{I} \right) \]

The minimum 2nd moment of inertia \( I \) is also at the center, and the distance from the neutral axis \( y \) is constant:
\[ I_{\text{min}} = \frac{(38.1 - 18.5)h^3}{12} \]

\[ y = \frac{h}{2} \]

\[ \frac{I_{\text{min}}}{y} = 3.27h^2 \]

F is the maximum compression force of the spring and is around 500N. The length between the supports: 120mm.

\[ \sigma_{\text{max}} = \frac{15,000}{3.27h^2} \approx \frac{4590}{h^2} \]

The hole at the center creates stress concentrations of factor 3:

\[ \sigma_{\text{max}} = \frac{\sigma_y}{3} \approx \frac{200}{3} \approx 67\text{MPa} \]

Hence, the minimum thickness of the plate is

\[ h_{\text{min}} = \sqrt{\frac{4590}{67}} = 8.3\text{mm} \]
Appendix IV – New Contact Interface Suggestion

- Zoom in on assembly with suggested new contact:
Drawings puncher and top surface:
SECTION A-A
SCALE 5:1