Combined Tensile-Compressive Biaxial Loading of Li-ion Battery Components

by

Nathaniel J. Byrd

B.S., Mechanical Engineering, North Carolina State University, 2007

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

Naval Engineer

and

Master of Science in Mechanical Engineering

at the

Massachusetts Institute of Technology
June 2018

© Nathaniel J. Byrd, MMXVI. All rights reserved.
DISTRIBUTION A. Approved for public release: distribution unlimited.
The author hereby grants to MIT permission to reproduce and to distribute publicly paper
and electronic copies of this thesis document in whole or in part in any medium now
known or hereafter created.

Signature redacted

Signature of Author..........

Department of Mechanical Engineering
May 2018

Signature redacted

Certified by............... Tomsz Wierzbicki
Professor
Thesis Supervisor

Accepted by.......................................................... Rohan Abeyaratne
Chairman, Department Committee on Graduate Studies
Department of Mechanical Engineering
ABSTRACT

Lithium-ion batteries under a mechanical load can develop failures in the internal multi-layer structure. Due to the flammable materials required for construction, the safety of the battery has been in question since it was first developed. Internal failures can create a short circuit which may lead to thermal runaway, resulting in fire and sometimes the explosion of the battery. Due to the increasing use of lithium-ion batteries in military and consumer products, the number of incidents involving batteries has also risen and with it a growing concern for safety. The larger batteries required for unmanned vehicles and electric vehicles increase the potential for damage due to a battery fire aboard a ship or a battery damaged in a collision.

This thesis investigates the failure mechanism of internal lithium-ion battery components when subject to a constant out-of-plane compression while applying and increasing a decoupled in-plane tension until failure. This thesis also describes the methods used to optimize the mechanical system designed to apply constant compression while increasing tension. The results will be used to characterize and anticipate the effect of lateral compression on the failure load of lithium-ion battery cells. The existing microstructural based finite element model can be modified to the experimental conditions in this thesis in order to compare the experimental and modeled results. This comparison will be used to refine and validate the micro model and ultimately bring us closer to improving the design of lithium-ion batteries.

Experimental results showed there is a quantifiable relationship between the amount of pre-compression internal battery components are exposed to and the maximum failure load. In the tested 0 to 30 MPa compression range was a 40% reduction in fracture displacement and a 20% reduction in load displacement. The change in failure order, predicted by the micro model was validated by this experimental data. However, failure did not occur in the area of compression which indicates friction due to the compression had a significant effect on testing therefore a design to eliminate friction was proposed.

Thesis Supervisor: Tomasz Wierzbicki
Title: Professor of Applied Mechanics
ACKNOWLEDGEMENTS

This thesis was completed with the guidance and support of several individuals, whom I would like to thank:

The guidance, instruction, and support from the MIT 2N Naval Construction and Engineering program provided by Captain Joel Harbour, Commander Weston Grey, Commander Johnathan Page and Mary Mullowney.

Professor Tomasz Wierzbicki, the founder of the MIT Impact and Crashworthiness Lab, for his mentoring, guidance, and enthusiasm which were essential in this research. Dr. Elham Sahraei for her support and insightful guidance in this research. The members of the Impact and Crashworthiness Laboratory for their instruction and assistance.

Finally, I would like to thank my loving family, my wife Kasey and sons Oliver and Alexander, for the love, support, encouragement, and so much more during our time in Cambridge.

BIOGRAPHICAL NOTE

Lieutenant Commander Nathaniel Byrd is from Richmond, Virginia and graduated from North Carolina State University in 2007 with a Bachelor of Science in Mechanical Engineering. He was commissioned as a Surface Warfare Officer in the United States Navy and served tours on the guided-missile destroyer USS Paul Hamilton and, after completing nuclear power training, the aircraft carrier USS Theodore Roosevelt.

In 2014, LCDR Byrd transferred in the Engineering Duty Officer (Nuclear) community and reported to the Massachusetts Institute of Technology in May of 2015 to earn a Naval Engineer’s degree and a Master of Science in Mechanical Engineering. After graduation from MIT, LCDR Byrd will report to the Supervisor of Shipbuilding Newport News in Newport News, Virginia to work in the aircraft carrier refueling and complex overhaul project office.
TABLE OF CONTENTS

ABSTRACT ................................................................................................................................. 3

ACKNOWLEDGEMENTS ........................................................................................................... 5

BIOGRAPHICAL NOTE ............................................................................................................. 5

TABLE OF CONTENTS ............................................................................................................... 6

LIST OF FIGURES ..................................................................................................................... 9

LIST OF TABLES ....................................................................................................................... 10

LIST OF EQUATIONS ............................................................................................................... 10

LIST OF ACRONYMS AND ABBREVIATIONS ......................................................................... 11

UNITS OF MEASUREMENT ...................................................................................................... 11

1. INTRODUCTION ................................................................................................................... 13
   1.1 Commercial Industry ....................................................................................................... 14
      1.1.1 Rapid Growth ........................................................................................................... 14
      1.1.2 Battery Incidents .................................................................................................... 16
      1.1.3 Policy and Regulations ......................................................................................... 18
   1.2 Department of Defense ................................................................................................... 18
      1.2.1 Planned Growth ........................................................................................................ 18
      1.2.2 DoD Policy ............................................................................................................. 21
      1.2.3 DoD Battery Incidents .......................................................................................... 22
   1.3 Problem Statement .......................................................................................................... 23

2. LITERATURE REVIEW ......................................................................................................... 24
   2.1 Lithium-Ion Battery Basics ............................................................................................ 24
      2.1.1 Construction and Operation .................................................................................... 24
      2.1.2 Lithium-ion Battery Properties ............................................................................... 26
      2.1.3 Safety Concerns ...................................................................................................... 27
   2.2 Impact Crashworthiness Lab at MIT ............................................................................. 28
      2.2.1 Progression of Research ....................................................................................... 28

3. TENSION COMPRESSION SYSTEM TEST DESCRIPTION ................................................. 29
   3.1 Test Equipment and Preparation .................................................................................... 29
      3.1.1 ICL Equipment ....................................................................................................... 29
      3.1.2 Specimen Geometry and Preparation ................................................................... 31
   3.2 Material Properties and Selection .................................................................................. 32
      3.2.1 Separator Selection ............................................................................................... 32
LIST OF FIGURES

Figure 1-1: Evolution of Rechargeable Batteries to Higher Energy Density [2] ......................................................... 14
Figure 1-2: Lithium-Ion Battery Price 2010-16 [3] ........................................................................................................ 15
Figure 1-3: Evolution of Global Electric Car Stock, 2010-2016 [4] .............................................................................. 16
Figure 1-4: Chevrolet Volt Fire [5] ............................................................................................................................. 16
Figure 1-5: 787 Fire in APU (left), APU Post Fire Comparison (right) [8] ................................................................. 17
Figure 1-6: Desired Capability Advancements to Support Unmanned Vehicles [13] ................................................... 19
Figure 1-7: DoD Roadmap for Unmanned Maritime Systems (2013-2038) [13] ........................................................ 20
Figure 1-8: DoD Roadmap for Unmanned Aircraft Systems (2013-2038) [13] ......................................................... 20
Figure 1-9: DoD Roadmap for Unmanned Ground Systems (2013-2038) [13] .......................................................... 21
Figure 1-10: NASA Spacesuit Li-ion Battery [8] ......................................................................................................... 21
Figure 1-11: ADS Mini-sub Damaged and Retired due to Li-ion Battery Fire [8] ....................................................... 22
Figure 1-12: Army BB-2590/U Li-ion Battery and PP-8489 Portable Charger [8] ....................................................... 23
Figure 2-1: Cross Sections of Li-ion Cells [19] ........................................................................................................... 25
Figure 2-2: Li-ion Battery Charge Discharge Cycle [21] .......................................................................................... 26
Figure 3-1: INSTRON Machine (left) with TCS Installed (right) ............................................................................... 30
Figure 3-2: Multi-Layered Specimen .......................................................................................................................... 31
Figure 3-3: Circular Specimen Geometry .................................................................................................................... 31
Figure 3-4: Cutting Template (left) and Tapered Specimen (right) ........................................................................ 32
Figure 3-5: (a) Illustration of MD, TD, +DD, -DD a separator roll; nominal stress-strain of ................................ 34
Figure 3-6: Force-displacement curves for biaxial punch test using 1", ½", ¼", & ⅛" diameter punches .................... 35
Figure 3-7: Compression test stress-strain curve (courtesy of Xiaowei Zhang [24]) .............................................. 36
Figure 3-8: Tension Mechanism ............................................................................................................................... 38
Figure 3-9: Compression Mechanism ........................................................................................................................ 39
Figure 4-1: Clamp Ring Material Discrepancy ............................................................................................................ 41
Figure 4-2: Specimen Interaction with Clamp Ring ................................................................................................. 41
Figure 4-3: Clamp Ring Lip (left) & Machined (right) .............................................................................................. 42
Figure 4-4: Fixed Compression Area .......................................................................................................................... 43
Figure 4-5: Polymer Punch Tips (left) and Polymer Sheet Cut to Specimen Size (right) .................................... 44
Figure 4-6: Foam Types Tested .................................................................................................................................. 46
Figure 5-1: Separator Load-Displacement Graph .................................................................................................... 48
Figure 5-2: Maximum Load and Displacement of Separator at Failure ................................................................. 48
Figure 5-3: Average Peak Load (top) and Displacement (bottom) at Different Levels of Pre-compression ........ 49
Figure 5-4: Typical Failures Observed in Separator ............................................................................................... 50
Figure 5-5: Tapered Multi-layer Load-Displacement Graph .................................................................................... 51
Figure 5-6: Failures of the Tapered Multi-layer Samples .......................................................................................... 52
Figure 5-7: Trial Circular Multi-layer Sample Load-Displacement Graph ........................................................... 54
Figure 5-8: Failures of Circular Multi-layered Sample ............................................................................................. 54
Figure 5-9: INSTRON and Bluehill© Output for Circular Multi-layer Sample Load-Displacement Graph ......... 56
Figure 5-10: Combined and Color Coded Circular Multi-layer Sample Load-Displacement Graph ............... 57
Figure 5-11: Maximum Load-displacement of Circular Multi-layer Samples at Failure .................................. 57
Figure 5-12: Average Peak Load (top) and Displacement (bottom) at Different Levels of Pre-compression .... 58
LIST OF TABLES
Table 3-1: TCS Strain Gage Panel Meter 500 N Load Cell Calibration..........................................................30
Table 3-2: Material Properties of Separators .................................................................................................32
Table 3-3: Material Properties of the Anode and Cathode .............................................................................37
Table 4-1: Punch Tips Tested.........................................................................................................................43

LIST OF EQUATIONS
Equation 1: Li-ion Chemical Reaction........................................................................................................26
Equation 2: Chemical Reaction Occurring at the Anode .............................................................................26
Equation 3: Chemical Reaction Occurring at the Cathode ..........................................................................26
Equation 4: Force conversion for compression ...........................................................................................30
Equation 5: Compression Area.....................................................................................................................39
Equation 6: Compression Spring Force .......................................................................................................39
LIST OF ACRONYMS AND ABBREVIATIONS

Al – Aluminum
APU – Auxiliary Power Unit
ARL – Army Research Laboratory
ASDS – Advanced Seal Delivery System
BEV – Battery Electric Vehicle
Cu – Copper
DoD – Department of Defense
DON – Department of the Navy
DLA – Defense Logistics Agency
EV – Electric Vehicle
FAA – Federal Aviation Administration
Li – Lithium
Li-ion – Lithium-ion
NDS – National Defense Stockpile
OPNAVINST – Office of the Chief of Naval Operations Instruction
PE – Polyethylene
PHEV – Plug-in Hybrid Electric Vehicle
PP – Polypropylene

UNITS OF MEASUREMENT

kWh – kilowatt hour
N – newton
min – minute
mm – millimeter
MPa – mega pascal
mV – millivolt
Whkg$^{-1}$ – watt hour per kilogram
Whl$^{-1}$ – watt hour per liter
1. INTRODUCTION

When selecting a thesis topic, my goal was to make a contribution towards an area of research that has applications in the U.S. Navy and the potential for the betterment and advancement of humanity. The Impact Crashworthiness Laboratory at MIT is focused on improving the safety of Lithium-ion batteries mainly for electric vehicle applications but I believe the end result satisfies both my personal and professional thesis criteria.

I have spent almost half of my life in the U.S Navy and am intimately familiar with the limited space, weight restrictions, and power limits faced when deploying and conducting operations on a ship. The benefits of a safe, lightweight, high power, rechargeable storage unit are too many to list. To give an example, the designs as well as operational capabilities of unmanned and autonomous vehicles not only benefit from the improvement but require power storage units such as the lithium-ion battery.

During my time deployed overseas, I realized how dependent we are on oil when a portion of my life was spent defending the Khawr al Amiyah and al-Bashrah oil terminals in the Persian Gulf, more commonly referred to as the KAAOT and ABOT oil platforms. Improving the lithium-ion battery is not going to reduce the world’s dependence on oil but replacing small engines with lithium-ion batteries is a step in shifting to a cleaner energy source.

Lastly, I have always been fascinated with space, and might be stretching a little to connect my research with space travel. However, it’s really not that far off, as lithium-ion batteries are already essential for power storage on the International Space Station and smaller versions are used to power NASA space suits during space walks.

I realize my thesis work is only a very small piece but according to Sir Francis Bacon, the father of the scientific method, progress is achieved through many small advances which accumulate to produce large leaps in science and technology [1] and this is my contribution.
1.1 Commercial Industry

Research and development for lithium-ion batteries began in the 1970's and less than 2 decades later lithium-ion batteries were available for commercial use. The potential applications for both Department of Defense (DoD) and consumer products were recognized immediately which started the integration into electronic designs. The initial integration was slow at first due to the high cost; when first introduced a lithium-ion battery cost about $3,000 per kWh which was about 20 times the cost of using a lead acid battery at $150 per kWh. However, a lighter and smaller lithium-ion battery could provide the same amount of power, Figure 1-1, making them ideal for portable and personal electronics driving further research and development that continues today [1].

![Figure 1-1: Evolution of Rechargeable Batteries to Higher Energy Density](image)

1.1.1 Rapid Growth

Lithium-ion batteries provide a compact high power source that has resulted in rapid growth of electric vehicles and small consumer electronic products such as cell phones, tablets, and laptops. As the demand for lithium-ion batteries continues to grow improvements are made to the material
supply chain, chemical composition, manufacturing process, price competition, and learning curve all resulting in reducing the cost per kWh, Figure 1-2. This allowed personal electronic devices to become part of everyday life and the lithium-ion batteries that power them.

These improvements have made large batteries affordable for consumers allowing the market for electric vehicles to expand. The Tesla model S and X demonstrated the capability of electric vehicles exceeded most expectations. This along with the movement to reduce carbon emission levels allowed the electric vehicles to gain more of the market share. Now most major automotive manufacturers are currently producing at least one version of electric vehicle. Sales of electric vehicles are growing exponentially each year in many countries and the global stock of hybrid and electric vehicles passed 1 million in 2015 and 2 million in 2016 [4], Figure 1-3. The use of lithium-ion batteries in consumer products continue to grow and larger, more powerful batteries are becoming more and more common.
1.1.2 Battery Incidents

In 2012 the U.S. Department of Transportation conducted an investigation on an incident involving the destruction of a 2011 Chevrolet Volt and three nearby vehicles, Figure 1-4. The vehicle was used for safety testing 3 weeks before the incident and no damage was noted to the battery. The investigation determined that the fire originated from the Volt’s Li-ion battery due to damage sustained during the testing [5].
Several other electric vehicle fires have occurred due to damage of the Li-ion battery and casing, including Tesla. In 2013 a Tesla Model S ran over a metal object creating significant force that punctured the quarter inch armor plate and the Li-ion battery. Designed safety features minimized the extent of the incident and include: an onboard alert system which directed the owner to stop and exit the vehicle, internal firewalls to minimize spreading, and vents which directed the flames toward the road [6].

The Federal Aviation Administration (FAA) Office of Security and Hazardous Material Safety maintains records involving “Aviation Cargo and Passenger Baggage Events Involving Smoke, Fire, Extreme Heat or Explosion Involving Lithium Batteries”. They currently have 191 incidents that include brief descriptions dating from March of 1991 to January 2018 [7]. These air travel and transit incidents have not been limited to passenger or cargo items, in 2013 and 2014 three separate incidents occurred involving the auxiliary power unit on the 787 planes, two on the ground in Boston, Massachusetts and Narita, Japan and another in flight Takamatsu, Japan. This resulted in the fleet being grounded for over 3 months while a world-wide investigation occurred. The National Transportation Safety Board determined the probable cause was an internal short that developed due to mechanical stresses. Thermal runaway allowed the failure to spread to multiple cells but the fires were contained in the units. This did result in the redesign of the auxiliary power unit (APU) and additional FAA certification process requirements[8].

Figure 1-5: 787 Fire in APU (left), APU Post Fire Comparison (right) [8]
1.1.3 Policy and Regulations

The number of incidents involving Li-ion batteries grows with number of Li-ion batteries in use and will lead to increased policy and regulations. The safety of EVs are getting a lot of attention and manufacturers are incorporating many safety features to produce a safe consumer product. The Department of Transportation is investigating vehicle and battery design and many additional elements including: new firefighting requirements, wrecked vehicle transportation and disposal, and even minimum noise for pedestrian awareness [9]. The FAA has restrictions in place for personal electronic device Li-ion batteries as well as large shipments of Li-ion battery cargo [10]. These policies and restrictions placed on Li-ion batteries increase overall safety of the products but could negatively impact the Li-ion battery market. On the other hand increasing the safety of Li-ion batteries by improving internal structure, optimizing casing, battery placement, and incorporating safety features like firewalls, temperature monitoring, and directed ventilation would reduce regulations and improve consumer confidence in the safety of Li-ion batteries.

1.2 Department of Defense

1.2.1 Planned Growth

The impact the Li-ion battery could have on different missions and operating environments of organizations within the Department of Defense (DoD) was recognized early in development. The light weight, compact, and high power density characteristics which drove success in the commercial industry also led to the integration of Li-ion batteries into many existing systems. The military applications have a broad range and provide energy storage and power for missiles, lasers, tactical vehicles, unmanned vehicles, and military personal electronic devices such as portable reconnaissance and communication equipment. The advantages of the compact size and reduced weight are magnified when operating in the air, on the ocean surface, under water, or launching the equipment into space, Figure 1-10, since adding either weight or size comes with a significant cost [8].

To operate in marine environments where vessels have very limited cargo space, weight is restricted by displacement, and increasing size comes with a significant hydrodynamic resistance penalty, all require a battery with advantages that Li-ion batteries offer. This is why the Department of the Navy (DON) began testing lithium-ion batteries in the late 1970’s and today the Office of Naval Research continues to focus on understanding the failure modes of lithium-ion batteries.
The Army Research Laboratory continues to enhance the capability of lithium-ion batteries. Recently, their research with the University of Maryland developed an aqueous lithium-ion battery with a higher power density comparable to non-aqueous lithium batteries [11]. Over the last few years the Defense Logistics Agency (DLA) has recognized the growing importance of lithium and has listed it as a strategic material and determined a minimum amount to maintain in the National Defense Stockpile [12].

Advancements in Li-ion batteries and several other areas of technology have led to the recent focus on unmanned and autonomous vehicles and systems. In 2013 the President’s Budget request for developing, testing, and evaluating unmanned vehicles and systems was $5.6 billion over 5 years [13]. The Unmanned Systems Integrated Roadmap (2013) describes the DoD’s plan for developing and integrating unmanned systems into operations from 2013 through 2038. An essential part of each phase of the plan are the improvements of energy storage and specifically, “Lithium-Chemistry Batteries”, shown in the highlighted portions of Figure 1-6. The many different types of systems, current, and potential applications for marine, air, ground, and space systems can be seen in Figure 1-7, Figure 1-8, Figure 1-9, Figure 1-10 respectively.

![Figure 1-6: Desired Capability Advancements to Support Unmanned Vehicles][13]
Figure 1-7: DoD Roadmap for Unmanned Maritime Systems (2013-2038) [13]

Figure 1-8: DoD Roadmap for Unmanned Aircraft Systems (2013-2038) [13]
1.2.2 DoD Policy

Department of Defense has strict guidelines for the responsible use of Li-ion batteries on ships since a fire on a vessel when submerged or at sea cannot only impact operations but risks the safety of equipment, the vessel, sailors, and their lives. The Navy established the Lithium Battery Safety Program in 1982 in response to several incidents the DON experienced during early research.
involving Li-ion batteries including a fatality [14]–[16]. The program provides detailed guidance to obtain approval to use or transport a specific Li-ion battery in a specified system. The Naval Sea Systems Command (NAVSEA) instruction states, “the purpose of the manual is to establish safety guidelines for the selection, design, testing, evaluation, use, packaging, storage, transportation and disposal of lithium batteries” [15]. The tests required for approval include:

- Short Circuit
- High-rate overcharge/discharge
- High temperature abuse: external heating, and internal heating
- Physical: nail penetration, crush, and impact

Due to the risks associated with Li-ion batteries the thorough testing and certification requirements limits the use of equipment with Li-ion power supplies for the DON as well as DoD.

1.2.3 DoD Battery Incidents

Even with strict certification requirements incident involving Li-ion batteries occur and each slows the integration of Li-ion batteries into more systems. In 2008 an Advance Seal Delivery System (ASDS) was connected to charging its Li-ion batteries at a charging station when a fire occurred. Severe damage occurred estimated over $200 million and SOCOM, Special Operations Command, determined it would be more cost effective to retire the mini-sub [8]. The ASDS hull is now used as a shipyard mini-sub trainer at Pearl Harbor [17].

Figure 1–11: ASDS Mini-sub Damaged and Retired due to Li-ion Battery Fire [8]
The Army uses BB-2590/U Li-ion batteries with the PP-8489/U portable charger for many different applications and all batteries must pass testing established by the Army. Three fires have occurred involving these using these units in 2010, 2011, and 2015 [8].

1.3 Problem Statement
Lithium-ion batteries under a mechanical load can develop failures in the internal multi-layer structure. Due to the flammable materials required for construction the safety of the battery has been in question since it was first developed. Internal failures can create a short circuit which may lead to thermal runaway, resulting in fire and sometimes the explosion of the battery. Due to the increasing use of Li-ion batteries in military and consumer products, the number of incidents involving batteries has risen and with it a growing concern for safety. Larger batteries required for unmanned and electric vehicles increase the risk associated with storing batteries onboard naval ships and severity of damage from a collision involving an electric vehicle.

In 2009 the Impact and Crashworthiness Lab (ICL) was established at MIT to study Li-ion batteries in order to produce a computational model and improve safety. One study in 2016 investigated the failure mechanisms leading to internal short circuits under different loading scenarios. From this
a microstructural based model was developed to predict load-displacement and failure of internal components under crushing loads with different ratios of tension and compression [18].

This thesis investigates the failure mechanism of internal Li-ion battery components when subject to a constant out-of-plane compression while increasing in-plane tension to the point of failure. This constant compression models the internal pressure a battery may be subject to due to certain designs, operating environments such as the ocean, or damage to the external protective casing. This thesis will also demonstrate the methods used to optimize a mechanical system designed to apply constant compression while increasing tension. The results will be used to characterize and anticipate the effect of lateral compression on the failure load of lithium-ion battery cells. The existing micro model can then be modified to the experimental conditions in this thesis in order to compare the experimental and modeled results. This comparison will be used to refine and validate the micro model and ultimately bring us closer to improving the design of Li-ion batteries.

2. LITERATURE REVIEW

The literature review provides the background information necessary to understand the experimental setup, material selection, and design. This section will introduce the components, operation, advantages, and safety concerns of the lithium-ion battery. Then provide details on the Impact and Crash Worthiness Laboratory at MIT including; the objective, research areas, and the progression of research that led to this thesis.

2.1 Lithium-Ion Battery Basics

2.1.1 Construction and Operation

All battery cells store chemical energy and convert it into electrical energy require four basic components: the anode, cathode, separator, and electrolyte. A battery consists of one or more cells and cells come a variety of shapes and may have a hard casing or soft pouch outer casing. The battery internal components described below are arranged in a multi-layered sample with 4 layers: separator, Al with double-sided coating of active material (cathode), separator, and Cu with double-sided coating of graphite (anode) which can be rolled to form the cylindrical cell in Figure 2-1 (a) or can cell (c) [19].
(+) Cathode - a lithium metal oxide coating such as cobalt oxide (CoO₂) on an aluminum (Al) conductor as the positive (+) electrode that is reduced by acquiring electrons

(-) Anode – carbon or graphite (C₆) on a copper (Cu) conductor as the negative electrode that is oxidized by releasing electrons

Electrolyte - a variation of Lithium (Li) salt dissolved in an organic solvent acts as a medium allowing the ions to move between anode and cathode

Separator – a porous polymer provides a physical barrier to prevent contact between anode and cathode while allowing Li-ions to pass through

Discharging a battery describes the conversion of stored chemical energy to electrical energy, shown in Figure 2-2 and governed by reaction in Equation 1. When discharging, the right arrows, Li deintercalates freeing itself from the graphite becoming a (+) Li-ion and releases an (-) electron, Equation 2. The anode’s conducting Cu electrode is connected through external circuitry providing the path for electrical energy to flow and provide power. The Li-ion travels in the electrolyte and passes through the porous separator to the cathode then intercalates into the metal oxide absorbing an electron in the process, Equation 3. To charge the battery power from external circuitry is
required and reverse occurs, following the left arrows. Li deintercalates from the cathode, Equation 3, causing the metal to oxidize producing a (+) Li-ion and an (-) electron. The electron travels from cathode to the anode via external circuitry. The (+) Li-ion travels in the electrolyte through the separator to intercalate into the graphite, Equation 2, at the anode [20].

Equation 1: Li-ion Chemical Reaction

\[ \text{LiC}_6 + \text{CoO}_2 \rightleftharpoons \text{C}_6 + \text{LiCoO}_2 \]

Equation 2: Chemical Reaction Occurring at the Anode

\[ \text{LiC}_6 \rightleftharpoons \text{C}_6 + \text{Li}^+ + \text{e}^- \]

Equation 3: Chemical Reaction Occurring at the Cathode

\[ \text{CoO}_2 + \text{Li}^+ + \text{e}^- \rightleftharpoons \text{LiCoO}_2 \]

Figure 2-2: Li-ion Battery Charge Discharge Cycle [21]

2.1.2 Lithium-ion Battery Properties

This section will describe properties of Li-ion batteries both advantageous and those that bring up safety concerns [20], [22]. This focus is on secondary battery cells, meaning they can be recharged
and reused a finite number of times as opposed to primary battery cell which can only be used once.

**More affordable:** The cost to manufacture a battery, shown in Figure 1-2.

**Fast charge rate:** The rate the battery can undergo the chemical reactions to store energy, usually described by the time it takes to fully charge a “dead” battery.

**Long cycle life:** The number of times a battery can be recharged.

**High energy density:** The amount of energy stored in the battery. For a certain type of battery specific energy is normally used to describe and compare the amount of energy a battery can hold, Figure 1-1.

**Wide operating range:** The range of temperatures the chemical reaction will occur so the battery can be used in.

**High power density:** The amount of power that can be drawn from a battery. For a certain type of battery specific power is normally used to describe and compare the amount of current a battery can supply.

**Growing safety concern:** Failures of the battery which can result in harm of personnel or equipment. Recently, Li-ion batteries have been experiencing failures resulting in a growing concern for safety.

**Low self-discharge:** Describes the rate a battery undergoes the chemical reactions at the anode and cathode without an external electrical connection. Meaning the energy stored dissipates over time providing no useful power or work. Li-ion batteries have low self-discharge compared to other types.

**Long shelf life:** The length of time a battery can be stored (years) and later used. Li-ion batteries have a long shelf life.

**Low Toxicity:** Standards are in place to protect people and the environment, the reason nickel cadmium batteries are no longer used.

**No Maintenance:** Some chemical compositions require filling electrolytic solution and venting gasses from the battery.

### 2.1.3 Safety Concerns

Except for safety, all the characteristics have resulted in the rapid growth of the Li-ion battery market which in turn has led to an increasing number of incidents that has shifted the focus to safety. Two main components tied to safety are the battery casing protects from external factors and the separator provides protection internally. To improve safety the different methods of
failures of Li-ion batteries resulting in fire or explosions are being investigated. These failures occur mainly due to manufacturing defects, operational heating, mechanical abuse, electrical abuse, or thermal abuse. Safety features have been and are continuing to be developed to prevent or minimize the occurrence of these types of failures. Over charging can produce a combustible gas to prevent this circuitry was developed and integrated into many battery systems. When a battery cell is damaged it may develop a short circuit producing heat which can ignite the flammable electrolytic solution in the cell. Even when the damage is minimal, the heat developed could cause further damage to the cell feeding on itself. This is known as thermal runaway and will produce a larger short and more heat which can spread in the form of heat or fire to other cells and lead to the combustion and sometimes explosion of the battery [22], [23].

2.2 Impact Crashworthiness Lab at MIT

Instead of focusing on improving performance, Professor Wierzbicki’s Impact Crashworthiness Lab (ICL) at MIT conducts research in several areas, one which focuses on modelling failure mechanics of Li-ion batteries. This is one of the few academic groups working to model battery cells, others include Princeton, University of Michigan, and Michigan State University.

2.2.1 Progression of Research

Several studies conducted at the ICL have led to selecting the research for this thesis. The external and internal battery components were studied to determine characteristics [24], [25]. Tests were then conducted on both dry cells and actual battery cells to determine failure mechanics. These results were used to build a micro-structural computational model for certain battery cell types. Further testing was conducted to improve and validate the results of the model. Next, compression was added to the model to simulate an area of the battery casing damage which caused localized internal out-of-plane compression [18]. This compression was proportional to the amount of in-plane tension applied and the modelled results were validated by experimental results. The next step is the subject of this thesis, to decouple the out-of-plane compression and the in-plane tension to further develop the micro model. A test device was designed but limited testing was conducted and significant problems were encountered [26]. This thesis will also investigate the issues with the current system to optimize the device and experimental technique to improve results.
3. TENSION COMPRESSION SYSTEM TEST DESCRIPTION

This section introduces the equipment used, material tested, and describes the mechanics of operation. A general description of specimen construction and preparation is included and the reasoning behind the different geometries is discussed in Section 4.

3.1 Test Equipment and Preparation

This section describes the equipment used to conduct the combined tensile-compressive biaxial loading testing and how the specimens were assembled to represent dry internal components of LI-ion battery cells. All testing was conducted at ICL using installed commercial machines fitted with equipment designed and manufactured at MIT.

3.1.1 ICL Equipment

All testing used the Tension Compression System (TCS), designed by Sagy Hakoon with guidance from Professor Tomasz Wierzbicki and Dr. Elham Sahraci, to apply a constant, static compressive load while allowing tension to increase \[26\]. The TCS is mounted to the base or lower testing area of the INSTRON machine, model 5944, and connects to the upper portion of the INSTRON using an adapter, designed by Xiaowei Zhang \[24\]. This allows the INSTRON to control vertical displacement of the samples installed in the TSC, Figure 3-1 shows the INSTRON without and with the TCS installed.
Separate load cells were used to provide accurate real time measurements of both the compressive and tensile loads. The constant compressive force was measured with a 500 N load cell installed in the base of the TCS. The load cell was connected to DP25-S Strain Gage Panel Meter and calibrated in accordance the user manuals [27], [28] to 0.25 newton per unit, shown in Table 3-1 and Equation 4.

<table>
<thead>
<tr>
<th>Input Unit</th>
<th>1</th>
<th>mV</th>
</tr>
</thead>
<tbody>
<tr>
<td>Unit Screen Resolution</td>
<td>100</td>
<td></td>
</tr>
<tr>
<td>Converting to force [N/unit shown]</td>
<td>0.25</td>
<td>N/unit</td>
</tr>
<tr>
<td>Conversion for 100 Units</td>
<td>25</td>
<td>N/100 units</td>
</tr>
</tbody>
</table>

**Equation 4: Force conversion for compression**

\[
\text{Force [N]} = (\text{meter reading in [mV]}) \times (0.25 \text{ [N/mV]})
\]

The INSTRON was equipped with a 2 kN load cell that connected and moved vertically with the adapter connecting to the TCS and measured the changing in-plane tensile force. The INSTRON
machine was connected to a computer that used Bluehill© software to measure, record and plot the force, displacement, and record the time for each.

3.1.2 Specimen Geometry and Preparation
Specimen tested consisted of single and multi-layered samples to model the internal components of a Li-ion battery. The main internal battery component that prevents short circuits is the separator which made up the single layer specimen and was mainly used for the earlier tests while optimizing the equipment configuration and test conditions. Next, multi-layered samples, shown in Figure 3-2, were used and layered to model the internals of a Li-ion battery cell as shown in Figure 2-1: Cross Sections of Li-ion Cells [19].

![Figure 3-2: Multi-Layered Specimen](image)

The circular sections were placed between two sheets of paper and cut with a circular cutting punch to a diameter of 44.45 mm. The specimen were inserted into a clamp ring for testing leaving a 30 mm inner diameter circular section for testing, Figure 3-3. The clamp ring removed the cut edge from the test.

![Figure 3-3: Circular Specimen Geometry](image)
Another sample geometry tested was made using the circular cutting punch with three cuts to produce a tapered specimen, shown in Figure 3-4. The major disadvantages of the tapered samples were they took longer to make and introduced the cut tapered edges into the test. Before testing each edge was visually inspected and samples with noticeable damage along the exposed edge were removed and discarded.

![Figure 3-4: Cutting Template (left) and Tapered Specimen (right)](image)

### 3.2 Material Properties and Selection

This section covers the properties of the materials tested and the method for selecting the type of separator.

#### 3.2.1 Separator Selection

Several different types of separators were considered for testing, these were purchased from MTI Corp, have been used in previous research, all are currently used in Li-ion batteries, and have properties listed in Table 3-2.

<table>
<thead>
<tr>
<th>Graph Reference</th>
<th>Type</th>
<th>Manufacturing Process</th>
<th>Thickness (µm)</th>
<th>Porosity (%)</th>
<th>Pore Size (µm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>b</td>
<td>PP</td>
<td>Dry</td>
<td>25</td>
<td>36-46%</td>
<td>0.01-0.1</td>
</tr>
<tr>
<td>c</td>
<td>PP-PE-PP</td>
<td>Dry</td>
<td>25</td>
<td>39%</td>
<td>0.5-0.2</td>
</tr>
<tr>
<td>d</td>
<td>Al2O3-PE-Al2O3</td>
<td>Wet</td>
<td>16 (2,12,2)</td>
<td>37%</td>
<td>0.1</td>
</tr>
<tr>
<td>e</td>
<td>Non-woven</td>
<td>E-spinning</td>
<td>31</td>
<td>46%</td>
<td>0.2</td>
</tr>
</tbody>
</table>

Table 3-2: Material Properties of Separators
The separator will be subject to a combined tension and compression in a biaxial punch test so the selection was based on the tensile tests, biaxial punch tests, and compression tests previously conducted at the ICL by Xiaowei Zhang. The results of the uniaxial tensile test produced the stress strain curves shown in Figure 3-5 [24]. The polypropylene (PP) in (b) and polyethylene (PE) used in PP-PE-PP, the tri-layer in (c) have very anisotropic properties so failure would occur much earlier in any direction other than MD (a). These separators would introduce another variable of alignment in the biaxial test. The Al₂O₃-PE-Al₂O₃, ceramic coated in (d) and the non-woven in (e) have close to isotropic properties and failure occurs in roughly the same stress range. The non-woven is very weak so the ceramic coated was selected for the higher, approximately 5 times the stress strain failure levels and for the isotropic properties better suited for biaxial testing [24].
Figure 3-5: (a) Illustration of MD, TD, +DD, -DD a separator roll; nominal stress-strain of (b) dry-processed PP separator, (b) dry-processed tri-layer separator, (c) wet-processed ceramic-coated separator, (e) nonwoven separator in the four directions (courtesy of Xiaowei Zhang [24])
The results of the biaxial punch test are shown in Figure 3-6 [24], and were used to select the separator. The PP and PE-PP-PE have two modes of failure, shown in (b) and (c), where the ceramic in (d) and non-woven in (e) have a single mode of failure. This is due to the isotropic properties resulting from the manufacturing and the Al_2O_3-PE-Al_2O_3, ceramic coated separator was selected due to the single mode of failure as well as the increased strength compared to non-woven.

**Figure 3-6:** Force-displacement curves for biaxial punch test using 1", ½", ¼", & ⅛" diameter punches (b) dry-processed PP separator, (b) dry-processed tri-layer separator, (c) wet-processed ceramic-coated separator, (e) nonwoven separator (courtesy of Xiaowei Zhang [24])
The results of the compression test are shown in Figure 3-7 [24] and were used to select the separator. The compression ranges being considered for the testing were from 0 to 50 MPa. The PP, PE, and tri-layer had the best stress-strain characteristics but the ceramic coated was selected based on tensile test and punch test results. Of note, the ceramic coated separator has an almost linear relationship in the ranges of compression tested.

![Figure 3-7: Compression test stress-strain curve (courtesy of Xiaowei Zhang [24])](image)

3.2.2 Anode and Cathode

Internal battery components were removed from dry cells to construct the multi-layered specimen. The anode and cathode have a metal foil center and are coated as described in Section 2.1.1 and have properties listed in Table 3-3.
Table 3-3: Material Properties of the Anode and Cathode

<table>
<thead>
<tr>
<th>Internal component</th>
<th>Mass Density (p) [kg/m³]</th>
<th>Young's Modulus (E) [GPa]</th>
<th>Poisson's Ratio (v)</th>
<th>Thickness [µm]</th>
<th>Combined Thickness [µm]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Anode Coating</td>
<td>879</td>
<td>2.5</td>
<td>0.010</td>
<td>63.5[1]</td>
<td>141</td>
</tr>
<tr>
<td>Copper (anode)</td>
<td>8930</td>
<td>110</td>
<td>0.343</td>
<td>14</td>
<td></td>
</tr>
<tr>
<td>Cathode Coating</td>
<td>2162</td>
<td>5.9</td>
<td>0.010</td>
<td>54[1]</td>
<td>128</td>
</tr>
<tr>
<td>Aluminum (cathode)</td>
<td>2680</td>
<td>385</td>
<td>0.330</td>
<td>20</td>
<td></td>
</tr>
</tbody>
</table>

Note: [1] Each side of the anode and cathode are coated, this is thickness per side
[2] Average values are provided

3.3 Mechanics of Operation

This section describes the mechanism of operation that allow the Tension Compression System (TCS), designed by Sagy Hakoon with guidance from Professor Tomasz Wierzbicki and Dr. Elham Sahraei, to apply constant in-plane compression that is decoupled from an increasing out-of-plane tension [26].

3.3.1 Tension Mechanism

The tension mechanism is a slight modification to a proven method that has been used to apply tension in the radial direction in previous research conducted at the ICL [24]–[26]. The specimen is held in a clamp ring which is lowered at a fixed rate onto a stationary punch while the time, load, and displacement are measured and recorded, as seen in Figure 3-8. These measurements are based on input from the INSTRON’s 2 kN load cell to the Bluehill© software. The punch tips can be changed and allows for experimentation with different diameter, material, and compression areas.
3.3.2 Compression Mechanism

To add a constant in-plane compression that is decoupled from the tension mechanism the TCS uses compression spring. The compression spring provides a relatively constant compression through its own tip to the specimen which is located between the spring tip and punch tip. This compression force is measured using the additional load cell located in the base of the TCS. In the cross section view at bottom you see the battery specimen is placed in the clamp ring on top of the
punch. The force is applied by compressing the spring and measured with the load cell located beneath the punch. The required force for the desired compression is calculated based on the area of the flat portion of the punch, Equation 5, then using the strain meter conversion, Equation 4, and adjusting force the compression spring to the desired compression, Equation 6.

![Compression Mechanism Diagram](image)

**Figure 3-9: Compression Mechanism**

**Equation 5: Compression Area**

\[
A_{compression} = \frac{\pi \times \text{diameter}^2}{4}
\]

**Equation 6: Compression Spring Force**

\[
F = \sigma_{compression} A_{compression} \text{[N]}
\]
3.3.3 Basic Procedure for the TCS
To simultaneously apply the decoupled tension and compression requires the equipment is set-up in a very specific order. Appendix A) describes the set up and operation of the TCS to aid in any future research involving the device.

3.4 Problems Encountered During Previous Testing
Previous testing did not produce the desired and hypothesized failure mode where the point of failure would be centered on the punch. Many of the failures occurred at the boundary of the clamp ring or outside the area of pre-compression. There was only a small amount of testing initially conducted with the TCS and the problems were not able to produce consistent expected results. Further testing was recommended to optimize experimental technique for more consistent results.

4. OPTIMIZATION OF EXPERIMENTAL TECHNIQUE
This section explores the experimental parameters varied in order to optimize the experiment for consistent repeatable results. To reduce time and material costs the initial optimization tests were conducted on the separator only samples. Once previous problems were corrected and consistent results were achieved the experiment was further optimized for the multi-layer samples.

4.1 Equipment and Design
The radial in-plane tension should be the highest at the center of the punch tip for any level of pre-compression. The samples were expected to fail on or near the center of the punch tip though in my initial testing many failed near the clamp ring boundary and this was independent of the level of pre-compression. This unexpected error had to be corrected prior to any optimization. The designer was contacted to ensure the TCS device was set up correctly and for any additional advice on optimization.

4.1.1 TCS Design
After running several tests and varying many different parameters there was no reduction in number of boundary failures. This indicated the unexpected failure location was the result of a design or manufacturing error. A thorough review of the CAD drawings were conducted to determine if the failure could be caused by a design error but nothing significant was noted. Next, the TCS components were examined and compared to the CAD drawings and the material
discrepancy was easily identified. The lip on the upper portion of the clamp ring had not been machined when manufactured, the lip is highlighted in green in Figure 4-1. As the clamp ring was lowered onto the punch, the rough edge would concentrate stress causing the failure of the specimen at the boundary, the specimen interaction with the clamp ring lip is shown in Figure 4-2 with the specimen added in red. This rough clamp ring lip and machined clamp ring lip are shown in Figure 4-3. Tests conducted after this correction moved the failure location closer to the expected and for no compression the failure occurred at the center of the punch tip.
4.1.2 Punch Tip Geometry

The TCS punch tip could be unscrewed to allow testing with different punch tip diameters, geometries, and materials which are listed in Table 4-1. The non-metal tips would deform slightly as the compressive force was increased and this produced the compression area. Small changes in the compression area significantly change the amount of force required to produce the desired level. This added another variable which could not easily be measure or accounted for. The metal punch tips with no fixed compression area experienced a similar effect though the compression areas produced using different types of foam to distribute the force.

Measuring and adjusting the force to apply a specific amount of pre-compression required a fixed area for the force to act on the specimen. Metal punch tips with small flattened areas were used to provide the fixed area for compression. Two different diameter compression areas, 6 mm and 3 mm, were used and the flat compression area is shown in Figure 4-4. The punches with the larger fixed compression area were not ideal for testing since the maximum compression that could be applied was much lower. Another issue was the compression spring acted on a tip which had a diameter of just over 7mm so ensuring proper alignment between the two tips was difficult with separator samples and would be even more difficult using multi-layer samples. Proper alignment means the upper, compression spring tip overlapped the lower, punch tip compression area on all sides to produce a uniform compression.

To ensure a repeatable test the metal punch tips with a 3mm fixed compressive area were used. The largest diameter radial punched were desired to produce in-plane radial tension. However, using the larger 1” diameter punches with separator only samples produced failure near the clamp.
ring boundary even after the surface was machined. This was due to the larger angle produced which can be seen in Figure 4-2, this was corrected by using the smaller ⅜” punch tip.

Table 4-1: Punch Tips Tested

<table>
<thead>
<tr>
<th>#</th>
<th>Material</th>
<th>Diameter [inches]</th>
<th>Compression Area Diameter [mm]</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>teflon</td>
<td>1</td>
<td>25.4 not fixed</td>
</tr>
<tr>
<td>2</td>
<td>teflon</td>
<td>3/4</td>
<td>19 not fixed</td>
</tr>
<tr>
<td>3</td>
<td>metal</td>
<td>1</td>
<td>25.4</td>
</tr>
<tr>
<td>4</td>
<td>metal</td>
<td>3/4</td>
<td>19 3</td>
</tr>
<tr>
<td>5</td>
<td>metal</td>
<td>1/2</td>
<td>12.5 3</td>
</tr>
<tr>
<td>6</td>
<td>metal</td>
<td>1</td>
<td>25.4 6</td>
</tr>
<tr>
<td>7</td>
<td>metal</td>
<td>3/4</td>
<td>19 6</td>
</tr>
<tr>
<td>8</td>
<td>metal</td>
<td>1/2</td>
<td>12.5 6</td>
</tr>
<tr>
<td>9</td>
<td>metal</td>
<td>1</td>
<td>25.4 not fixed</td>
</tr>
<tr>
<td>10</td>
<td>metal</td>
<td>3/4</td>
<td>19 not fixed</td>
</tr>
<tr>
<td>11</td>
<td>metal</td>
<td>1/2</td>
<td>12.5 not fixed</td>
</tr>
<tr>
<td>12</td>
<td>metal</td>
<td>1/4</td>
<td>6.4 not fixed</td>
</tr>
</tbody>
</table>

Figure 4-4: Fixed Compression Area
4.2 Friction

Friction is present and increases as the sample is lowered onto the punch and friction is also produced by the compression mechanism. This is a possible explanation for the failure occurring off center since the fracture locus will change depending on the friction coefficient. In order to reduce the friction coefficient different materials, lubrication, and contact interfaces were tested.

4.2.1 Material

A fluorocarbon polymer was used to reduce the friction coefficient, the punch tip and sheet are shown in Figure 4-5. First, it was incorporated into the punch tip but due to the compression of the punch tip described in Section 4.1.2 the tips could not be utilized for testing. In order to gain the reduced friction benefits of the polymer a thin sheet was tested in several locations. When placed between the punch tip and the sample, it would deform or fold as the sample was lowered onto the punch. To prevent this, the polymer sheet was cut to the sample size and tested in the clamp ring with the specimen. However, the polymer failed before the specimen so could not be incorporated into a multi-layer sample. The polymer was used to reduce the friction between the foam and the specimen tested which also provided the same contact surface with the specimen regardless of which foam was tested.

![Figure 4-5: Polymer Punch Tips (left) and Polymer Sheet Cut to Specimen Size (right)](image)

4.2.2 Lubricant

Lubricant was applied in several locations to reduce the friction coefficient. First, the grease was applied to the samples but the changes in sample properties depended on amount of grease, time
between application & testing, and spread area. These were difficult to control and experimentation variations did move the failure into the desired location. In order to reduce the friction while designing a repeatable experiment the grease was only used between the foam and the polymer sheet.

4.3 Additional Optimization Methods

4.3.1 Specimen Geometry
The tapered specimen described in Section 3.1.2 was designed to produce increase stress towards the center at the expected failure location. The smallest cross section is located in the center of the specimen and the intention was try to shift the failure into the compressed area. The tension produced is no longer radial but uniaxial in-plane tension. This type of geometry had increased labor costs, each tapered specimen requires 3 times as many cuts so a multi-layered sample would require 12 cuts. The circular geometry maintained the cut edge outside of the test boundary, however, the tapered geometry exposed two of the cut edges to the test. This introduced a new variable and required cleaner precise cuts to minimize the effect. Each specimen was visually inspected for damage and resulted in about 25% being discarded. Based on this, the tapered section was only tested using multi-layered samples with the experimental set up based on circular sample testing.

4.3.2 Force Distribution using Foam
Foam was incorporated into the test to evenly distribute the force between the compression spring tip and the punch tip to produce a uniform compression field. Several different types of foams were tested and a few examples are shown in Figure 4-6. Foam was also used to test the punch tips with no fixed compression area. This was designed to distribute the force over an area and produce a compression area with maximum compression at the center which gradually decreased as you moved outward in the radial direction. This was not used in the experiment since using the thicker foams to distribute the force created alignment issues between the compression spring tip and the punch tip. This also produced a varying force distribution and an unknown amount of pre-compression based on the foam used, size of the foam, alignment, and force applied.

A thin hard foam was used to minimize alignment issues between the two tips. For the final experiment setup the foam was placed in direct contact with the compression spring tip and
lubricant was applied to the bottom of the foam this was then positioned on the polymer sheet on top of the sample.

![Foam Types Tested](image)

**Figure 4-6: Foam Types Tested**

### 4.3.3 Rate of Displacement

Changing the rate the clamp ring was lowered onto the punch produced noticeable results; rates of 2, 4, and 6 mm per minute were tested. During initial testing, the rate of 4 mm/min was mainly used based on time to run a single test. The separator only tests allowed visual identification of the failure origin before the failed section propagated. However, the first part of multi-layer testing involved interrupting the test to identify which layer failed and where the failure initiated. The time it took to stop the test once a layer had failed allowed the fail to propagate which and made the origin of failure difficult or impossible to identify depending on the propagation path. In order to minimize the displacement occurring post failure, the rate was reduced by half to 2 mm/min.

### 5. Results

This section includes the results for the circular separator, tapered multi-layered, and circular multi-layered tests conducted after the experimental design parameters were determined as described in Section 4.
5.1 Circular Separator

5.1.1 Experiment Description
The separator only testing was conducted using the following: a circular cut ceramic coated separator, 20mm metal punch with compression area diameter of 3mm, hard foam to distribute the load, grease to reduce the coefficient of friction, and a displacement of 4mm/min. Three tests were conducted for each level of pre-compression with a range from zero to 20 MPa.

5.1.2 Data Collection
The raw data, experimental summary, and photographs are maintained at the ICL at MIT. The experimental summary for this test was included in Appendix B) as an example of the data collection. The Appendix was included to show how each test was tracked and it includes basic parameters, a table for individual tests, comments on specific tests, the photograph numbers associated with each test, generic plots the BlueHill© software produced from raw data, notes on the current test, and notes describing the future work.

5.1.3 Analysis
The load-displacement plot was color coded based on amount of pre-compression, Figure 5-1. Next, the raw data was used to determine the maximum load and displacement that occurred at failure, plotted in Figure 5-2 on two different scales. Finally, the maximums were averaged for each level of compression and plotted, Figure 5-3. There is a 15% reduction in load-displacement, based on the maximum extension which occurs at 0 MPA and the minimum extension which occurred at 20 MPa. There is a 15% reduction in fracture-displacement, based on the maximum load before failure at 0 MPa and 20 MPa. The failures occurred within 2mm of each other so the pre-compression has an effect on load-displacement. This slight trend in load-displacement could indicate the friction produced in the compression area is having a significant effect.
Failure of Ceramic Coated Separator due to In-Plane Tension & Pre-Compression
(Circular Specimen, 3/4 in Metal Punch, Hard Foam, and Grease)

**Figure 5-1:** Separator Load-Displacement Graph

Max Load of Ceramic Coated Separator due to In-Plane Tension & Pre-Compression
(Circular Specimen, 3/4 in Metal Punch)

**Figure 5-2:** Maximum Load and Displacement of Separator at Failure
Examples of typical failures of the separator are shown in Figure 5-4. With no pre-compression the separator normally fails dead center. Once compression is added the failure occurs near the compressed area but even with the optimizations the failure did not occur or propagate through the compressed area. In the example photograph from a 20 MPa test, the failure propagation appears to have been slowed or stopped by the compressed area.
5.2 Tapered Multi-layer Samples

5.2.1 Experiment Description

The tapered multi-layer testing was conducted using the following: tapered cut samples assembled as shown in Figure 3-2, 20mm metal punch with compression area diameter of 3mm, hard foam to distribute the load, grease to reduce the coefficient of friction, and a displacement of 4mm/min. Enough tapered layers were cut to run 3 tests at each level of pre-compression with a range from zero to 20 MPa. The initial run for each level of compression the test was interrupted to determine the failure order of the different layers.
5.2.2 Results
The tapered multi-layer load-displacement plot was color coded based on amount of pre-compression, Figure 5-5. With no pre-compression applied the failure order was cathode, anode, outer separator, and then the inner separator. When pre-compression was applied the failure order changed to cathode, anode, inner separator, and then outer separator. This shift in failure order is interesting since it indicates an interaction is occurring between the layers under compression which causes the inner separator to fail earlier when under compression. The different failure orders observed also confirm the results from the microstructural finite element model where the same order shift occurred when the sample was under in-plane compression, [18].

![Failure of Ceramic Coated Separator due to In-Plane Tension & Pre-Compression](image)

Failure of Ceramic Coated Separator due to In-Plane Tension & Pre-Compression
(Metal Punch. Hard 30-60 psi Foam. Cut Samples. and Grease)

5.2.3 Experiment Modifications
This was the first multi-layered as well as the first tapered set of experiments and additional modifications to the experiment were made based new problems. First, the failure of the anode and
cathode occurred at the boundary of the clamp ring, this can be seen in Figure 5-6. The top image from right to left shows the anode, inner separator, cathode, and outer separator, the circled portion shows where the cathode failed at the clamp ring boundary before the anode failed. Failure of the anode or cathode was observed in almost half the samples tested. For the separator only tests this was corrected by machining the contact surface and reducing the size of the punch tip as described in Section 4.1.2. Applying the same reasoning, the angle produced at the clamp ring boundary, Figure 4-2, was likely too large for the brittle material of the anode and cathode which caused the failure. Based on this testing, future testing used a reduced punch tip size of ½", down from ¾". This was also the first test using layers and since the sample could not be observed while testing the rate of displacement was too high. For future testing the displacement rate was reduced by half, from 4 mm/min to 2 mm/min, to reduce the damage occurring post failure until the test is stopped.

The bottom photographs in Figure 5-6 show typical failures of the multi-layered tapered samples. The tapered geometry did not shift the failure into the compressed area as intended, introduce a cut edge into the test, have increased production time and costs so they are not used in future testing.

![Figure 5-6: Failures of the Tapered Multi-layer Samples](image)
5.3 Trial Circular Multi-layered Sample Test

5.3.1 Experiment Description
The tapered multi-layer testing was conducted using the following: circular cut samples assembled as shown in Figure 3-2, 12.5mm metal punch with compression area diameter of 3mm, hard foam to distribute the load, grease to reduce the coefficient of friction, and a displacement of 2mm/min. The initial run for each level of compression the test was interrupted to determine the failure order of the different layers.

5.3.2 Results
The trial circular multi-layer load-displacement plot was color coded based on amount of pre-compression, Figure 5-7. With no pre-compression applied the failure order was cathode, anode, outer separator, and then the inner separator, shown in top of Figure 5-8. When pre-compression was applied the failure order changed to cathode, anode, inner separator, and then outer separator, shown in bottom of Figure 5-8. This shift in failure order is interesting since it indicates an interaction is occurring between the layers under compression which causes the inner separator to fail earlier when under compression. The same failure order and changes based on pre-compression was observed in the tapered multi-layer samples and also confirm the results from the microstructural finite element model where the same order shift occurred when the sample was under in-plane compression.
Failure of Ceramic Coated Separator due to In-Plane Tension & Pre-Compression
(Multi-Layer Circular Specimen, 1/2 in Metal Punch, Hard Foam, and Grease)

Figure 5-7: Trial Circular Multi-layer Sample Load-Displacement Graph

Figure 5-8: Failures of Circular Multi-layered Sample
5.3.3 Experiment Modifications
The adjustments made based on the first multi-layered tests worked well and no additional modification was required. No failures occurred at the ring clamp boundary after replacing the ¼” punch tip with the ½”. The slower 2mm/min displacement rate reduced the propagation post failure which allowed identification of failure point. An additional benefit of this was the failure of the cathode then anode are easily identified in Figure 5-7 compared to Figure 5-5.

5.4 Final Circular Multi-layered Sample Test

5.4.1 Experiment Description
The final circular multi-layer testing was conducted using the following: circular cut samples assembled as shown in Figure 3-2, 12.5mm metal punch with compression area diameter of 3mm, hard foam to distribute the load, grease to reduce the coefficient of friction, and a displacement of 2mm/min. An initial test with no compression was run, then a full set, 4 to 5 runs, was conducted at each compression level. The range for compression was increased to the maximum, 30 MPa, limited by the compression spring design. No test interruptions were required since the failure order was already determined.

5.4.2 Data Collection and Processing
The raw data, experimental summary, and photographs are maintained at the ICL at MIT. The Bluehill© software displays a graph from the INSTRON data and is updated in real time, these graphs are shown in Figure 5-9. The raw load-displacement from all 18 runs were plotted and color coded based on amount of pre-compression, Figure 5-10. Next, the maximum load and corresponding displacement were identified using the raw, this was plotted in Figure 5-11 using two different scales. Finally, the maximum loads were averaged for each level of compression and the same was done for the corresponding displacements then both were plotted in Figure 5-12.
Figure 5-9: INSTRON and Bluehill® Output for Circular Multi-layer Sample Load-Displacement Graph
Failure of Ceramic Coated Separator due to In-Plane Tension & Pre-Compression
(Multi-Layer Circular Specimen, 1/2 in Metal Punch, Hard Foam, and Grease)

Figure 5-10: Combined and Color Coded Circular Multi-layer Sample Load-Displacement Graph

Max Load at Failure of Ceramic Coated Separator
(Multi-Layer Circular Specimen, 1/2 in Metal Punch, Hard Foam, and Grease)

Figure 5-11: Maximum Load-displacement of Circular Multi-layer Samples at Failure
5.4.3 Analysis

The color coded load-displacement plots, Figure 5-10 and Figure 5-11, show a downward trend in both load and displacement as compression is increased. This is not as distinguishable with the lower levels of compression, 0 – 10 MPa. When failure load and corresponding displacements were averaged for each level of compression, Figure 5-12, there is a slight reduction in both as pre-compression is increased at low levels and then an almost linear relationship from 10 to 30 MPa. Over the entire range there was a 40% reduction in fracture-displacement and a 20% reduction in load-displacement. The failures were spread over a 4mm displacement of each other so the pre-compression has an effect on load-displacement. This slight trend in load-displacement could indicate the friction produced in the compression area is having a significant effect on testing.
6. SUMMARY AND CONCLUSIONS

This final section summarizes the process developed and optimized to use the TCS to simultaneously apply in-plane compression and out-of-plane tension to dry internal battery components used to investigate the failure mechanics of Li-ion batteries. Also included is a comparison between the experimental results and the micro-structural finite element model and a new equipment design that should be considered to apply simultaneous uncoupled tension and compression.

6.1 Objective

This research investigated the failure mechanism of internal lithium-ion battery components subject to a constant out-of-plane compression while increasing in-plane tension to the point of failure. This thesis built on research previously conducted by MIT's ICL and used the TCS which was designed and built the previous year but did not undergo significant testing. The TCS is a mechanical system used to apply constant compression while increasing tension until the point of failure. The results will be used to characterize and anticipate the effect of lateral compression on the failure load of lithium-ion battery cells. This will be done using a general comparison between this experimental data and the existing microstructural finite element model. The micro model can then be modified to the experimental conditions in this thesis in order to provide a numerical comparison between the experimental and modeled results. This comparison will be used to refine and validate the micro model and ultimately bring us closer to improving the design of Li-ion batteries.

6.2 TCS Experiment Optimization

This thesis presented the background and methods used to improve the mechanism, repeatability, and results of the TCS. The process for exploring various testing parameters to optimize the experimental technique were specific for the TCS. These changes to the device and experiment setup improved the repeatability and results of the experiment. However, even with the modifications the failure occurred outside of the compression area.
6.3 Conclusion

The experimental results showed there is a quantifiable relationship between the amount of pre-compression internal battery components are exposed to and the maximum failure load. In the tested 0 to 30 MPa compression range was a 40% reduction in fracture displacement and a 20% reduction in load displacement. However, the failure never occurred in the area of compression which indicates friction due to the compression had a significant effect on testing. Experimental techniques did not reduce the friction desired levels, therefore the TCS may not the best method to investigate the failure mechanism of constant internal battery compression subject to an in-plane biaxial or radial tension.

A general comparison between the experimental results and the micro-model was conducted. Both the tapered and circular multi-layered tests experience the change in failure order when subject to pre-compression. This was predicted by the micro model and validated by the experimental data. A numerical comparison may not be as beneficial since an unknown level of friction has been introduced to the experiment when applying the compression.

6.4 Future Work

To continue researching the failure mechanism and reduction in failure load due to an existing compression a new method for applying compression will be discussed. To remove the friction produced by the compression mechanism would require a pressurized chamber. The chamber would need to be large enough to house the punch and conduct the experiment in. The design of this machine would be more complicated but the MIT machine shop has the equipment required to manufacture a pressure chamber. This could be designed for a specific test pressure range and provide several benefits over the TCS, such as:

- Entire specimen subject to the same compression
- Realistically models the compression of internal battery components
- Friction is only produced by the punch, tension mechanism
- No alignment issues
- Produce the desired failure mode

Once the friction is removed, the failures should occur near the center of the punch tip which is the desired failure mode. When this occurs, the micro model can be adjusted to match the
experimental conditions and a numerical comparison between experimental and modeled results can be conducted.
BIBLIOGRAPHY


APPENDICES

A) TCS Basic Operating Procedure

This Appendix describes the basic set up and operation of the TCS to aid in any future research involving the device. This assumes the operator is familiar with the INSTRON machine and software.

1. Samples are prepared as described in Section 3.1.2.
2. Install the 2 kN load cell in the INSTRON.
3. Install the 500 N load cell in the TCS and connect it to the strain meter using and calibrate if required as described in Section 3.1.1.
4. Mount the TCS to the base of the INSTRON.
5. Use the adapter to connect INSTRON load cell to the upper portion and clamp ring of the TCS.
6. Remove the clamp ring and insert the desired specimen and tighten.
7. The clamp ring is then placed onto the guide rods but held above the punch tip.
8. Use the INSTRON to lower the upper portion of the TCS until the clamp ring can be reattached while on the guide rods.
9. Install the mount for the compression spring before lowering the clamp ring.
10. Use the INSTRON normal then fine adjustment to lower clamp ring until the specimen is just touching the punch tip.
11. Zero the INSTRON at this location so all tests will start from the same location.
12. If desired, foam and/or grease should be added.
13. Apply compression by inserting the spring and tip into the mount and tightening to the desired level. To help maintain spring alignment use a level on-top the compression spring mount, count and maintain equal number of turns for each side, use calipers to measure the thread height of each side of the mount or amount of thread remaining.
14. Verify amount of pre-compression on the strain meter.
15. Select the correct program for the INSTRON to use and verify the displacement rate.
16. Verify the displacement and tension loads on the Bluehill© software.
17. Commence testing.
B) Example of an Experiment Summary

This Appendix was included to show how each test was tracked and it includes basic parameters, a table for individual tests, comments on specific tests, the photograph numbers associated with each test, generic plots the BlueHill software produced from raw data, notes on the current test, and notes describing the next steps.

TCS tests on Separator samples
Date: 10/13
Operator(s): Nathaniel Byrd
Load Frame: INSTRON
Load Cell: 2kN
Speed: 4mm/min
Specimen type: circular
Condition of sample: Greased punch with greased foam under PC

<table>
<thead>
<tr>
<th>Test</th>
<th>Punch</th>
<th>Layers</th>
<th>Separator</th>
<th>Failure</th>
<th>Load cell</th>
<th>Pre Comp Mpa</th>
<th>Grease</th>
<th>Foam</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>L-Metal</td>
<td>1-S</td>
<td>Ceramic Coated</td>
<td>100.7</td>
<td>32</td>
<td>0</td>
<td>y</td>
<td>y</td>
</tr>
<tr>
<td>2</td>
<td>L-Metal</td>
<td>1-S</td>
<td>Ceramic Coated</td>
<td>96</td>
<td>173</td>
<td>5</td>
<td>y</td>
<td>y</td>
</tr>
<tr>
<td>3</td>
<td>L-Metal</td>
<td>1-S</td>
<td>Ceramic Coated</td>
<td>90.1</td>
<td>173</td>
<td>5</td>
<td>y</td>
<td>y</td>
</tr>
<tr>
<td>4</td>
<td>L-Metal</td>
<td>1-S</td>
<td>Ceramic Coated</td>
<td>92.1</td>
<td>314</td>
<td>10</td>
<td>y</td>
<td>y</td>
</tr>
<tr>
<td>5</td>
<td>L-Metal</td>
<td>1-S</td>
<td>Ceramic Coated</td>
<td>91.3</td>
<td>314</td>
<td>10</td>
<td>y</td>
<td>y</td>
</tr>
<tr>
<td>6</td>
<td>L-Metal</td>
<td>1-S</td>
<td>Ceramic Coated</td>
<td>87.1</td>
<td>597</td>
<td>20</td>
<td>y</td>
<td>y</td>
</tr>
<tr>
<td>7</td>
<td>L-Metal</td>
<td>1-S</td>
<td>Ceramic Coated</td>
<td>90.3</td>
<td>597</td>
<td>20</td>
<td>y</td>
<td>y</td>
</tr>
<tr>
<td>8</td>
<td>L-Metal</td>
<td>1-S</td>
<td>Ceramic Coated</td>
<td>96.7</td>
<td>32</td>
<td>0</td>
<td>y</td>
<td>y</td>
</tr>
<tr>
<td>9</td>
<td>L-Metal</td>
<td>1-S</td>
<td>Ceramic Coated</td>
<td>95.8</td>
<td>173</td>
<td>5</td>
<td>y</td>
<td>y</td>
</tr>
<tr>
<td>10</td>
<td>L-Metal</td>
<td>1-S</td>
<td>Ceramic Coated</td>
<td>91.2</td>
<td>314</td>
<td>10</td>
<td>y</td>
<td>y</td>
</tr>
<tr>
<td>11</td>
<td>L-Metal</td>
<td>1-S</td>
<td>Ceramic Coated</td>
<td>83.5</td>
<td>597</td>
<td>20</td>
<td>y</td>
<td>y</td>
</tr>
<tr>
<td>12</td>
<td>L-Metal</td>
<td>1-S</td>
<td>Ceramic Coated</td>
<td>97.7</td>
<td>32</td>
<td>0</td>
<td>y</td>
<td>y</td>
</tr>
</tbody>
</table>

Comments
Test 1  Failed center IMG_7344 - 46
Test 2  Failed just off center not under foam (48-50)
Test 3  Failed near center (51-54)
Test 4  Failed near center (56-58)
Test 5  Failed just off center near edge of foam (59-61)
Test 6  Failed near center under foam (62-68)
Test 7  Failed center stopped immediately (69-73)
Test 8  Failed center (74-76)
Test 9  Failed center (77-79)
Test 10 Failed off center not under foam, specimen tore when removing (80-81)
Test 11 Failed off center not under foam, specimen tore when removing (82-86)
Test 12 Failed center (87-89)

Specimen 1 to 11

Switched to the metal tip for this run and used thinner foam in order to more accurately position the pre compression. Initially ran one with no pre-compression then 2 of each 5, 10, and 20 MPa and was pleased with the results so ran 3 total of each. No failures began at boundary but some failure was off-center. In all but T-3 failure occurred lower at higher pre compression. The next
step is to put the data into excel or matlab and produce plots showing 3 runs at same pre-compression then average the failures and plot pre-compression versus average failure.

The next tests will be using multi-layered samples with interruptions to determine the failure order.