

LITHIUM - BATTERY - SAMPLE PREPARATION

By Dr. E. Mogire

Introduction

Lithium ion batteries (LIB) have found wide application due to their high energy and power densities compared to conventional batteries. They find wide application in portable electronics, hybrid/full electric vehicles (H/EV), power tools among others. Electric vehicle technology is increasingly being adopted as the preferred mode of transport over traditional Internal combustion engine (ICU) technology, due to their perceived lower impact on green-house gas emissions. This is primarily driven by low emission targets for ICU engines targeted at less than 95g/Km by 2021, (Miller, 2015) (Tsiropoulos I., 2018). For automotive applications, the lithium ion batteries are found in hybrid, plug-in hybrid electric vehicles or fully electric vehicles. The selection of a particular type of a lithium ion battery depends on its battery chemistry, which has a direct bearing on its safety during use, the power or energy density, charging/discharging time, and lifetime behaviour. Individual cells are normally assembled into packs containing hundreds/thousands of cells to form battery packs that will provide the required voltage and current density for efficient running of H/EV cars. Individual cells and/or battery packs assemblies can be evaluated metallographically to validate battery chemistries and construction, and the inspection of various joining techniques used to make the packs. The following article will highlight the procedures that can be adopted for metallographic assessment of various battery components.

Sample Preparation Techniques

Sectioning

Before sectioning is carried out, ensure the battery is fully discharged and no nominal voltage is still present. This is important to minimise risks associated with short circuiting of the cell. Sectioning of Lithium batteries should be carried out in dry state to avoid chemical reaction with the coolant and its subsequent absorption into the cell laminate structure. The active components in both the anode and cathode laminates consists of multi-materials that have high affinity to water resulting in corrosion products that are harmful. Sectioning was carried out on IsoMet High speed with the aid of double saddle chuck for cross-sections. The blade used was a non-ferrous SiC abrasive blade. Ensure there is an extraction system fitted to the back of the machine for the dust generated. For sectioning of the outer metallic casing without the internal active cell elements, wet sectioning can be carried out. Wet sectioning was also carried out on welded terminal points, and since the materials are mainly metallic, a diamond wafering wheel can also be used instead of abrasive blade, (SumMet, 2018).

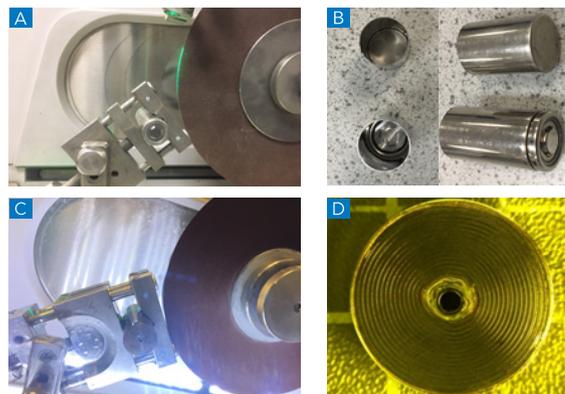
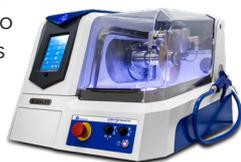


Figure 1 showing sectioning on IsoMet high speed with (a) illustrating clamping configuration, (b) sectioned casing material without rolled cell element, (c) showing battery with cell element for cross-sectional view and (d) illustrating sectioned cell with characteristic laminate structure.

Mounting

Lithium ion batteries can be prepared in mounted and unmounted configurations. For unmounted samples, these should be hand prepared and for ease of handling the cross-sections, one can design a jig fixture to hold them in place when carrying out coarse to fine grinding steps. If the samples are to be mounted, castable mounting technique should be adopted using epoxy-based resins, such as Epothin 2 or EpoxiCure 2. Epoxide resins are preferred due to their low peak exotherm temperatures during curing as well as their low reactivity with battery constituent elements. Castable mounted samples are ideal for terminal weld evaluations, dimensional checks and battery cap assembly region check, as the technique ensures weld integrity without distortion and/or damage compared to a compression (hot) mounting process (SumMet, 2018).

Grinding and Polishing

Cell grinding should be carried out using SiC papers for several reasons; the material making up the lithium ion batteries have different abrasion rates and consists of various metallic components, polymeric materials, compacted powders among others. For uniform grinding and to ensure good planarity, SiC papers offer the best alternative without causing too much subsurface damage of these materials. For the initial grinding step, a fine grit SiC paper is recommended as long as it doesn't compromise the material removal process. If high stock removal is desired, coarser grits can be adopted, however, do not use grits coarser than P600(360grit). Using finer grits for initial grinding has the added advantage of minimizing the number of subsequent

Step No.	Surface	Abrasive	Lubricant/ Extender	Force (per specimen)	Time (min:sec)	Platen Speed (rpm)	Head (rpm)
1	SiC Paper	P600	Dry	Moderate Pressure	Until Plane	120	Manual
2	SiC Paper	P1200	Dry	Moderate Pressure	Until Plane	120	Manual
3	SiC Paper	P2500	Dry	Moderate Pressure	Until Plane	120	Manual
4	SiC Paper	P4000	Dry	Moderate Pressure	Until Plane	120	Manual

Table 1 shows the preparation routine used to prepare the LiCoO battery. This is applicable to other types of lithium ion battery chemistries.

steps required. Dry grinding is recommended at relatively low revolutions per minute of the platen to ensure better control of flatness without causing subsurface damage. Safety precautions must be considered during dry grinding due to the risk of dust inhalation. Lower platen speeds and using dust mask significantly minimizes the risks associated with dry grinding. Buehler EcoMet 30 was used to prepare the mounted micro-sections as per the procedure in Table 1, (SumMet, 2018)

The internal cell components are made up of the cathode, a separator and the anode laminates are wound to make a cell element. The cathode terminal is then connected to the battery lid with its built-in safety valve mechanism and a PTC (positive temperature coefficient) switch, Figure 3. During abnormal use of the battery, such as excessively drawing the current from the battery, the conductive polymer making up the PTC heats up becoming resistive, stopping the current

Step No.	Surface	Abrasive	Lubricant/ Extender	Force (per specimen)	Time (min:sec)	Platen Speed (rpm)	Head (rpm)	Rotation
1	SiC Paper	P400	Water	25N	Until Plane	250	60	<<
2	SiC Paper	P1200	Water	25N	04:00	150	60	<<
3	PoliCloth	3µm MetaDi	MetaDi	30N	04:00	150	60	<<
4	Chemomet	0.05µm MasterMet, 0.05 MasterPrep	Water**	25N	02:30	120	60	>>

Table 2 shows the preparation routine used to prepare battery casing components and welded interconnects for battery pack assembly.

Microscopy analysis

The as-received cell, sectioned cells and micro-sections were analysed using both optical and electron microscopical techniques. Optical metallography involved using a Nikon LV150 upright microscope for detailed view of the laminate structure, and a macro stand for photomicrography of the cell and its cross-sections. For Electron microscopy, FEI electron microscope was used for energy dispersive spectroscopy (EDS) work. Unmounted samples were used for electron microscopy, as the cell components were conductive ensuring no charge build-up effect occurred for good elemental analysis. For external cell geometry without the active components, traditional techniques of metallographic preparation were carried out.

Cell Structure & Description

The internal cell structure of a lithium ion battery consists of an outer cylindrical casing protecting the internal active cell elements made of a graphite anode, a lithium metal oxide cathode, an electrolyte of a lithium salt and an organic solvent. The internal cell structure is common to all lithium ion batteries with the main variations being on the active anode/cathode chemistries. The outer casing, Figure 2 (a-e), houses the rolled cell element. Depending on the end application of the battery, a polymeric outer sheath can be used (d and e) or the battery can be uncovered, which offers ideal surface for battery cooling when assembled into a battery pack.

flow and acts as short circuit protection. On cooling down, the PTC returns to conductive state allowing the battery to operate. The safety valve mechanism functions to relieve

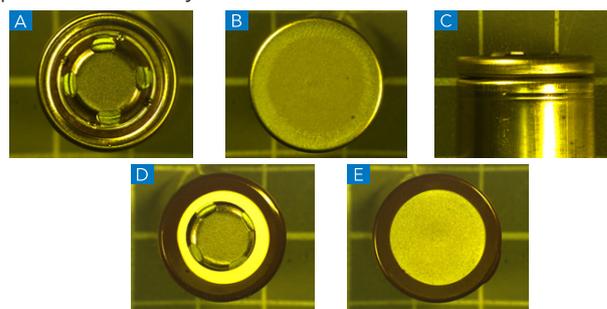


Figure 2 illustrates the outer case structure of a cylindrical solid body lithium ion battery without the polymeric sheath with (a) showing the cathode, (b) anode side and (c) is a side view of the battery lid cap region, (d) and (e) illustrate the polymeric outer sheath.

pressure build-up in the cell and consists of membrane seal that ruptures under high pressure, which might cause cell leakage and dry-out. Newer battery designs have mitigated this issue by utilising a re-sealable vent. The battery lid, the safety valve mechanism and the PTC (the positive temperature coefficient) element are fixed to the open end of the battery casing by caulking them through the insulation sealing gasket and the inside of the battery can is thus sealed, Figure 3(a).

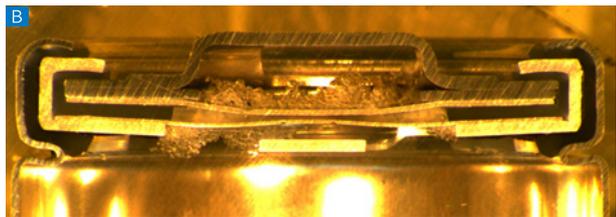
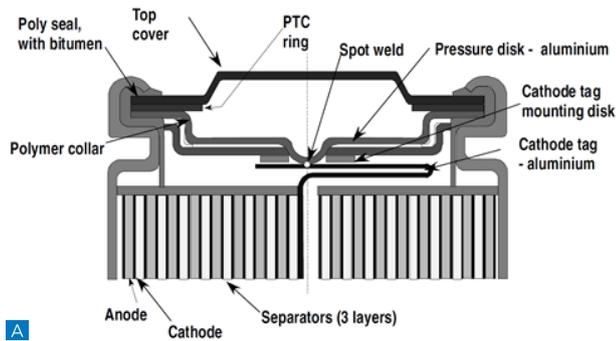


Figure 3 (a) schematic illustration of battery cross-section at the cap (open) end side showing the various components making up the outer casing, and (b) illustration of actual cell cross-section.

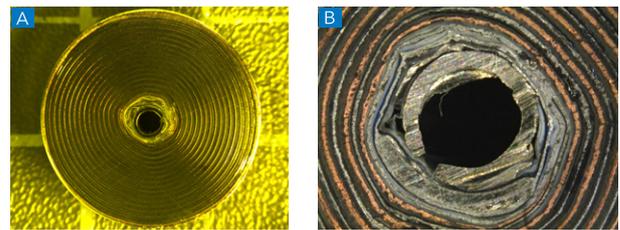


Figure 4 Battery cross-section showing the laminate layers consisting of both the cathode and the anode, outer casing and centre pins. (B) shows a close-up of the active cell element layers around the centre pin.

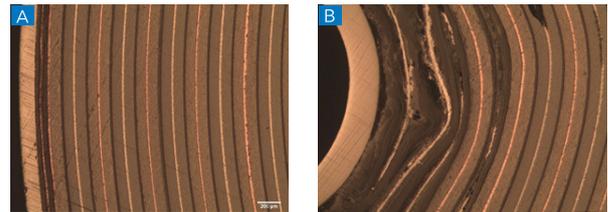


Figure 5 (a) Optical micrographs above illustrate the cell cross-sectional view. Figure A shows the outer casing and alternating laminate layers composed of anodic and cathodic components, (b) shows the structure towards the centrepin of the battery

The laminate layer on the other hand is wound around a metallic centrepin consisting of a cathode and anode with their respective active elements, Figure 4. The cathode consists of a sandwich cathode active material layer on either side of a cathode current collector. The current collector is a metal foil, which can be aluminium foil, nickel foil or stainless steel foil. The cathode active material layer consists of a formulation with ceramic, conductive agent and a binder. The ceramic is an inorganic oxide, which can be Al_2O_3 , SiO_2 , ZrO_2 , MgO , Na_2O , TiO_2 , among others. Alumina is particularly preferable because of superior diffusion of lithium. The active cathode materials include lithium oxides, lithium sulphides or complex compounds containing lithium such as lithium phosphate compounds. These include lithium/cobalt oxides ($LiCoO_2$), lithium/nickel oxides ($LiNiO_2$), lithium/nickel/cobalt oxides ($LiNiCoO_2$) and lithium/nickel/cobalt/manganese oxides ($LiNiCoMnO_2$). Also, phosphoric acid compound based include lithium-iron-phosphoric acid compounds ($LiFePO_4$) and lithium-iron-manganese-phosphoric acid compounds ($LiFeMnPO_4$), Figure 6. Selection of battery chemistries depends on their safety, power and energy densities, and life time. The Titanate, $LiFePO_4$, $LiCoO_2$ and NCM have the widest application in automotive.

The conductive agent consists of carbon-based materials such as graphite or carbon black for better conductivity. For binders, these are fluorine-based polymers such as polyvinyl fluoride, polyvinylidene fluoride and polytetrafluoroethylene, fluorine type rubbers among others.

The anode consists of a sandwich laminate made up of anode active material layer between a current collector. The current collector is a metallic foil such as a copper or nickel foil. The anode active material layer consists of a conductive carbon doped with lithium such as graphite and bound with the same materials as found on cathode laminate, Figure 7.

The laminate structure can also be assessed optically for evaluation of anode/cathode layers materials thicknesses, and volume or area fraction analysis. Area/volume fraction analysis can be insightful on energy density evaluation of active materials in Li-ion batteries, Figure 6. (Du, 2017) modelled electrode thickness as a function of energy density, and found that the energy density had a maximum point at a critical electrode thickness for a given discharge rate. They concluded that cell polarisation and underutilisation of the active materials was the limiting factor affected by Li-ion diffusion in active materials and its depletion in the electrolyte phase, (Yu, 2013).

OmniMet image analysis software was used to evaluate area/volume fractions and thickness measurement, the software can be scripted to do overlay of multiple lines on the anode and cathode layers irrespective of geometry and will calculate feret diameter at various angles for an in-depth analysis of the thicknesses, (SumMet, 2018).

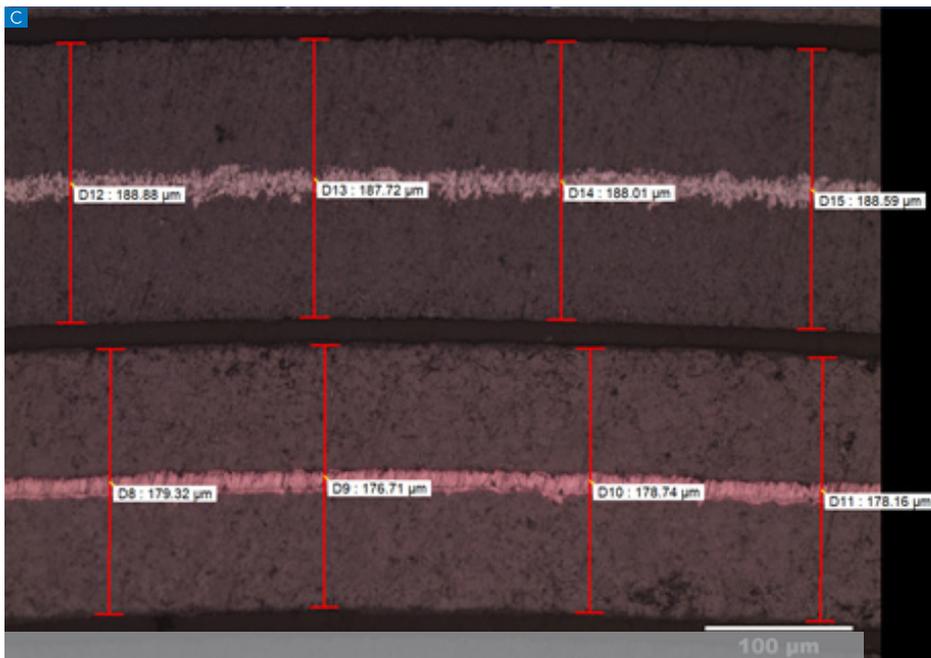
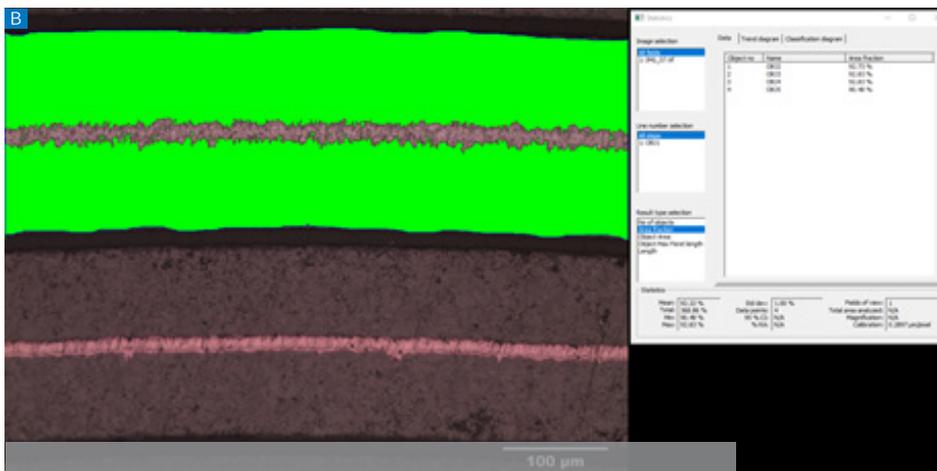
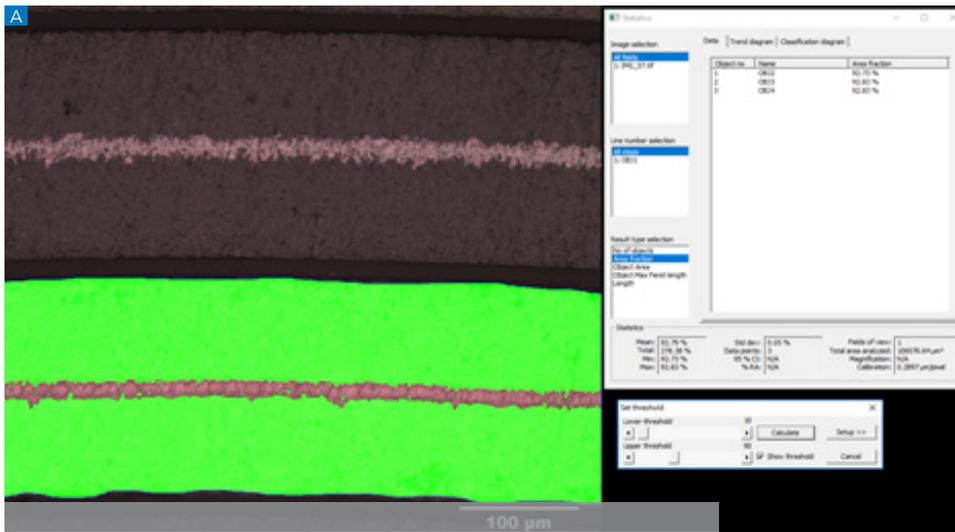


Figure 6 illustrating active material area/volume area fraction analysis of (a) anode and (b) cathode layers, and (c) showing corresponding approximate thicknesses.

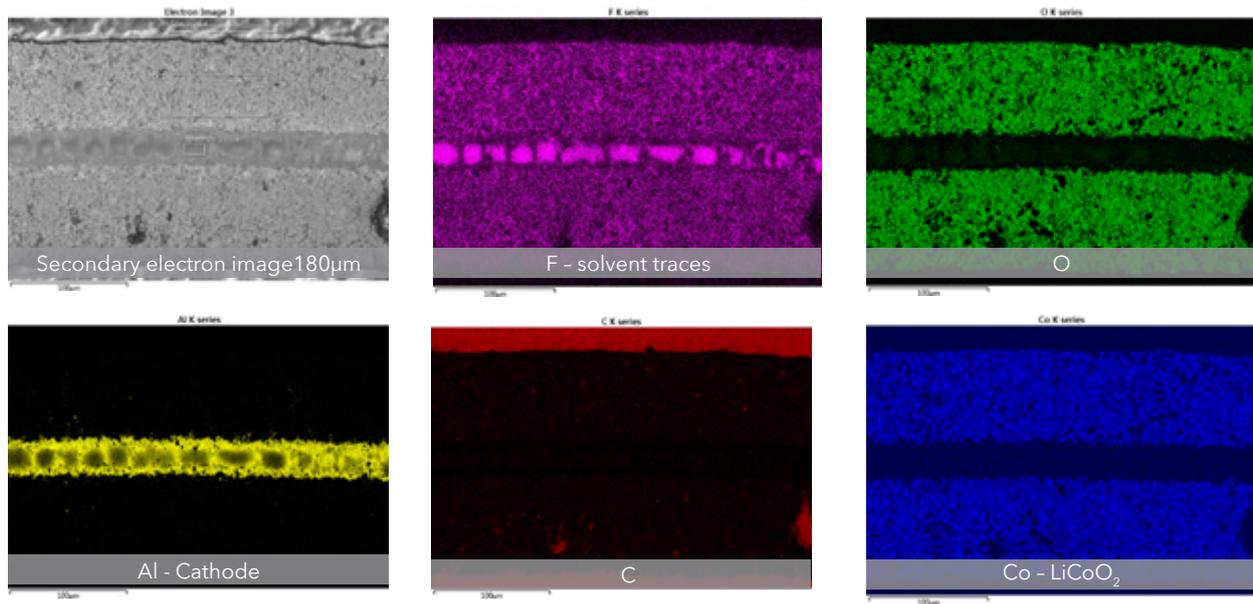


Figure 7 illustrates a higher magnification electron microscopy (EDS) analysis around the cathode laminate layer showing its compositional chemistry. It's evident that remnant active and binder agent compositions have also been picked up.

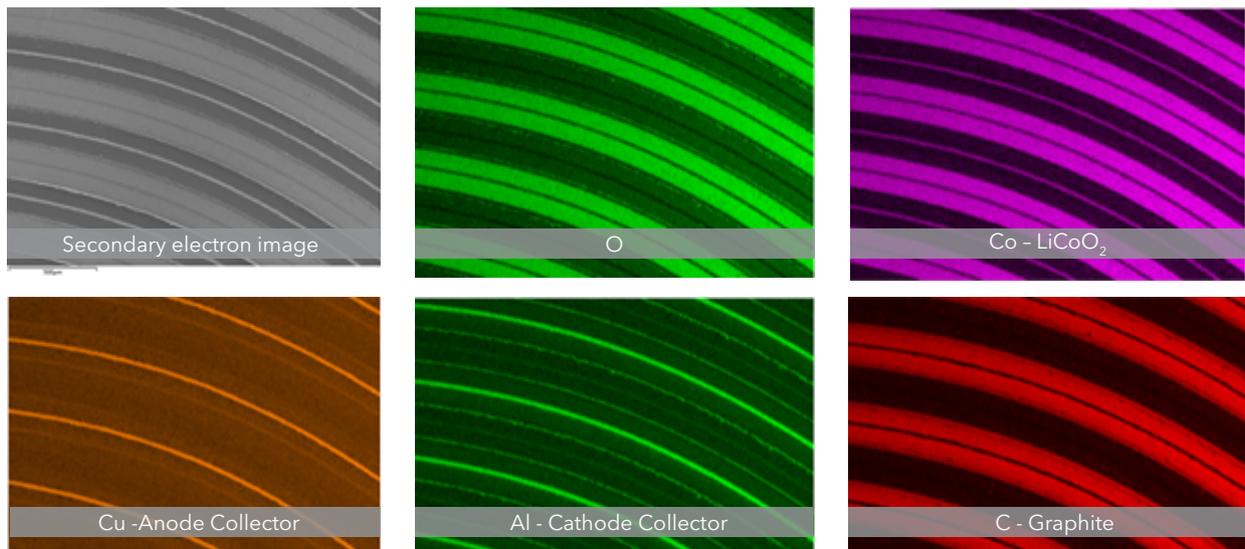


Figure 8 illustrates a lower magnification electron microscopy (EDS) analysis of the laminate structure of a Lithium ion battery with the main constituent components elemental map analysis shown. The micrographs clearly highlight the anode (carbon rich with Cu charge collector and cathode layers (cobalt rich) with Al charge collector).

Cell Interconnects - welding

Welding or joining technologies are key areas of development with ever increasing demand for higher energy densities, low self-discharge, and portability of battery packs as alternative power sources for H/EV's. A battery pack typically consists of hundreds and/or thousands of individual Li-ion batteries that are connected in series or parallel configurations to achieve the required power and energy. A battery pack can either be a single module or several modules connected in series or parallel configurations, with sensor and battery control systems encased in housing structure. The module and/or packs do require robust and reliable interconnects and the joining methodology depends on the type of cell used but also ensuring good electrical, thermal and mechanical behaviour in use, (Das, Li, Williams, & Greenwood, 2018).



Figure 9 illustrates the common welding techniques of lithium ion batteries with left - spot weld, middle - ultrasonic weld and right - laser beam welds interconnects on Li-Ion cylindrical cells, (Martin J. Brand, 2015).



Figure 10 shows examples of (a) ultrasonic and (B) laser beam welds before cross-sectioning and castable mounting to investigate the quality of the weld.

The most common method of connection (welding) of battery packs is either through resistance spot welding, ultrasonic welding and laser beam welding. The welding parameters and resultant welded area must be of good quality without

imperfections (voids) that are likely to occur due to the welding technique. They concluded that different methods had distinct advantages and disadvantages from an application viewpoint, with spot welding being ideal for joint partners with low conductivity such as nickel-plated steel conductors and the 18650 sized cells. Ultrasonic was suitable for nearly all battery cell connections when welded components were clamped during welding process. For unclamped battery configuration before welding, ultrasonic technique led to crack formation in the conductors and subsequent damage to the battery cell. Laser welding was deemed ideal for better weld quality and any battery cell could be connected with fewer risk factors relating to surface reflections and spatters (Steen, 2010).

For the weld tests carried out, empty battery cans welded using ultrasonic, spot and laser welding as illustrated in Figure 9 were metallographically prepared to investigate their microstructures and/or identify defects that affect electrical contact resistance. Copper interconnect samples were used in the tests due to copper's high conductivity, which would pose a considerable challenge during welding as reported by (Zhou, 2000) and (Martin J. Brand, 2015), with a high likelihood of exhibiting the welding defects.

Weld preparation

Preparation of the welded joints was carried out semi-automatically as per Table 3 to ensure all the components materials types were metallographically prepared.

Figure 10 shows metallographic sections of a spot weld and a laser weld of a copper interconnect onto a battery can. Figure 11 (a) shows a spot weld with its characteristic roots attributed to the induced eddy current affecting the weld profile. There is presence of micro/macro cracks within the roots of the spot welded interconnect, the length of which would compromise the cell casing integrity. On the other hand, laser welded interconnect, Figure 11 (b) had fewer microvoids, and on certain areas did not show weld defects that would affect cell casing integrity confirming the observations made by (Martin J. Brand, 2015).

Step No.	Surface	Abrasive	Lubricant/Extende	Force (Per Specimen)	Time (min:sec)	Platen Speed (rpm)	Head Speed (rpm)	Rotation
1	SiC/DGD equivalent	P400	Water	5lbs	Until Plane	250	60	<<
2	SiC/DGD equivalent	P1200	Water	5lbs	01:30	250	60	<<
3	PoliCloth	3µm	MetaDi Fluid	5lbs	04:00	150	60	<<
4	ChemoMet	0.06 µm MasterMet	Water**	5lbs	02:30	150	60	<<

Table 3 shows the preparation routine used to prepare the LiCoO battery. This will be applicable to other types of lithium ion batteries.

compromising the battery casing architecture. (Martin J. Brand, 2015) investigated these techniques and showed that laser welding technique had the lowest electrical contact resistance, and with spot and ultrasonic techniques showing considerably higher contact resistance attributed to

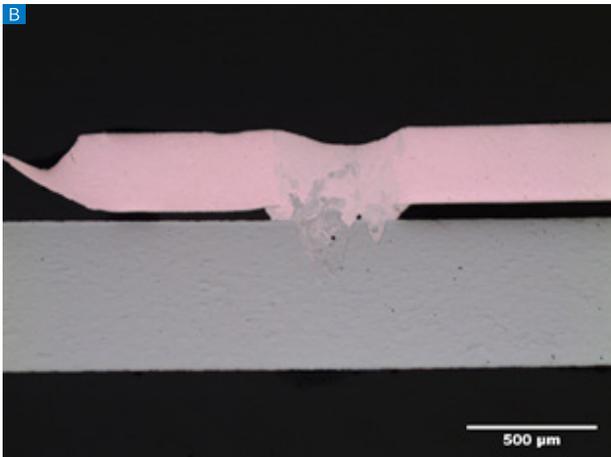
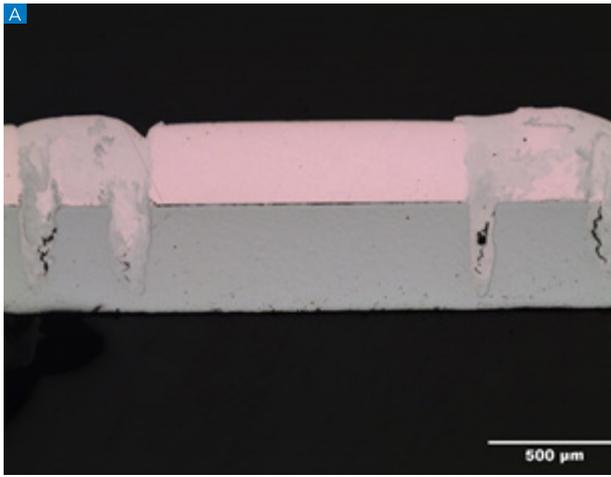


Figure 11 shows examples of polished cross-sections of (a) spot weld and (b) Laser weld. It's evident that spot weld has more voids and appears to have penetrated deeper into the casing material and could compromise battery casing structure whereas the laser weld is moderately penetrating. This of course depends on welding parameters used and could be optimised to minimise observed defects.

Summary

The article highlights metallographic preparation of Li-ion battery cells found in battery modules/packs of cylindrical 18650 cell type. The methodology showed herein is also applicable to other battery types such as prismatic and pouch shape design. The paper also highlighted the safety precautions that need to be taken before sectioning of cells and associated challenges in mounting and subsequent grinding/polishing procedures. The metallographically prepared cell was also characterised optically, and using electron microscopy technique, elemental composition mapping the cell structure was made possible. The cell interconnectivity through spot, ultrasonic and laser welds was also metallographically prepared and optically characterised. The defects, such as voids and cracks relating to the weld methodology used were also shown with the intent of highlighting their effect on battery module/pack performance behaviour.

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For further queries please contact us on techsupport@buehler.com or lab.eu@buehler.com or visit our website at <https://www.buehler.com>

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**BUEHLER Worldwide Headquarters
North America-South America Offices**
41 Waukegan Road
Lake Bluff, Illinois 60044-1699 USA
P: 800 BUEHLER (800-283-4537)
P: (847) 295-6500
www.buehler.com | info@buehler.com

European Headquarters
BUEHLER Germany
info.eu@buehler.com
BUEHLER France
info.eu@buehler.com
BUEHLER United Kingdom
info.eu@buehler.com

BUEHLER China
info.cn@buehler.com
BUEHLER Japan
info.japan@buehler.com
BUEHLER Asia-Pacific
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