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Understanding the limits of rapid charging using instrumented commercial 18650 high-energy Li-ion cells

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ABSTRACT

The charging rates of commercial high-energy Li-ion cells are limited by the manufacturer's specifications leading to lengthy charging times. However, these cells are typically capable of much faster charging, if one ensures that the thermal and electrode-specific voltage profiles do not exceed safety limits. Unfortunately, precise and *in-situ* measurements of these parameters have not been achieved to date without altering the operation of these cells. Here we present a method to assess the maximum current for commercial 18650s, using novel instrumentation methods enabling in operando measurements. We found the maximum charging current that could be safely applied to the evaluated high-energy cells is 6.7 times higher than the manufacturer-stated maximum. Subsequently a rapid-charging protocol was developed that leads to over five-fold reduction in charging times without compromising the safety limits of the cells. We anticipate our work to be a starting point for a more sophisticated understanding of commercial Li-ion cells through deployment of diverse *in-situ* sensor systems. This understanding will enable advances in battery materials science, thermal engineering and electrical engineering of battery technology. Furthermore, this work has the potential to help the design of energy storage systems for high performance applications such as motor racing and grid balancing.

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1. Introduction

Rechargeable Lithium-ion (Li-ion) cells are widely regarded as the technology of choice for the electrical energy storage and power delivery solutions. With formats ranging from small portable devices to large high energy packs, one of the key applications of this technology is in the automotive industry as a battery pack for electric and hybrid vehicles [1–4]. Fast charging of these batteries is becoming increasingly important as consumers demand reductions in charging time. Concurrently, the applied charging regime has a significant impact on cycle life, thermal performance and safety [5]. Charging guidelines given by cell manufacturers are generally conservative and so significant testing is required before deploying in an application that requires rapid charging.

The most commonly used charging strategy is constant-current constant-voltage (CC-CV), although alternative charging modes are being explored [5–7]. Unfortunately, these attempts are usually lacking experimental *in-situ* thermal measurement or electrode-specific data [7–9]. When assessing the maximum performance limits of the cell, the risk of internal overheating resulting in catastrophic thermal runaway is greatly increased [10]. The commonly agreed five sources of heat generation in Li-ion cells [11] – electrolyte, anode and cathode resistances (R_e, R_a and R_c), anode material and cathode material entropy change (ΔS_a and ΔS_a), can be summarised as Joule (resistive) heating and exothermic reactions [10]. Both of these phenomena are C-rate related and accelerate when a cell experiences heavy load. Subsequently, as the cell temperature increases, the reaction rates for the decomposition of the electrolyte increase, which can lead to the electrolyte breakdown and gas formation, resulting in pressure build-up in the cell [12]. Additionally, if a cell is charged too fast, lithium metal can electroplate on the anode, which may grow in the form of dendrites and eventually pierce the separator, causing an internal short circuit and subsequent catastrophic failure [13]. This is most pronounced in the case of high-energy cells which, while providing significantly more gravimetric and volumetric energy density than their high-power counterparts, suffer from significantly limited charge rates [14,15].

To be able to overcome these obstacles, information about each electrode's potential is required, complemented by the surface and internal thermal load responses of the cell. Here we use a novel

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instrumentation design for commercial 18650 cells that minimises the adverse and previously unavoidable alterations to the cell geometry [16]. This includes an *in-situ* reference electrode coupled with an optical fibre temperature sensor. This enables the measurement of each electrode's potential, supplemented by the cell's internal and external temperature profiles. In addition to performance and safety optimisation, monitoring of internal cell temperature and each electrode potentials can be an important asset when trying to minimise aging effects and enable degradation effects early onset detection, e.g. by observing the negative electrode voltage slippage as the cell ages [17]. Introduction of a reference electrode was previously attempted by other researchers on Li-ion pouch and cylindrical cells [18–20]. The latter was more challenging due to assembly constraints, often resulting in extensive modification of the cell which can affect its internal resistance and electrolyte stability.

In-situ thermal instrumentation of Li-ion cells using thermocouples has been explored by other researchers [21–23]. However, this technology has intrinsic limitations that restrict its applicability. Thermocouples measure only relative temperature changes, therefore requiring a cold junction element and extra calibration. A thermocouple is not capable of multiplexing to a single wire, which means additional signal lines are required, thickening the sensor and adding points of failure to obtain distributed sensing. Conversely, FBG sensors support multiplexing using a single fibre thread with multiple gratings. Finally, the sensitivity of thermocouples is relatively low, requiring additional analogue conditioning circuits. Therefore, fibre optic temperature sensors are evaluated instead as a more promising alternative.

In this work, we use the instrumented Li-ion cell to assess the maximum charging current for the commercial 18650 high-energy cell and derive expedient charging parameters which remain within the thermal and potential safety limits of the cell. The approach utilised offers an unprecedented insight into the performance characteristics of the cell, allowing a much better understanding of actual electrochemical and thermal limitations of Li-ion batteries for the benefit of scientists researching new battery materials and engineers designing electric vehicle and grid storage systems.

2. Experimental

2.1. Cell instrumentation – reference electrode

Lithium metal foil was used as an in-situ reference electrode. Bare metal reference electrodes called quasi-references are often used in certain situations when it is impossible to introduce standard references due to mechanical [24] and/or chemical [25] constraints [26]. Lithium metal electrodes were previously reported to work successfully in pouch cells, where the geometry of the pouch cell format allows for easier modification [27]. This is due to the soft, flexible pouch material, which can accommodate additional elements inside the cells, while the electrodes in cylindrical cells are surrounded by a stiff metal can. Additionally, pouch cells have a substantially sized and easily accessible flat area at the top and bottom of the electrode stack, while cylindrical cells are built with very tightly wound electrodes - this allows for the reference placement only at the top of the jellyroll, and also requires the metal can to be cut open for cell modification.

As lithium metal reference electrodes can suffer from polarizability at high current densities, bespoke lithium titanate and lithium iron phosphate reference electrodes were also evaluated by other researchers [28,29]. In this work, a high-impedance input connection to the potentiostat is used to prevent any current from passing through the electrode, while maintaining the simplicity of the design and preventing any new foreign materials from influencing the system [26].

A strip of Li foil was used, wrapped in separator material to prevent it short-circuiting against the cell can or the electrode jellyroll, subsequently disturbing and reacting with the electrode materials. The electrode was then inserted into the top of the cylindrical cell, below the cathode cap, in contact with the cell's jelly roll. A spacer made from Kapton tape was placed on the upper side of the reference electrode to ensure good ionic contact with the jelly roll and prevent shorting with the cathode current collector once assembled inside the cell. After the modification procedure, the anode, cathode and full cell potentials were monitored for 24 h to confirm stability of the modified cell and the reference electrode readings. This setup allows for simultaneous observation of the cathode and anode potentials of a working Li-ion cell *vs.* Li/Li^+.

2.2. Cell instrumentation – thermal sensors

The selected thermal sensing method involves the use of Fibre Bragg Gratings (FBG) [30]. The FBG element was obtained from *Smart* Sensors Inc. in its bare form, sealed with a polyamide recoat stable in a temperature range of −270 °C to +300 °C. The element has a nominal wavelength of 1545 nm at 25 °C when no strain is applied. The bare fibre was then threaded through an aluminium tube, forming a strain protection layer. An outer skin of fluorinated ethylene propylene heat-shrink was applied over the fibre and aluminium, adding protection from the electrolyte. Elements prepared in such a way can withstand electrical, chemical and mechanical stress inflicted during the instrumentation procedure and during cycling. Fig. 1 shows the complete element.

An optical spectrum analyser, broadband laser source and a three port optical circulator (*Thor Labs Inc.*) were used to interrogate the wave shift of the FBG. The analyser was subsequently connected *via* RS-232 to a data logging computer. The observed wave shift was being translated to a temperature shift using an equation specific to a given FBG element based on its sensitivity

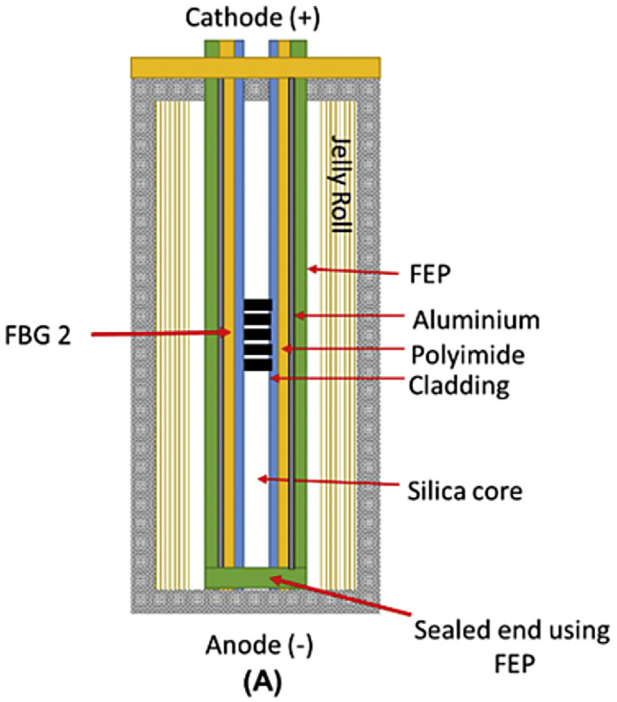

Fig. 1. Schematics of the FBG sensing element embedded into a Li-ion cylindrical cell.

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