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Thermal Runaway: Can Ultrasound Finally Solve Li-ion Cells' Most Dangerous Challenge?

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As part of European Automobile Manufacturers' Association-funded projects, the Mobility Innovation Hub has been collaborating with the University College of London to explore the use of ultrasound for battery diagnostic. This article reports on experimental studies aimed at evaluating the application of ultrasonic sensors to detect abuse conditions, internal defects, and early thermal runaway signals in battery cells. The changes in acoustic behaviours of battery cells have been monitored and evaluated during temperature-controlled, nail penetration, and overcharge as well as homogeneous and localised heating tests. Finally, ultrasound's ability to capture key events during thermal runaway propagation scenarios has been assessed by triggering thermal runaway in a cell of a small prototype module.

Introduction

The occurrence of several battery-related accidents over the years has risen public awareness of risks and safety issues around electric vehicles (EVs). These accidents can be due to a cell or a battery pack failing due to mechanical, thermal, or electrical abuse or as a result of cell manufacturing defects. Both classes of safety accidents can trigger the thermal runaway (TR) of a cell, a potentially catastrophic event. During TR the cell temperature increases due to exothermic reactions. Temperatures higher than 1000 ° C can be reached and high amounts of flammable and harmful gases are released. In a battery pack, the heat generated from a cell entering TR can trigger TR in the surrounding cells, causing a chain reaction within the pack with potentially disastrous consequences which is known as thermal runaway propagation (TRP). Current safety features implemented on battery management systems (BMSs) heavily depend on thermocouples instrumented to the surface of the cell. However, for an effective early warning system, the ability to record or detect internal changes in a battery cell is vital. A novel technique that is beginning to attract attention for the study of electrochemical devices is ultrasound [1-3]. Ultrasonic testing is based on monitoring how ultrasound waves propagate through an object of interest. Its main advantages are that it is non-destructive, can be conducted *in-operando*, is relatively cheap and measurements are very quick, taking only microseconds to complete.

This study explores the usage of ultrasound as a tool to detect cell abuse conditions, internal defects as well as identify early steps leading to TR at cell and module level. For the interested reader, a thorough description of the experimental studies here reported has been published in specialized scientific journals [4-5].

Working Principle

When conducting an ultrasonic measurement, an ultrasonic pulse wave is generated by a piezoelectric transducer. While the acoustic signal travels through the object of interest, the wavefront is influenced by the properties of the object's component materials. One of the most commonly measured properties of the acoustic wave is the time of flight (ToF) which is the time taken for the generated wave to travel through the object and be reflected back to the transducer. The ToF is affected by the speed of sound through the materials through which it propagates. Typical waveforms obtained are shown in Fig.1.

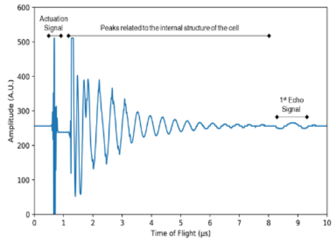

Figure 1 - Typical acoustic waveform

The initial signal at a ToF below 1 μs is due to the generation of the initial ultrasonic pulse and is not influenced by the cell under study. The peaks at ToF values higher than 1 μs are reflection peaks related to the internal structure and the interfaces present within the cell. Acoustic peaks corresponding to higher ToFs reflect the properties of interfaces located farther from the acoustic emitting device. The amplitude of these internal reflection peaks decreases steadily with penetration depth due to signal attenuation and reflection in previous layers. At ca 8.7 μs a peak with a slightly higher amplitude is observed. This is referred to as the first echo peak and corresponds to the part of the ultrasonic signal that has travelled all the way through the cell and hit the back wall of the cell.

Internal temperature and defects

To determine how the temperature of the cell affects the waveform generated by ultrasonic analysis, a 210 mAh pouch cell has been set up in an environmental chamber and the acoustic signal has been monitored as the temperature varies according to the profile shown in Fig 2a. To investigate the cell behaviours in mildly abusive conditions, the temperature has been varied beyond the window temperature assigned by cell manufacturers (0 - 40 ° C). A colourmap plot of how the acoustic signal changes during this period is shown in Fig. 2b. This plot is a top-down view of how the typical signal varies with temperature. Each line shows a peak with the colours indicating the amplitude and its position in the y-direction indicating the ToF.

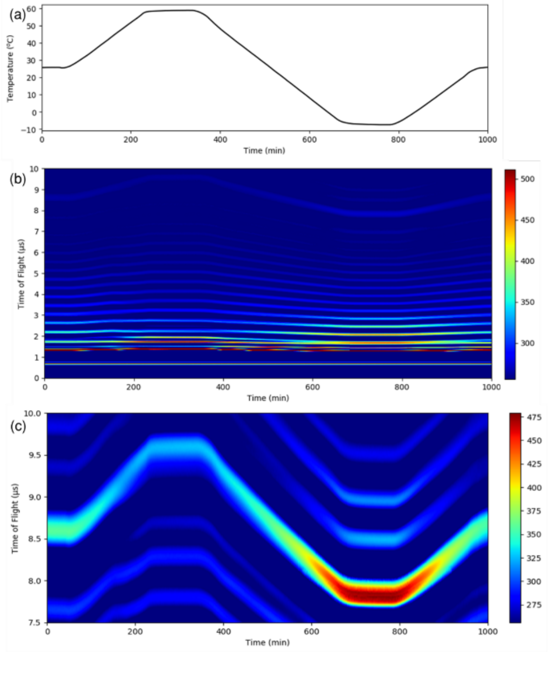

Figure 2 - (a) Cell temperature over the course of the experiment. (b) colourmap plot showing acoustic waveform variation. (c) Magnification of colourmap plot for the first echo peak ToF

As the temperature is initially held constant, no variation in amplitude or ToF is observed with any peak. During heating, the ToF of each peak increases due to the individual anode and cathode layers expansion, causing the entire cell to swell accordingly. A linear correlation between the first echo peak ToF and the temperature is observed proving the sensitivity of the acoustic measurements to ambient and internal temperature variations. Small deviations from the predicted linear relationship are observed at temperatures exceeding the manufacturer's limits. This may represent a powerful tool for predicting the onset of irreversible cell degradation and TR.

To determine the ability of acoustics to detect any defects that may be present in cells or formed during operation/abuse, acoustic tests have been run on two 400 mAh pouch cells. One is a pristine cell whereas the other had been cycled multiple times during which a sudden failure had occurred. Typical waveforms obtained from acoustic experiments are shown in Fig. 3. The pristine cell shows a high amplitude transmission peak whereas the defective cell also shows the transmission peak at the same ToF, however the amplitude has dropped significantly. These results suggest the presence of a defect since the formation of any gases would inhibit good acoustic contact between the electrode layers, increasing the signal attenuation. Later X-ray computed tomography experiments show evidence of internal layer separation, confirming how small internal defects can lead to significant changes in the acoustic signal.

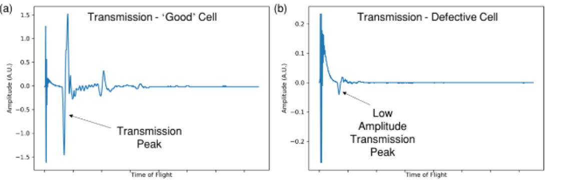

Figure 3 – Transmission results for 'good' and 'defective' cells

Cell and module TR tests monitoring

To acoustically monitor TR in individual cells and its propagation through a module or pack, a reliable, repeatable, and representative method of EVs in real-world applications for triggering TR is required. Nail penetration, overcharge as well as homogeneous and localised heating have been investigated and monitored through acoustic transducers. While repeatable and consistent acoustic signals are observed for cells operating under normal conditions, when the cells are pushed outside these limits, clear changes in acoustic signals are visible. During the homogeneous heating test, at lower temperatures, additional acoustic peaks are observed indicating the appearance of new interfaces. At high temperatures, the ToF of the first echo peak shifts quite drastically before disappearing completely at ca. 100 ° C, when the electrolyte likely evaporates.

Importantly, all these distinct and measurable changes occur long before the cell enters irreversible TR.

Homogeneous heating of the cell is less representative of the type of thermal abuse that would likely occur in a real-world application caused by an internal short, where higher thermal gradients would be more localised within the cell. To make the tests more representative of real-world scenarios the test has been adapted to a local or point heating experiment where a heating cartridge is used to heat a small area of the cell to represent the area where a short is occurring. A fully charged cell has been heated from the bottom of the cell with a heating cartridge, while an acoustic transducer is placed on the top of the cell. During the experiment the voltage remains relatively consistent until the cell enters TR, proving that monitoring cell voltage does not reveal damaging processes occurring within the cell. During initial heating, while there is little to no change observed in the temperature recorded by the thermocouple, significant changes in the acoustic signal are observed. About 500 seconds before the cell enters TR the echo peak (close to the heating cartridge) disappears, followed sequentially by peaks at lower ToFs. Conversely, readings from the thermocouples only show a mild temperature rise proving the superior sensitivity of acoustics to localised internal cell damage.

For high-power operations, battery modules or packs are used. Understanding the operation of these and the inherent risks associated with operating many cells together is of vital importance. Failure of a single cell can easily trigger the rapid heating and consequential failure of neighbour cells causing a dangerous thermal propagation chain reaction. To study the effectiveness of ultrasound in pack applications, a small four-cell demonstration module has been constructed (Fig. 4). The cells are placed in a side-by-side arrangement with acoustic transducers placed on each cell and thermocouples on the top and bottom of each cell. Initially, all cells are held at room temperature for ca. 400 seconds. Then, the heating cartridge in contact with the base of Cell 4 is switched on. The results of this test are summarised in Fig. 5, with the acoustic behaviours of all four cells shown along with the temperature of the bottom of each cell. During rest at room temperature, no shift in any of the acoustic signals is observed.

As soon as the localised heating on Cell 4 is initiated, the acoustic signal of this cell begins to change. For clarity, the change in ToF of the first echo peak has been plotted for all cells in Fig. 6 and compared to the temperature of the top and bottom of the cell. During heating, the ToF of the first echo peak of Cell 4 rapidly increases beyond any change that would be expected under normal operations.

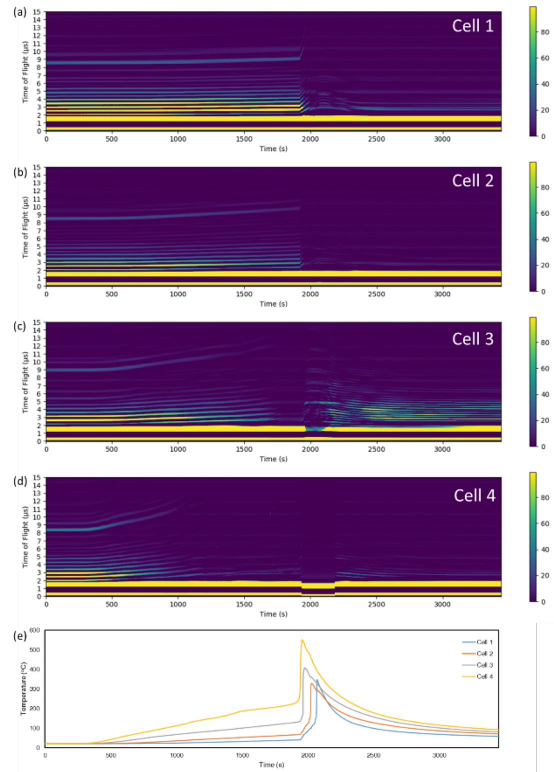

Figure 5 - The acoustic behaviours of Cell 1 (a), Cell 2 (b), Cell 3 (c) and the thermally abused cell 4 (d) are shown as well as the temperature of the bottom of each cell (e) during the TRP test

After ca. 1000 seconds the peak amplitude drops and the signal is lost (Fig. 6d). Meanwhile, the top of the cell is at 70 ° C and the bottom at 117 ° C, enough for the electrolyte to evaporate. At this point the acoustic signal has indicated that a significant thermal event and/or thermal damage has occurred to Cell 4. The cell, however, is heated for further 900 seconds (15 minutes) before the cell enters TR. After cell 4 enters TR, other cells also enter TR in short time.

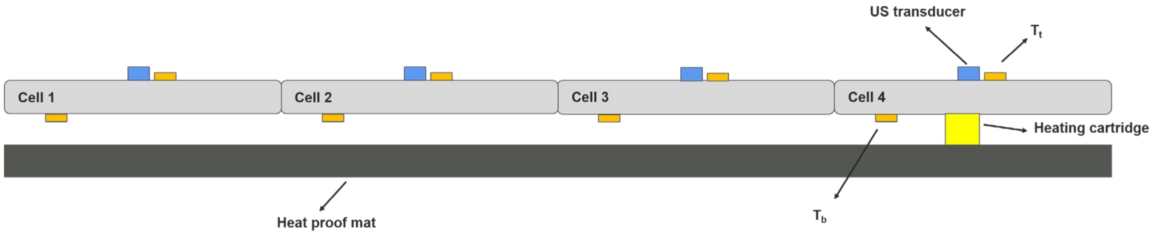

Figure 4 – Lab-scale module used for TR propagation studies

Cell 3 follows similar dynamics and considerations to cell 4 with the acoustic sensor showing significant damage well before the cell enters TR. For the farthest cells (cells 1 and 2) the signal is lost only when they undergo TR, indicating that they were still relatively undamaged until the flames coming from Cell 3 and 4 reached them.

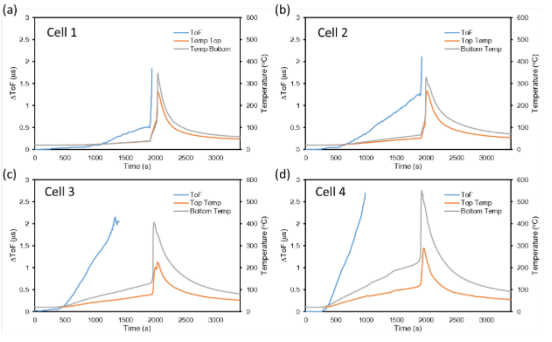

Figure 6 - The temperature and ToF variation of the module cells (a) Cell 1, (b) Cell 2, (c) Cell 3, (d) Cell 4

Conclusions

Ultrasound has been demonstrated to have the ability to access internal states and changes of battery cells, detect rapid heating and internal defects, and provide early TR warning up to several minutes before conventional methods based on voltage and temperature readings. Propagation of TR through a small module of cells has been monitored using multiple acoustic sensors with the acoustic readings flagging internal damage long before the first cell failure and showing clearly identifiable patterns for neighbour cells too. Remarkably, the acoustic measurements indicate the presence of internal damage 900 seconds before the cell entered TR. With the new mandatory GB standards introduced in China specifying that a battery system must provide a warning to allow egress 5 min prior to the presence of a hazardous condition that could lead to TR (*GB 18384-2020*), these results are certainly promising. With regards to onboard battery pack applications, to take the technology forward on the journey to mass-production, more comprehensive ultrasonic tests on a wide variety of batteries, looking across the complete range of operational variables shall be performed and a cost-effective integrated ultrasonic transducer system will need to be developed for each class of cell/battery. An analysis also needs to be made as to what proportion of cells within the battery pack needs to be monitored. As an immediate application, thanks to its unique ability to identify steps leading to TR, ultrasound could be incorporated into battery abuse testing to shut down testing safely and prevent irreversible TR to occur. Overall, these results open many opportunities to sensor suppliers but also industries relating to the testing, certification, simulation, and design of battery packs for EVs.

Acknowledgments

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