Modelling of thermal events in Lithium-ion batteries

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Abstract
Lithium-ion batteries are seen as part of the solution to meet the environmental concerns for many areas including the automotive sector. The Li-ion technology has many good properties such as a high energy-density but also drawbacks such as its narrow window of stable operation. If the cell is e.g. heated up it might go into a thermal runaway in which the cell rapidly heats itself up, a process that might spread also to adjacent cells. In order to investigate whether a thermal event will progress to adjacent cells, it is important to be able to model the heat transport within a battery module properly. A first attempt to model the spreading has been made using Comsol Multiphysics for a test case where one cell is exposed to a heating source and then the heating spreads to other cells.

Introduction

Lithium-ion (Li-ion) batteries offer great performance in form of e.g. energy and power densities, enabling their use for a wide range of applications including the use in electrified vehicles, an application that is steadily growing. If the temperature in a Li-ion cell is increased beyond a certain threshold, a thermal runaway can occur, resulting in a rapid temperature increase and possibly other adverse effects such as, release of gas, smoke, fire and rupture/explosion. There are numerous types of abuse situations that can result in elevated temperatures inducing a thermal runaway. For example, mechanical abuse, electrical abuse and thermal abuse [1-11]. In case of a thermal runaway in a single Li-ion cell it is important to stop or at least delay its spreading to adjacent cells, since the effects from a cascading thermal runaway scenario of a complete battery pack could be devastating. Battery packs in electrified vehicles can consist of thousands of battery cells. In general, the design of a battery pack plays an important role for fire propagation, e.g. the thermal management system, mechanical support structures between cells and modules, possible use of fire walls and division of the battery pack into different sub-packs are important aspects.

There are relatively many simulation studies regarding general heat generation and cooling of Li-ion batteries during normal cycling (battery charge/discharge) within the battery specifications. Furthermore, Li-ion cells and battery packs with multiple-cells have been simulated during abusive conditions; by external heating in oven, short circuiting, overcharging and deformation/crash. For example, Spotnitz et. al. [12] performed a numerical study on the influence of various heat transfer modes for the propagation cell-to-cell for a battery pack with 8 cylindrical cells of type 18650.

However, only a limited number of simulations studies of lithium-ion battery fire abuse situations are available. For example, Anderson et. al. [7] used CFD simulations with subsequent thermal modelling to study the fire resistance of a battery pack in a gasoline pool fire test according to UNECE Regulation No. 100 in the EU project Smartbatt. The purpose of that modelling work was however primarily focused on evaluating the casings possibility to protect the battery from the 2 minutes gasoline pool fire as required in UNECE Reg 100.

The work has then continued in smaller scale considering both the external heating and the heat transport from cell to cell. Tests have been conducted on five Li-ion pouch cells stacked on top of each-other and exposed to a propane flame from underneath and a first attempt to model the heat propagation from of the cell-to-cell was conducted Anderson et. al [8]. The simulations have so far been able to predict the temperature profiles to some extent, but further work is needed and ongoing. This paper presents tests performed on a mock-up to get a better characterization of the flame, including heat transfer coefficient, to rule out any errors in the boundary conditions and heat exposure of the cells. The mock-up test has been simulated and verified against the test and the previous simulations have been updated with the correct boundary conditions and heat transfer parameters.

Heat Exposure of Li-ion cells

Commercial Li-ion (EiG ePLB-F007A 7 Ah) pouch cells were exposed to a 15kW propane burner that initiated a thermal runaway event in the cell closest to the burner. During the tests heat release rates (HRR) and temperatures, measured by thermocouples between the cells, were measured. The resulting HRRs have been published as well as one preliminary study on modelling one of the tests [6, 8]. The tested EiG cell had a lithium-iron phosphate, $LiFePO_4$, cathode and a carbon based anode. Each of the tests consisted of five cells tightly packed together with steel wires as seen in Figure 1. The cell terminals (tabs) were cut-off prior to the tests for all but the middle cell.

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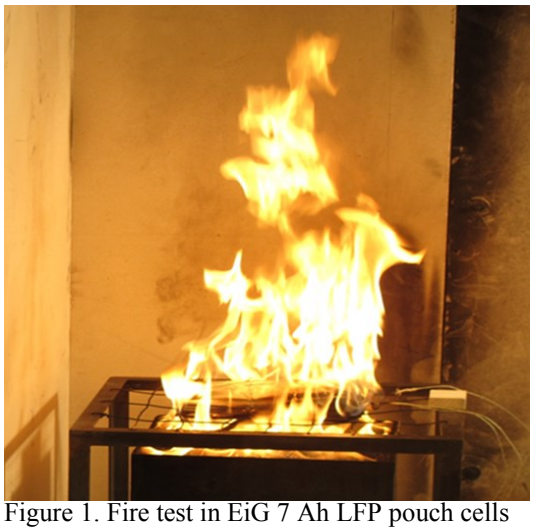

Figure 1. Fire test in EiG 7 Ah LFP pouch cells

Several tests with varying state of charge (SOC) were conducted yielding cases with varying reactivity. Figure 2 shows the HRR values for SOC levels between 100 % and 50 %. The propane burner is started at 2 minutes and the contribution from the burner to the HRR is subtracted. The modeling in this paper is based on the 75% SOC case, which is indicated by the red dashed line in Figure 2, while in Anderson et. al. [8] the case with 100% SOC was used. For 100% SOC the HRR shows energetic peaks, as seen in Figure 2, which is not seen for lower SOC values. During the test temperatures between some of the cells were carefully monitored in four positions as indicted in Figure 3. The measured temperatures from the test with 75% SOC, the test case that will be simulated here, are presented in Figure 4.

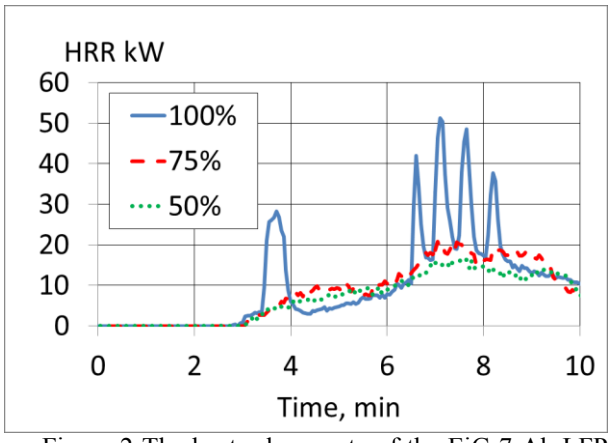

Figure 2 The heat release rate of the EiG 7 Ah LFP pouch cells where the heat from the propane burner is subtracted.

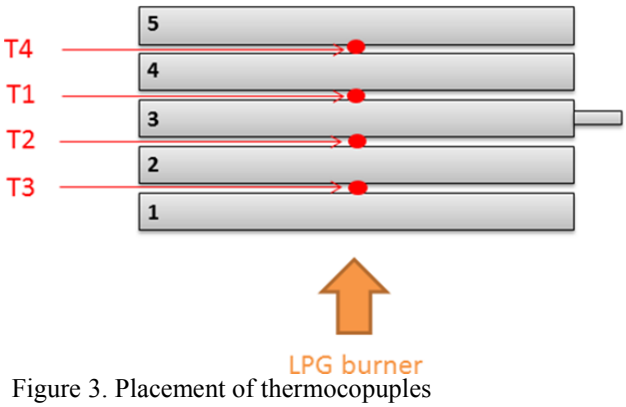

Figure 3. Placement of thermocopuples

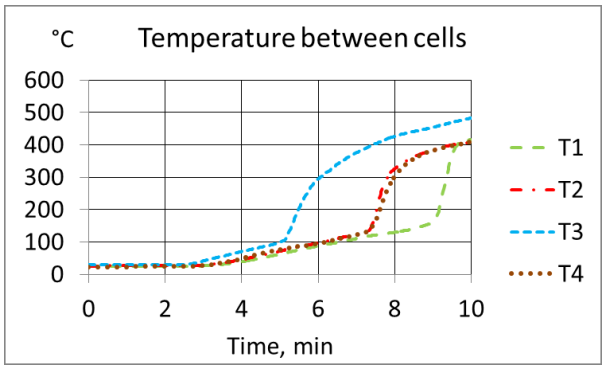

Figure 4. The measured temperatures between the cells.

Mock-up test

In the previous paper by Anderson et al [8] it was identified that a well-defined fire source is needed in order to more accurately describe the evolution of the temperatures between the cells. For modelling purposes one test with a mock-up was performed where the impact of the burner was assessed. The mock-up as shown in Figure 5 was constructed of Promatect clad with custom made plate thermometers. Although the moisture level in Promatect is very low it was dried for around 48 h in order to further reduce the moisture content. The plate thermometers were approximately 40 mm x 100 mm in size to fit on the side of the mock-up. The objective of the mock-up test is to estimate the fire source's impact on each side of the mock-up by measuring the adiabatic surface temperature (AST). The AST is an artificial effective temperature that replaces the gas and radiation temperature that describe the local conditions around the test object. It is defined as the temperature of a surface that cannot absorb any heat. However, due to the limitations of the physical extensions of the object the size of the plate thermometers were smaller than usual indicating that they may experience somewhat different convection radiation equilibrium since the estimation of the AST becomes more sensitive with smaller devices.

Figure 5. Mockup for determining thermal impact

Mock-up simulations

The simulation work was conducted along a similar path to the work in Smartbatt [7], i.e. the fire source was modelled with Fire Dynamics Simulator 3FDS [13] to determine the gas temperatures around the cells and the heat transfer coefficient. Then the gas temperatures and heat transfer coefficients as determined in the FDS simulation were used to simulate the temperature in between the cells as indicted in Figure 3 in a COMSOL model.

The FDS software solves the Navier-Stokes equations in the limit of low-speed, thermally-driven flow with an emphasis on smoke and heat transport from fires. The algorithm used is an explicit predictor-corrector scheme that is second order accurate in space and time where turbulence is treated by means of Large Eddy Simulation (LES) in the Deardorff form. The FDS software with default settings uses structured, uniform staggered grid in order to utilize the efficiency of the Fast Fourier transforms in the pressure solver. The combustion chemistry is simplified and a generalized lumped species approach together with the eddy dissipation concept is used for a single step reaction between fuel and oxidizer. In the default setting radiation is calculated using 100 discrete angles in a finite volume approximation of the radiation transport equation with gray gas. The FDS model is not limited to these simple algorithms however any additional physics included incur increased computational costs. The default model options have been selected based on results from a wide variety of full-scale validation experiments [13].

The propane fire source was modelled in a 5*5*5 cm^3 grid using default values for propane with 6*6 cells over the burner surface. The fire source was modelled using the HRRPUA option in FDS, i.e. the HRR per unit area over the burner surface was defined. The gas temperatures on each side of the mock-up were monitored together with the heat transfer coefficient, the results of which are presented in Figure 6 and Figure 7 respectively.

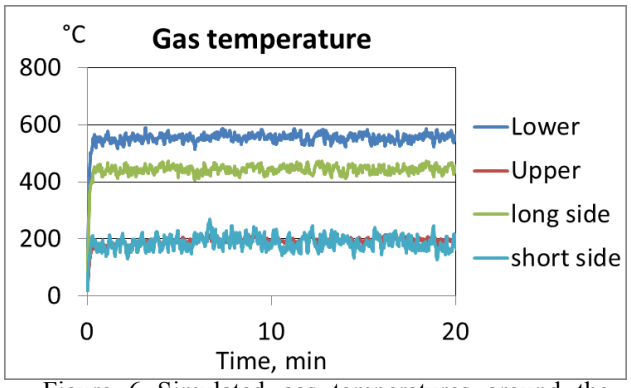

Figure 6 Simulated gas temperatures around the mpck-up

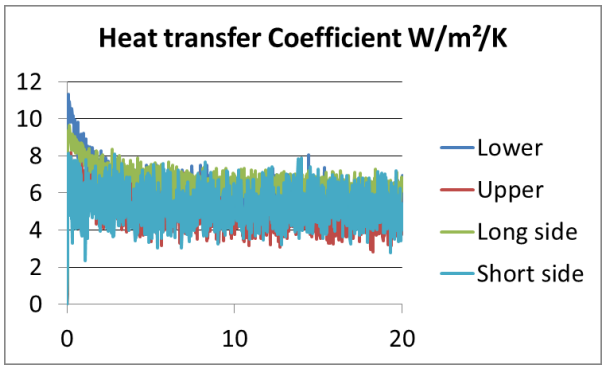

Figure 7. Simualted Heat Transfer Coefficient

The gas temperatures and heat transfer coefficients found in the modelling were then used in a COMSOL model of the mock-up. The heat transfer to the mock-up was modelled assuming radiation from the surrounding gas with emissivity=1 and convective heat transfer from the surrounding gas with the heat transfer as calculated by FDS and the gas temperatures as calculated by FDS. The thermal conductivity k = 0.099W/mK, density ρ= 280kg/m^3 and C_p = 2200J/kgK of the promatect were taken from previous measured data at 100 ˚C. The model includes thin Inconel plates on the side to simulate the conditions of the experiment, however to have reliable results at the plates an increased number of grid cells are needed at these locations. In total the model consists of approximately 70000 cells. The result from the simulation is shown in Figure 8. As seen the comparison is reasonable on the top and bottom side while the temperature on the sides is underestimated. This is probably due to that in the test there was a distance between the steel plates and the Promatect on the sides which allowed much higher heat transfer to the steel plates on the sides.

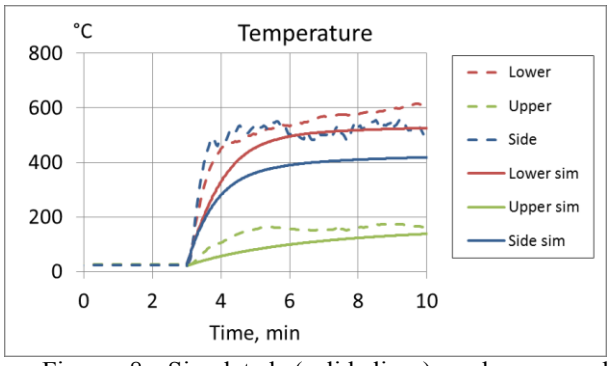

Figure 8. Simulated (solid lines) and measured (dashed lines) temperature from the mock-up test.

Material data

The physical structure of the pouch cells is complex with a repeatable layering of anode and cathode material. The total numbers of layers is of the order of 150 layers. The thermal properties of the materials vary significantly in terms of thermal conductivity, density and specific heat along and across the cell due to the layered structure. To be able to accurately compute the temperatures in the 5-cell-pack realistic values of the thermal properties are needed in combination with a well-defined fire source in terms of the measured heat release rate. This complex structure of the cell makes detailed modelling impractical however for our purposes bulk values of the thermal properties are enough to capture the essential propagative features. In the present study the values of Wu et. al [14] are used. Ideally, careful measurements of these quantities at elevated temperatures are needed but generally difficult to obtain. The heat conductivity coefficient, k, was varied in the simulations since the multi-layered structure of the battery cell allows for this value to be highly anisotropic. Table 1 shows the thermal material properties of the cells used in the thermal simulations.

Table 1. Material Data

Density ρ [kg/m^3]	Specific heat Cp [J/(kg K)]	Thermal conductivity k [W/(m K)]
1895	1243	kx = 21 and kz = 0.48

Battery simulations

The model was built in the multi-physics software COMSOL suitable for studying heat transfer problems. In the model the five battery cells was implemented with bulk values of density, specific heat and thermal conductivity. The test specimen was modelled in 3D with half the amount of simulated cells. The simulation time is only a few minutes. In the model, the battery cells are in the same ambient conditions (T0) as the surroundings of that particular test and the cells are assumed to always be in perfect contact to each other. The ambient conditions give the boundary conditions for the thermal exchange between the cells and the surroundings by conduction, convection and radiation. The incident heat flux form the propane flame is modelled as for the mock-up case using the gas temperatures and heat-transfer coefficients found in the FDS simulation together with emissivity 1 for incident radiation from the surrounding gas. In addition was the measured heat release rate from the battery cells themselves used as input in the model for the thermal energy which was deposited homogenously into the bottom cell.

Results

The result from the simulation is shown in Figure 9 where the temperature range of most interest is shown, i.e. up till the onset of the thermal runaway around 120°C. As seen the temperature is over-predicted in most cases, i.e. on the safe side as compared to the results from Anderson et al. [8], but still the temperature profiles are different from the experimentally measured values.

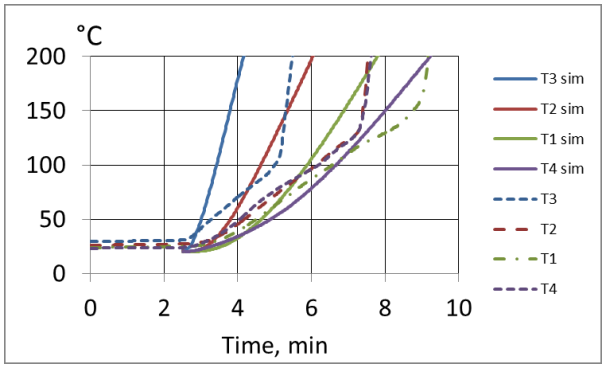

Figure 9. Comparison between experimental results (dashed liens) and simulated temperatures (solid lines) at 75% SOC using the thermal model. A comparison of temperatures T1, T2, T3 and T4 found in the experiment and the simulation assuming anisotropic heat conductivity (kx =21 W/(m K) and kz = 0.48 W/(m K))

The temperature rise is higher in the simulations as compared to the measured values. In the experiments however the rate of temperature rise changes suddenly when the temperature has reached a value between 100 and 150 °C. This rapid change in temperature could be due to that the contact between the cells and the thermocouple changes as the cell swells and the heat can reach the thermocouple also from the side. This is a feature that the simulations cannot capture.

Conclusions

This paper presents an attempt to predict the progress of a thermal runaway/fire in a cell to neighbouring cells by simulating the temperature development on neighbouring cells. Simulations and experiments have been conducted for varying SOC, this paper focuses on the case with 75% SOC as an example of results.

Modelling battery cells in such harsh conditions as that of a fire is a rather difficult task due to the lack of knowledge of the precise local conditions around the test object. This gives an uncertainty in the boundary conditions to be used. In this paper the boundary

conditions were estimated from a mock-up test where the temperature on the upper, lower and sides of a Promatect object of about the same size of the cells were used. The mock-up had larger exposed sides than the cells and thus experienced a larger heat transfer. On the same token the cells allowed for gases reaching in between the cells which would increase the heat transfer in the cell case. Comparing the results from the heat transfer simulations with the experimental data it seems however that the heat transfer is higher in the simulations than in the experiment.

Another difficulty with the simulations is the lack of knowledge of the material data. In order to have a reliable model the precise temperature dependency of the heat conductivity, specific heat and density are needed.

The simulations are in reasonable agreement with the experimental result up to the point where the cell starts to swell and a thermal runaway is initiated but still the simulations over-predict the temperature in the neighbouring cell somewhat in all cases. However the model can be further developed by:

1. Investigate in more detail different boundary conditions. Since the dimensions of the battery pack is rather small the influence of the boundary is important and can be a source for uncertainties. Both geometry at the edges and different forced convection boundaries should be investigated. In addition should radiation losses be taken into account.
2. Simulating different contacts between the cells using a thin contact element with variable thermal resistance should be investigated.
3. More adequate values of the thermal properties data including temperature variation.

With these improvements there is potential for the simulations to be able to better predict the temperature development in a neighbouring cell.

Acknowledgements

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