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Understanding and Modeling the Thermal Runaway of Lithium-ion Batteries

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Executive Summary

The main safety issue pertaining to operating Li-ion batteries is relating to its sensitivity to thermal runaway. This project aims to go deeper into the understanding & modeling of this complex multiphysics phenomenon at cell scale, taking into account the influences of novel highly reactive technologies and ageing (calendar and due to use) with 2 target degradation mechanisms: SEI formation/growth and Lithium Plating.

In this presentation, the methodology of investigating the thermal runaway through experimental study are presented. The obtained results will be used to calibrate & validate the 3D extended Thermal runaway model in the future works of the project.

1 Project introduction

Safety control of Lithium-ion (Li-ion) battery is essential. All the more energy/power demanding applications act as incentive towards highly reactive Li-ion batteries. Already dominating the consumer market, Li-ion batteries are expected to dominate also e-mobility and stationary applications in near future [1]. Incidents involving batteries undergoing thermal runaway are still recurrent and highly advertised by the media [2]. The thermal runaway is characterized by a deficit of energy evacuation versus energy accumulation in the cells leading to uncontrollable overheating of the battery system. This complex event involves multi-scale phenomena ranging from internal physico-chemical to battery components reactions (electrodes, electrolytes & separator) and further to the thermal propagation of cell core & safety devices (CID, PTC disk, Pressure disk, Button vent, steel can).

In the context of emerging higher capacity Li-ion batteries including Ni-rich high energy technologies, lithium plating also seems to have a notable impact on the behavior of lithium-ion cells in thermal abuse conditions through increased risk of short circuit, and hence higher sensitivity to thermal runaway [3,4].

Former IFPEN/INERIS project on the thermal runaway of Li-ion batteries has focused on the previous generation of Li-ion batteries (essentially LFP/Graphite) [5,6] which has shown that thermal runaway is triggered through a series of exothermic reactions which occur in cascade at higher or lower temperatures [6]. The work led to the development of a methodology to model the thermal runaway at cell scale and

subsequently to the prediction of the battery state of health. It was also found that SEI Growth driven calendar ageing seems to play a critical role in thermal runaway. However, the impact of SEI driven cycling ageing was not covered.

Accordingly, this project aims to go deeper into the understanding & modeling of the thermal runaway phenomenon of Li-ion batteries at cell scale, taking into account the influence of novel highly reactive technologies and the influence of ageing with 2 target degradation mechanisms: SEI formation/growth and Lithium Plating, in order to understand what are the keys to inherently safer highly reactive Li-ion batteries during usage.

3 main work packages (WPs) in the project have been defined and started:

- The first WP deals with the study of the effect of high energy density technologies on safety: Ni-rich positive electrode materials with a novel negative electrode technology: SiOx-Graphite Composites.
- The second WP aims at understanding underpinning mechanisms and relationship between calendar & cycling ageing and safety focusing on 2 target degradation mechanisms: SEI formation/growth as a function of temperature & Lithium Plating (under low temperature conditions).
- The final WP deals with the development of a consolidated thermal runaway 3D model and the implementation of test protocols for requested calibration and validation purposes of the model.

This three step research strategy relies on the achievements of several previous completed projects and on the combination and synergisms offered by combined experimental and modeling studies:

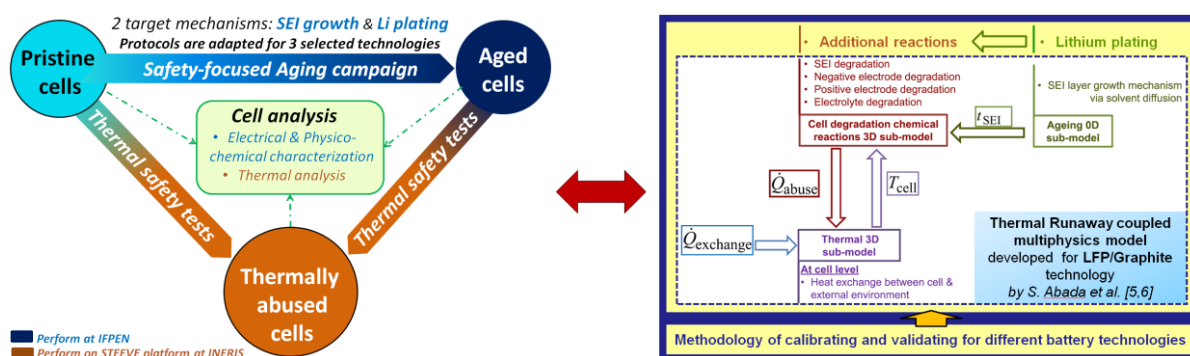


Figure 1: Project strategy: Combination of Experimental study (left) & Modeling study (right)

The modeling study is included in the final WP of the project which leads to the development of a 3D extended thermal runaway model in order to predict the behaviors of different Li-ion batteries nearby and during thermal runaway. This coupled multi-physics model will improve & extend the previous thermal runaway initial model build at IFPEN [6] by integrating the impact of lithium plating & SEI-driven cycling ageing.

At this stage, the first part of our project which is defining the methodology to investigate the thermal runaway through experimental study has been achieved and will be presented. This obtained results will be used to calibrate & validate the 3D extended Thermal runaway model build in the future WP of the project.

2 Methodology for thermal runaway investigation through experimental study

In this presentation, we will focus on the methodology of the experimental study. This study includes a safety-focused ageing campaign in order to artificially age battery samples, focusing on each target mechanism in a controlled and measurable way, the thermal safety tests in order to perform and calibrate the thermal runaway test. Pristine, aged and thermally abused cells will be analyzed by a complete multi-scale cell analysis.

2.1 Multi-scale cell analysis:

The cells will be analyzed at all stages (pristine, after certain level of ageing and after thermal runaway). Thermal safety test will also carry out the analysis of the composition of gases and particles emitted.

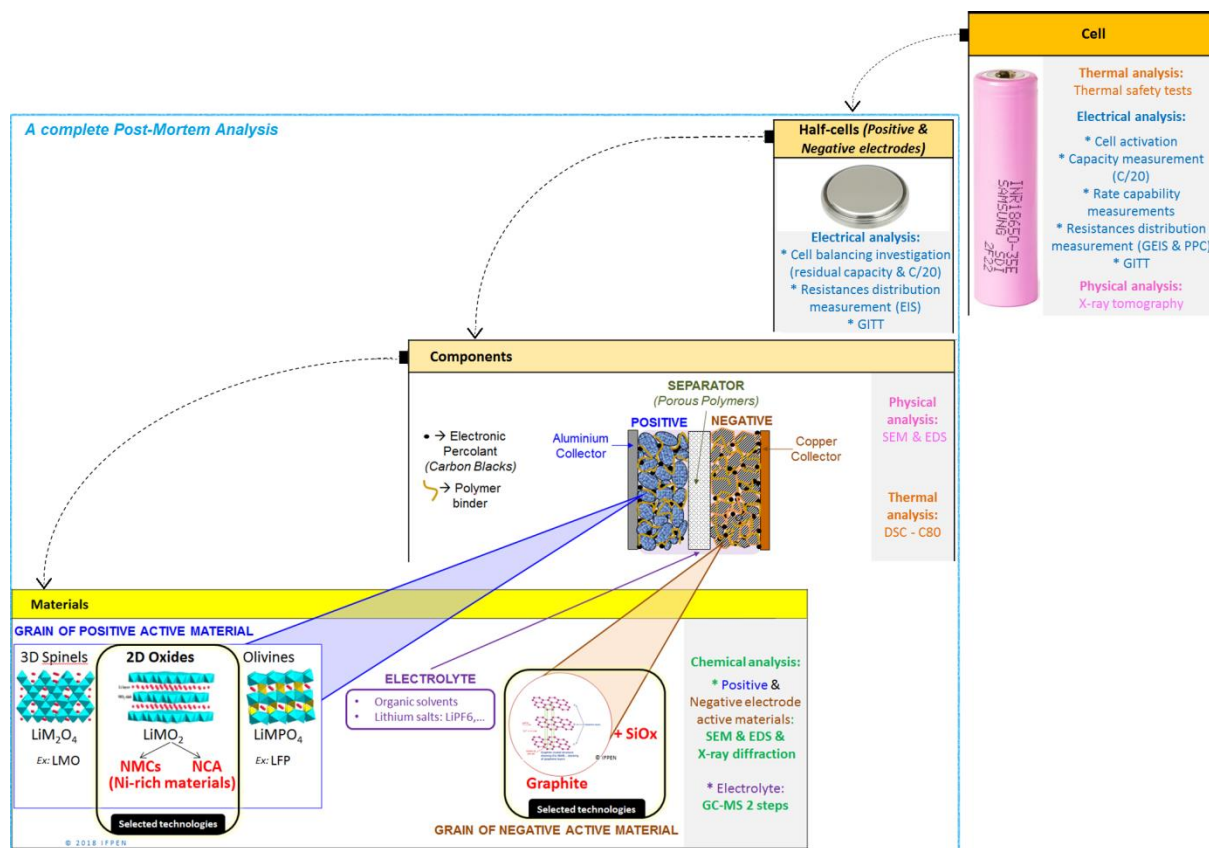


Figure 2: A complete multi-scale analysis of Li-ion battery cell

2.2 The impact of technology on Safety:

We will study different novel technologies of the new Lithium-ion battery generation. We selected 18650 Ni-rich high energy technologies:

SAMSUNG INR18650-35E, Panasonic NCR18650 GA and LG 18650 HG2.

A complete analysis are performed on pristine cells in order to identify the cell chemistry. The choice of Li-ion batteries technologies studied is well confirmed.

2.3 Safety-focused Ageing campaign:

Several pretests have been carried out to define a complete safety-focused ageing campaign with 2 target degradation mechanisms. Lithium plating ageing protocol is the cycling conditions at low temperature which accelerate the occurrence of lithium plating and SEI-driven ageing protocols are the storage/cycling conditions which accelerate the formation/growth of SEI but also minimize the occurrence of lithium plating (at high temperature). These protocols are customized to different technologies studied and to each other in order to have a comparable results.

Non destructive techniques of qualification and quantification of ageing state are also studied [7,8].

2.4 Thermal safety tests:

Pristine and aged cells obtained from the ageing campaign, in known and quantified degradation states, will undergo thermal safety tests on the STEEVE platform of INERIS. These tests will make it possible to

understand the processes involved in the thermal runaway of the batteries and to calibrate and validate the developed models. To go further in understanding the phenomenon, cells that have undergone runaway tests will also be analyzed, post-mortem.

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