32nd Electric Vehicle Symposium (EVS32)
Lyon, France, May 19 - 22, 2019

Understanding and modeling the Thermal Runaway of Lithium-ion Batteries

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Summary

Safety control of Lithium-ion battery (LIB) is essential. 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 relating properties of novel highly reactive technologies and aging (calendar and due to use), considering both SEI formation/evolution and lithium plating degradation mechanisms.

In this presentation, the methodology of investigating the thermal runaway through experimental study are presented. It has not only explored the effect of high energy Ni-rich LIBs on the thermal runaway but also investigating the underpinning mechanisms and relationship between calendar/cycling aging and safety. The experimental results will be used to calibrate & validate the 3D extended Thermal runaway model in the future research works.

Keywords: battery, safety, battery aging, battery model, energy storage

1 Introduction

Lithium-ion batteries (LIBs) is one of the most important energy storage technologies today thanks to their high specific energy densities and stable cycling performance [1, 2]. The challenging requirements for LIBs technology are i) targeting lower cost systems, ii) achieving higher performance with a long life-time (> 10 years for automotive applications), iii) allowing fast-charging (< 20 min for 80 % SOC), and iv) bearing low temperature cycling. In the meantime, all these expected improvements shall not compromise safety performance which must remain excellent in all situations, i.e. during the whole life-time including all weather and abuse conditions [3]. However, if Li-ion batteries are operated improperly, outside of the specification of its manufacturer or due to cell defects, electrical and chemical energies inside the cells can

be unintentionally released and lead to gassing, fires or even explosions. During these incidents, the most energetic catastrophic failure of a LIB system is the cascading thermal runaway event. It is characterized by a deficit of energy evacuation versus energy accumulation in the cells leading to uncontrollable overheating of the battery system. In general, this energetic failure occurs when an exothermic reaction goes out of control. As the temperature of the battery rises to a certain threshold, the exothermic chemical reaction rate inside the batteries increases and further heats up the cell. The continuously rising temperatures may trigger cascading chain reactions [4, 5] and result in uncontrolled flammable and toxic gassing, and/or fires and explosions, especially critical for large battery packs.

Since the commercialization of LIBs by Sony Inc. in 1991 until today, the recurrent incidents involving LIBs undergoing thermal runaway have been reported worldwide: on electronics devices such as cell phones (Samsung Note 7…), laptops…, electric vehicles (various trade models including the Tesla) and even auxiliary power units (APU) powering commercial aircrafts [6]. Although these incidents are highly unlikely, they are reminders that safety is a prerequisite for batteries, whatever the level of innovation, and understanding the causes and process of thermal runaway of high energy LIBs before their applications is essential to guide the design of functional materials and improve the safety & reliability of LIBs.

Battery safety becomes even more critical with the emerging of highly reactive Ni-rich Li-ion batteries into the market. They are commercialized to meet the novel energy/power demanding applications and even expected to dominate the market in the coming years, until the occurrence of a new technological breakthrough. This novel battery generation of such high energy density and more intrinsically reactive materials can possibly lead to more catastrophic events involving thermal runaway. Inspired by the works of Abada et al [7–9], our research aims to go deeper into the understanding & modeling of the thermal runaway phenomenon of Li-ion batteries at cell scale, taking into account new generation high energy cells influencing properties as well as the aging phenomenon as resulting into SEI evolution and Li Plating degradation mechanisms. The final aim in our work, supported by combined experimental and modeling approaches, is to find out the keys to inherently safer highly reactive Li-ion batteries during usage.

At this stage, we have defined, and started to deploy a complete experimental methodology that can fully investigate the thermal runaway of LIBs. It establishes the link between the battery technology (cell design, the materials used in the electrodes, electrolyte, separators, …), the degradation products during cell aging (mainly SEI evolution during cell lifetime and Li deposition during cold charging) and the thermal runaway kinetics.

2 Experimental methodology of investigating the thermal runaway

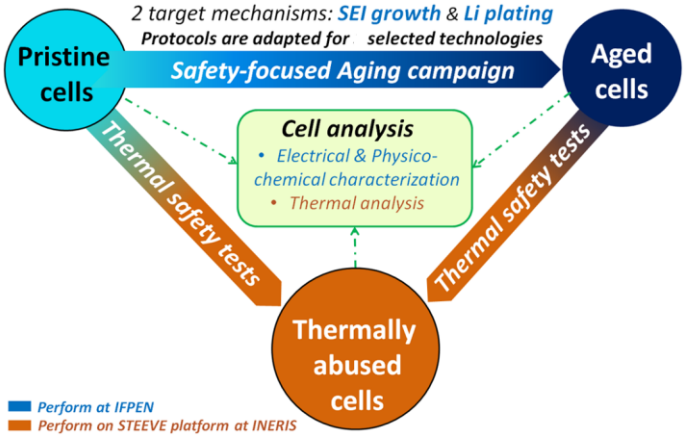


Figure1: Research experimental methodology

The complete methodology (model validation excluded) includes

- the technology selection
- a complete multi-scale cell analysis in order to analyse the pristine, aged and thermally abused states of LIBs,

- a safety-focused aging 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 understand the thermal runaway phenomenon.

These interconnected experimental processes are illustrated in Figure1.

The complete multi-scale cell analysis will support the interpretation of the 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 features (CID, PTC disk, Pressure disk, Button vent, steel can) involved in the thermal runaway process.

The safety-focused aging campaign and the thermal safety tests aged cells will explore the role of aging on the thermal runaway, especially the impact of SEI evolution aging occurring throughout cell lifetime and Li deposition/plating aging occurring during cold recharges.

Both detailed aspects of these experimentally supported analyses will help to adjust and upgrade as needed the multiphysics modeling tool under COMSOL so far calibrated for LFP cells developed by [7].

2.1 Technology selection

With the massive commercialization of electric vehicles, the supply of Ni-rich LIB technologies today presents less difficulties than before. The size (in term of capacity) & format of a cell can have a significant effect on safety behaviour [6]. For simplification, the cell format studied in this research is fixed as the cylindrical 18650 cell format because:

- cylindrical format is one of the most common cell formats and basic phenomenology of the same provided battery chemistry is the same while difficulties may arise from mechanical aspects;
- 18650 size: 18650 cells (e.g. from Japanese/Korean manufacturers) are currently widely used in consumer and EV cars products for quite a while and according to international regulations for transport of dangerous goods (UN TDG Model Regulations) are subject to reportable control quality procedures. Therefore, functional and safety performance repeatability could be assumed as very representative of best products currently available on the market. Another aspect is that the detection of a temperature rise could be easier in case of small cells, since only one or two sensors (thermocouples) might be enough. In contrast, for larger cells, more sensors are needed.

Two 18650 Ni-rich high energy technologies from the new Li-ion battery generation: LG 18650 HG2 and Panasonic 18650 GA have been selected. A complete analysis has been performed on pristine cells in order to carefully check the cell chemistry and thereby to confirm the choice of highly reactive Ni-rich Li-ion batteries technologies studied. LG HG2 and Panasonic NCR GA base on NMC811 and NCA, respectively, as the positive electrode active materials and Graphite-SiO_x composite technologies as the negative electrode active materials. More details can be found in Table 1.

Table1: Selected Ni-rich commercial batteries

	LG 18650 HG2	**Panasonic NCR18650GA**
Cell chemistry:	**NMC811/(Graphite-SiO_x)**	**NCA/(Graphite- SiO_x)**
Nominal Capacity:	**3000mAh**	**3450mAh**
Charging voltage:	4.20V+/-0.05V	4.20 +/-0.03V
Cut off voltage:	2.5V	2.5V
Standard charge:	1500mA (C/2)	1725mA (C/2)
Standard discharge:	600mA (C/5)	690mA (C/5)
Operating temperatures (from manufacturer):	Charge 0 °C ~ 50 °C Discharge: -20 °C ~ 75°C	Charge: +10 ~ +45 °C Discharge: -20 ~ +60 °C

2.2 Multi-scale cell analysis

The cells will be analysed at all stages (pristine, after predefined levels of aging and after thermal runaway occurrence). The complete multi-scale analysis of Li-ion cell is illustrated in Figure2.

At cell scale, electrical analysis activates the battery and provides the information on cell state of health (such as actual capacity, impedance distribution, …) and cell performance (rate capability, …). X-ray Tomography is the preferable analysis method to investigate the internal structure of the battery, especially cell safety features that directly link to the venting mechanism and the type of battery central core metallic structure.

At component scale, SEM & EDS methods can give the knowledge about the morphology and geometry of cell components, especially the cell electrode active grains and different layers of separator.

At material scale, chemical mapping in combination with X-Ray diffraction analysis can be used to indicate the appearance of the existing chemistry in the cell electrodes and to identify the active material stoichiometry in the electrode grains.

Besides, techniques based on DSC measurements should be implemented to study the thermal stability of the materials/components used in selected LIBs (electrode materials, electrolytes, separators, …) and the degradation products.

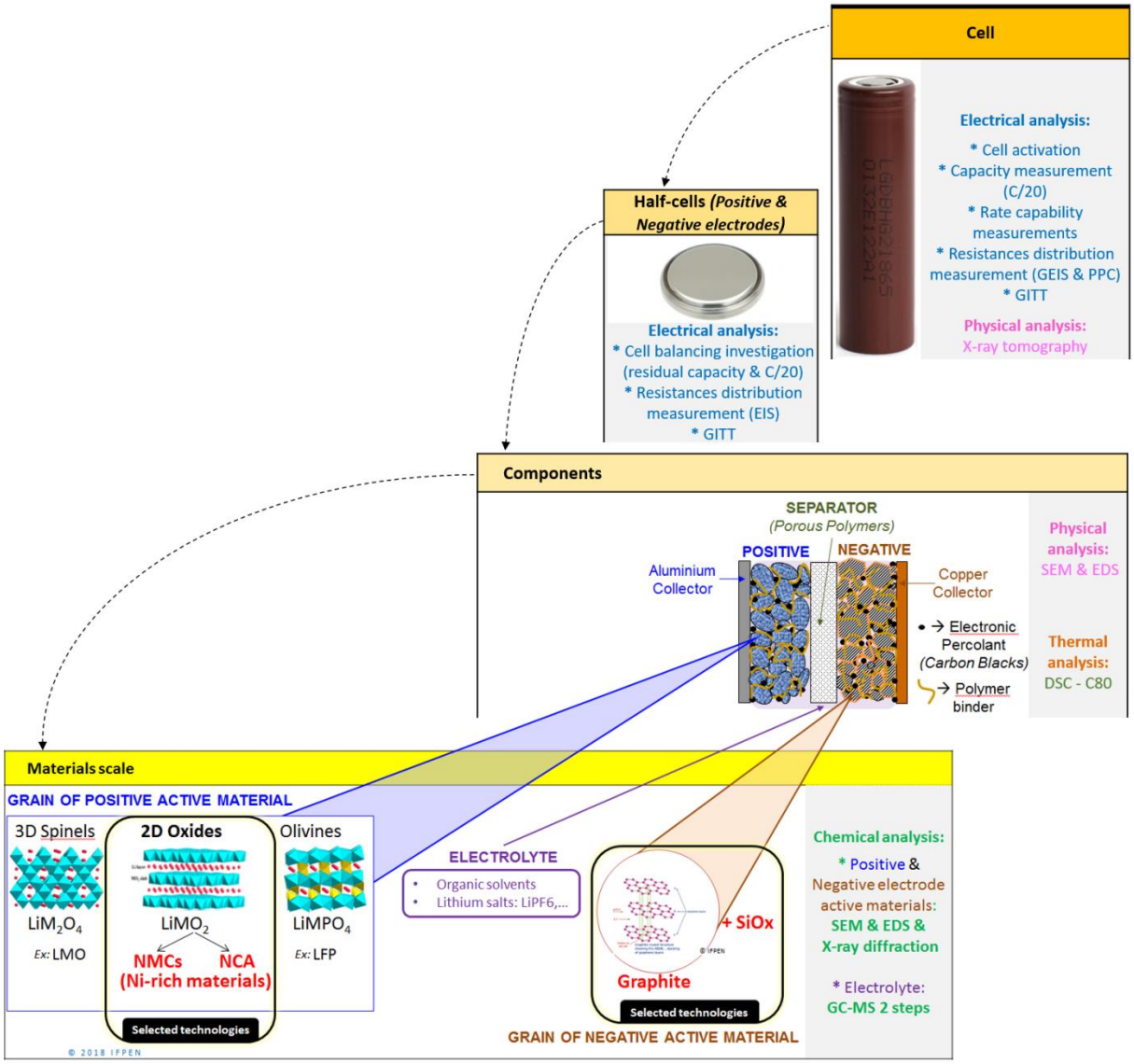


Figure2: Process of the complete multi-scale analysis of Li-ion battery cell

2.3 Implementation of Safety-focused Aging campaign

Aging of LIBs has a notable impact on the abusive behaviour of thermal runaway according to recent literature [3, 5, 9–12]. It is an inevitable degradation process which leads to capacity loss and internal impedance increase, thereby, loss of rate capability and cell performance during the whole lifetime of Li-ion batteries. It links closely to the inherent safety issues inside Li-ion batteries. Several studies confirmed that SEI growth driven aging process seems to play a critical role in thermal runaway. In the context of emerging higher capacity Li-ion batteries including Ni-rich high energy technologies, Li plating also seems to have a notable impact on the behaviour of Li-ion cells in thermal abuse conditions through increased risk of short circuit, and hence likely higher sensitivity to thermal runaway [3, 5, 11, 13, 14]. Both described aging related phenomena in high energy LIB cells interacting with their safety performance are illustrated in Figure3.

Several pre-tests have been carried out in IFPEN to define a complete safety-focused aging campaign with 2 target degradation mechanisms. Li plating aging protocol is based on abuse cycling conditions at low temperature which accelerate the occurrence of Li deposition/plating. SEI-driven aging protocols have been developed based on storage/cycling conditions which accelerate the evolution of SEI but also minimize the occurrence of Li plating. These protocols are customized to the different technologies studied.

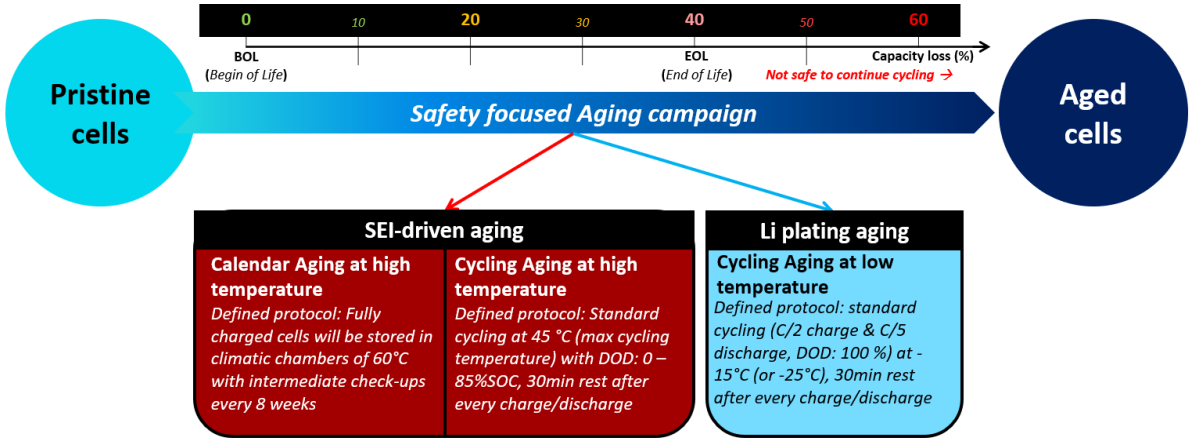


Figure3: Safety-focused aging campaign

To allow better understanding and calibration of runaway phenomena, non-destructive techniques of qualification and quantification of aging state based on [15–17] have been used to knowledge the state of aging of the cells before carrying out runaway tests.

2.4 Thermal safety tests:

Thermal abuse is the most direct way to exceed the thermal stability limits of a Li-ion cell. Therefore, thermal safety tests will be carried out to subject the cell to external heating. Pristine and aged cells obtained from the aging campaign, in known and quantified degradation states, are currently progressively undergoing thermal abuse tests on the STEEVE platform at INERIS. These tests will make it possible to understand the processes involved in the thermal runaway of the batteries and to calibrate the models.

Thermal runaway process requires not only elevated temperature, but an adiabatic (highly insulated) environment, and extended times to reach a self-sustaining thermal runaway condition. The quasi-adiabatic conditions in Accelerating rate calorimetry (ARC) tests can be regarded as the case of perfect thermal insulation. Therefore, the results of ARC tests represent a worst-case scenario where the safety behaviour is mainly characterized by the onset of thermal runaway (*T_onset*), the onset of rapid thermal runaway (*T_rapid*) and the self-heating temperature rate.

ARC tests are typically operated by the heat-wait-search (or seek) (HWS) algorithm. In such experiments, a cell is heated to a certain temperature (e.g. 30 °C) and if significant self-heating of the cell is detected (SHR> 0.02 °C/min) after defined wait and search periods, the ARC changes into the exothermic tracking mode where it follows the temperature on the cell sample. The heat is not allowed to be transferred from

the cell to its surrounding. In case the self-heating rate is not significant, the temperature is increased, for example by 5 °C and this step is repeated until significant exothermic reactions are detected. ARC HWS algorithm are illustrated in Fig. 4.

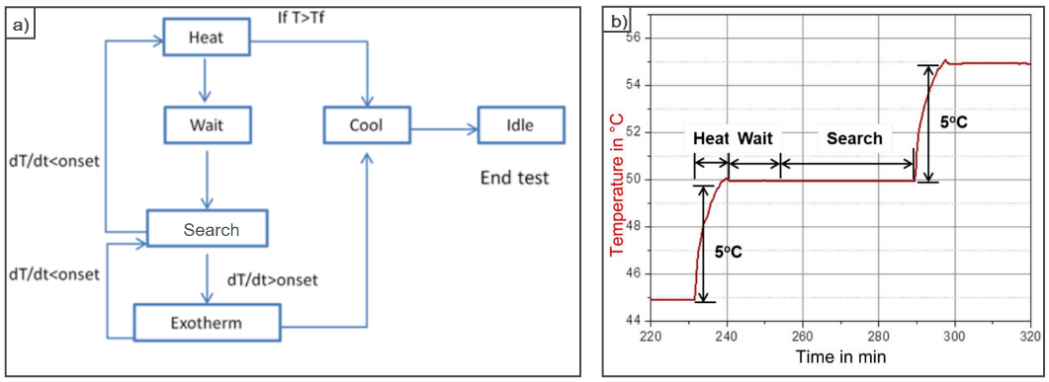


Figure4: a) The heat-wait-search (HWS) method and b) Example of cell surface temperature curve during HWS experiment in an ARC. [18]

3 Results

These ARC HWS test results presented below were performed in BTC500 E1735 with the cell setup as shown in Fig. 5.

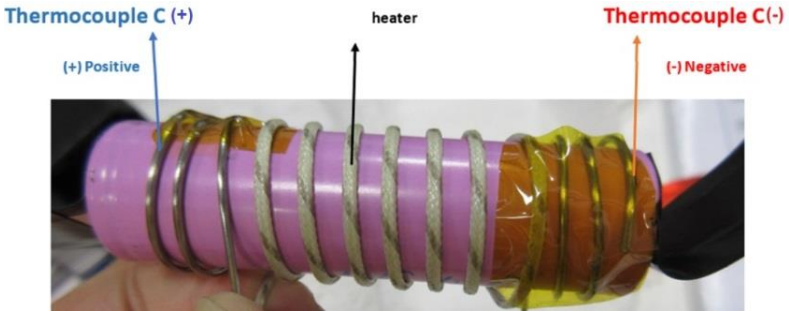


Figure5: ARC test sensors (thermocouples) and heater setup on 18650 cells

The thermocouple C(+) is near cell cap (positive side), therefore, its temperature can present the influence of the safety features during venting & combustion. In the other hand, the results of thermocouple C(-) will be more representative for the behaviours and thermal stability of the cell chemistry.

3.1 The thermal runaway of selected technologies at pristine state 100%SOC

The thermal runaway chain exothermic reactions of LG HG2 and Panasonic NCR GA technologies at 100%SOC can be divided into three clear stages as shown in Fig. 6. The temperature threshold between stage 1 and stage 2 is called the onset temperature of thermal runaway (*T_onset*) and the temperature threshold between stage 2 and stage 3 is called the onset temperature of rapid thermal runaway (*T_rapid*). The duration of these stages is independently on the HWS protocol and therefore representatives for the characteristic of the cell. Inspired by [5, 7, 9, 19], the thermal runaway process of these cells can be analysed as below:

Stage 1 refers from the end of the “safe zone” corresponding to the initial self-heating detected (*T_self-heating*) to the temperature onset of the thermal runaway (*T_onset*) where the cell internal temperature rate starts to strongly increase. *T_onset* of LG HG2 is ~145°C and *T_onset* of Panasonic NCR GA is ~151°C. During this stage, the battery operation changes from a normal to an abnormal state, and the reactions caused the battery to overheat are:

- Initial decomposition of SEI (> 60°C) (dominant reaction),

- Starting exothermic reduction of electrolyte at the lithiated negative electrode or so-called the SEI regeneration reaction,
- Endothermic reaction of double-layer separator melting & fusion (T > ~120 °C). This leads to internal short circuits (ISCs).

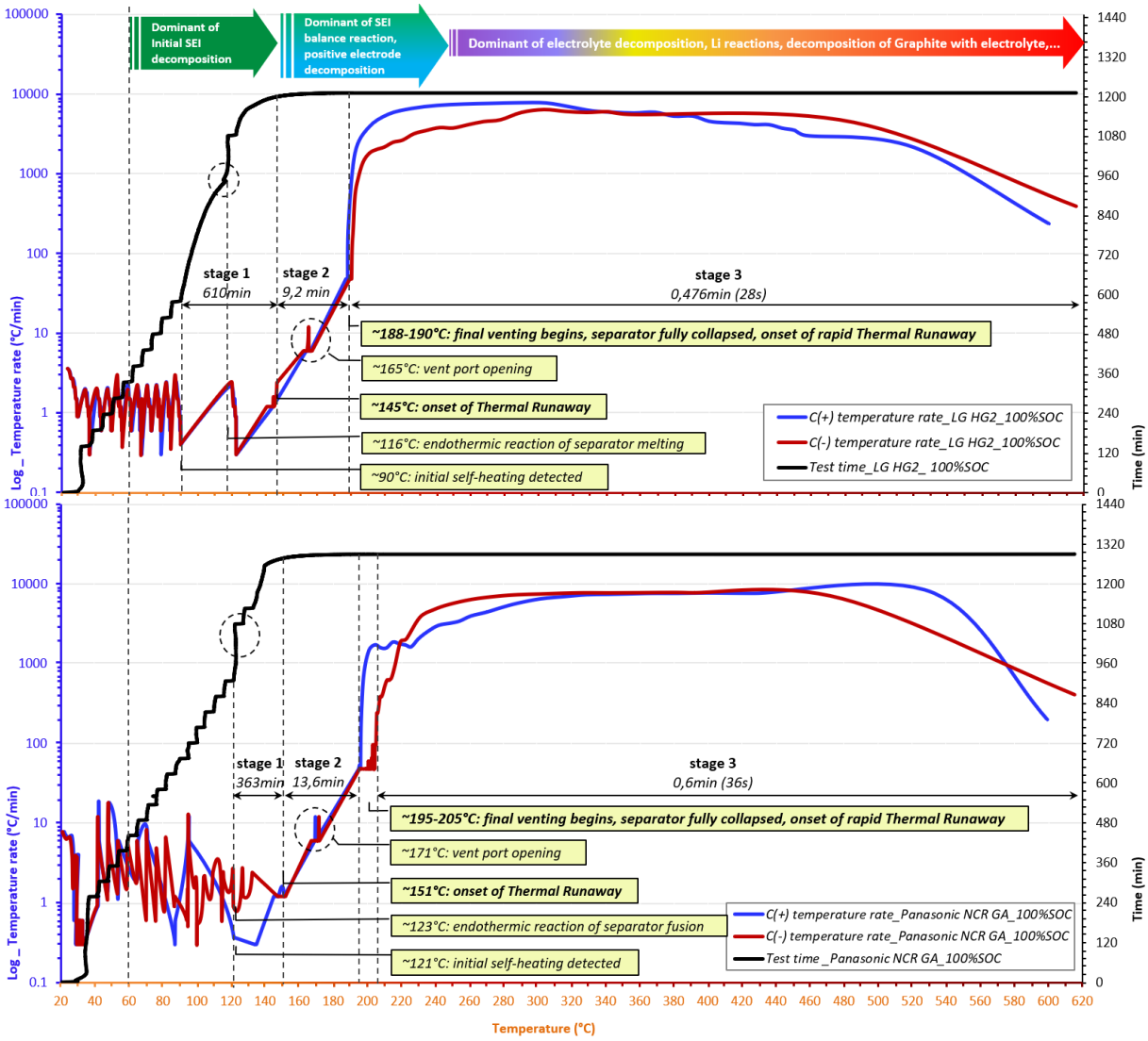


Figure6: Temperature rates and test time versus temperature plot of the thermal runaway of 100%SOC LG HG2 (top) and 100%SOC Panasonic NCR GA (bottom) pristine cells

Stage 2 occurs from *T_onset* to the temperature onset of the rapid thermal runaway (*T_rapid*) where the cell internal temperature rate suddenly accelerates. During this stage, the temperature violently increases, and oxygen accumulates inside the battery due to these exothermic reactions:

- Further electrolyte balance reaction of SEI regeneration and decomposition at negative electrode (dominant reactions at the beginning of stage 2),
- Starting electrolyte oxidation at positive electrode (dominant reactions at the end of stage 2),
- Venting: gases & smoke formation. Once the cell vents, the ultimate severity of the reaction is dominated by the ignition of flammable vent gases.

Therefore, stage 2 is the heat accumulation and gas release process. It lasts about 9.2 min in case of LG HG2 and about 13.6min in case of Panasonic NCR GA. To proceed from stage 2 to stage 3, the hard ISCs

after ceramic layer collapsed will accelerate the heat accumulated and activate the battery combustion as soon as there is enough oxygen (mainly from the positive electrode decomposition reactions and from the air). *T_rapid* of LG HG2 is ~188°C and *T_rapid* of Panasonic NCR GA is ~195°C.

Stage 3 occurs from *T_rapid* and leads to maximum temperature of ~615°C for both cells. During stage 3, combustion occurs, vessel pressure aggressively increases due to final venting, thereby causing fire & chemical explosion hazards due to the strong exothermic reactions below:

- Decomposition of positive electrode (highly exothermic reactions),
- Strong exothermic reaction between oxygen (released from positive electrode) and electrolyte,
- Decomposition of electrolyte (combustion reactions),
- Reactions of intercalated Li with air (H2O) diffused into cell after venting and/or with binder,
- Decomposition of Graphite with electrolyte (the balance reaction of SEI decomposition and regeneration is broken, graphite structure collapsed),
- Binder reactions.

This intensive stage is the combustion and explosive decomposition stage. It last ~28.5s in case of LG HG2 and ~36s in case of Panasonic NCR GA. During this stage, venting and cell component ejection accelerated.

It should be noted that these reactions do not strictly happen one after another in an order. They are, rather, complex and systematic issues.

3.2 The factors impacting the thermal runaway of selected technologies

3.2.1 The impact of separator

As presented in Fig. 7, the two selected technologies have similar double-layer separator technology: one layer of polymeric based carbon-rich and one layer of ceramic.

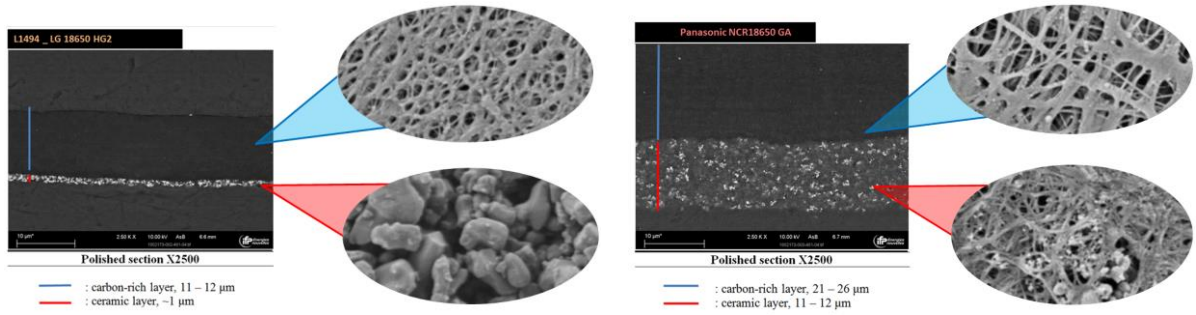


Figure7: SEM images of LG HG2 (left) and Panasonic NCR GA (right) double-layer separator.

In case of LG HG2, the polymeric layer has homogeneous porosity and the ceramic layer is the mixture of mainly grains. Thickness ration of ceramic layer over polymeric layer is ~1/12 and the separator total thickness is ~13 µm.

In case of Panasonic NCR GA, the separator polymeric layer has inhomogeneous porosity. The ceramic layer composes of long fibers in mixture with different grains. Thickness ration of ceramic layer over polymeric layer is ~1/2. This separator total thickness is ~38 µm, significantly thicker than the separator of LG HG2. Therefore, the Panasonic NCR GA's separator collapsed at higher temperature and its hard ISC has also delayed (~205°C compared to ~190°C of LG HG2's) which lead to the delay of stage 3 activation as observed in Fig. 8 (top). This influence of different separator thickness also impacts the moment of cell venting (Fig. 8 (bottom)).

3.2.2 The impact of electrode materials

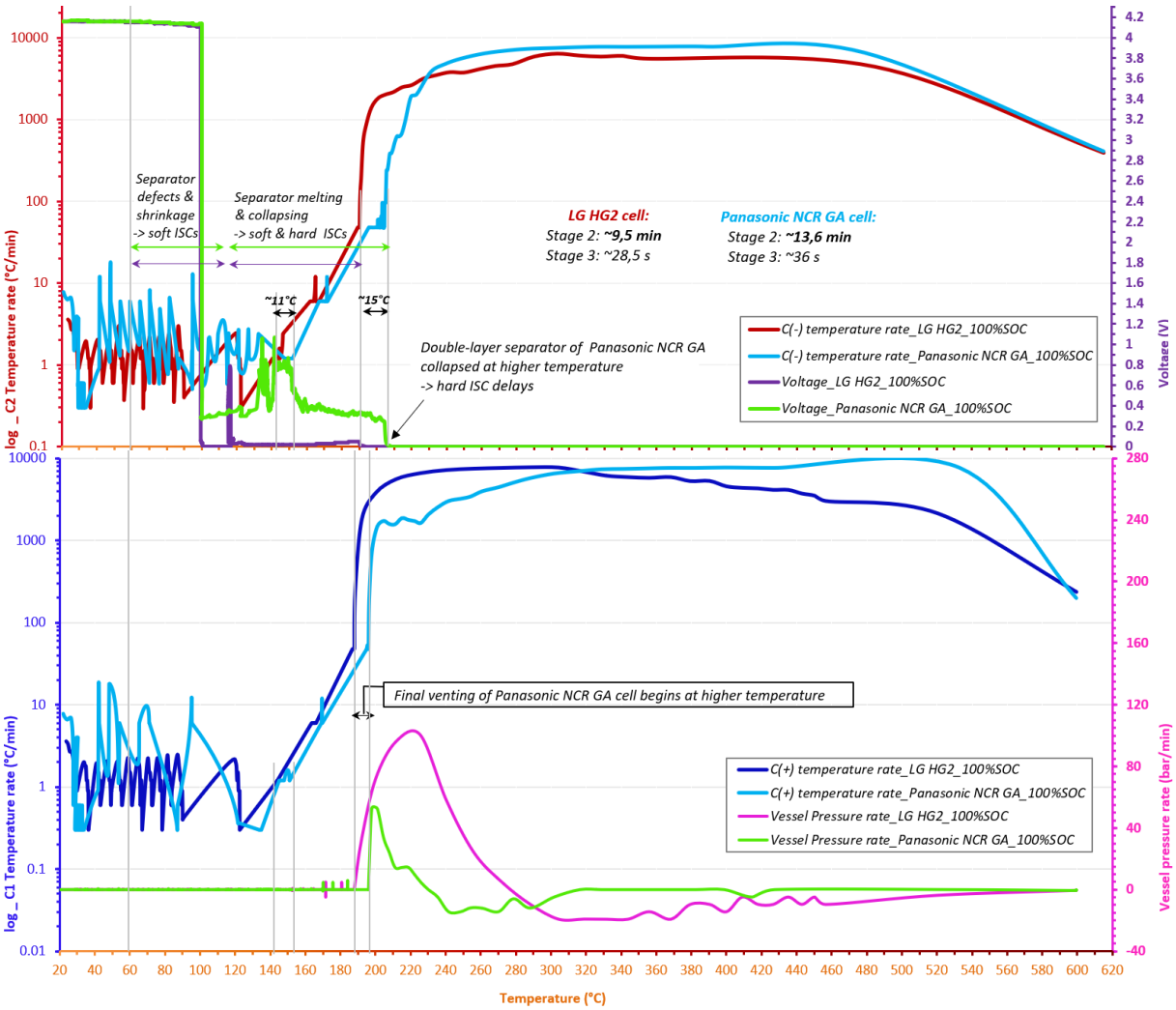


Figure8: Temperature rate of C(-) and cell voltage of LG HG2 and Panasonic NCR GA (at 100% SOC) versus C(-) temperature plot (top). Temperature rate of C(+) and vessel pressure rate of LG HG2 and Panasonic NCR GA (at 100% SOC) versus C(+) temperature plot (bottom).

Having similar technology of negative electrode (Graphite-SiO_x composites), different positive electrode technologies (NMC811 in LG HG2 and NCA in Panasonic NCR GA) have impacted the total duration and the severity of stage 3: With higher temperature rate, NCA cell lasts ~36s while stage 3 of NMC811 lasts 28.5s as shown in Fig. 8 (top). We also found that the temperature rate during stage 3 of NCA technology is higher with longer duration regardless of 100% SOC or 50%SOC. In conclusion, at these SOC levels, NCA technology appears to be more violent during the final stage of the thermal runaway than that of NMC811 technology.

3.2.3 The impact of SOC

The thermal runaway of LG HG2 and Panasonic NCR GA have been investigated at two different level of SOC: 100% and 50%. The impact of SOC on the activation of stage 2 and stage 3 is significantly observed in Fig. 9-10.

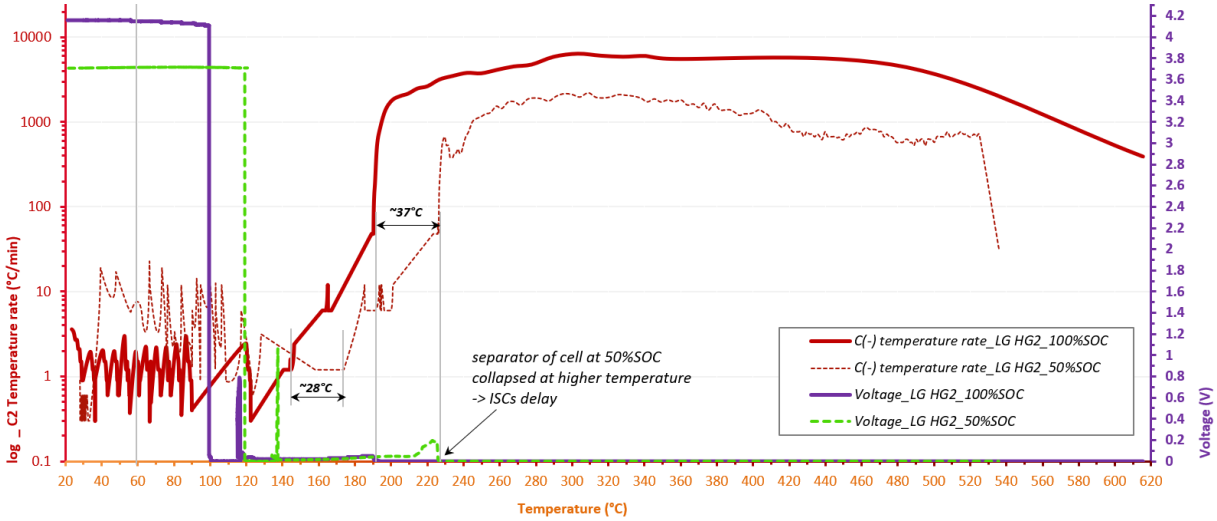


Figure9: Temperature rate of C(-) & voltage of LG HG2 at 50 and 100%SOC versus C(-) temperature plot.

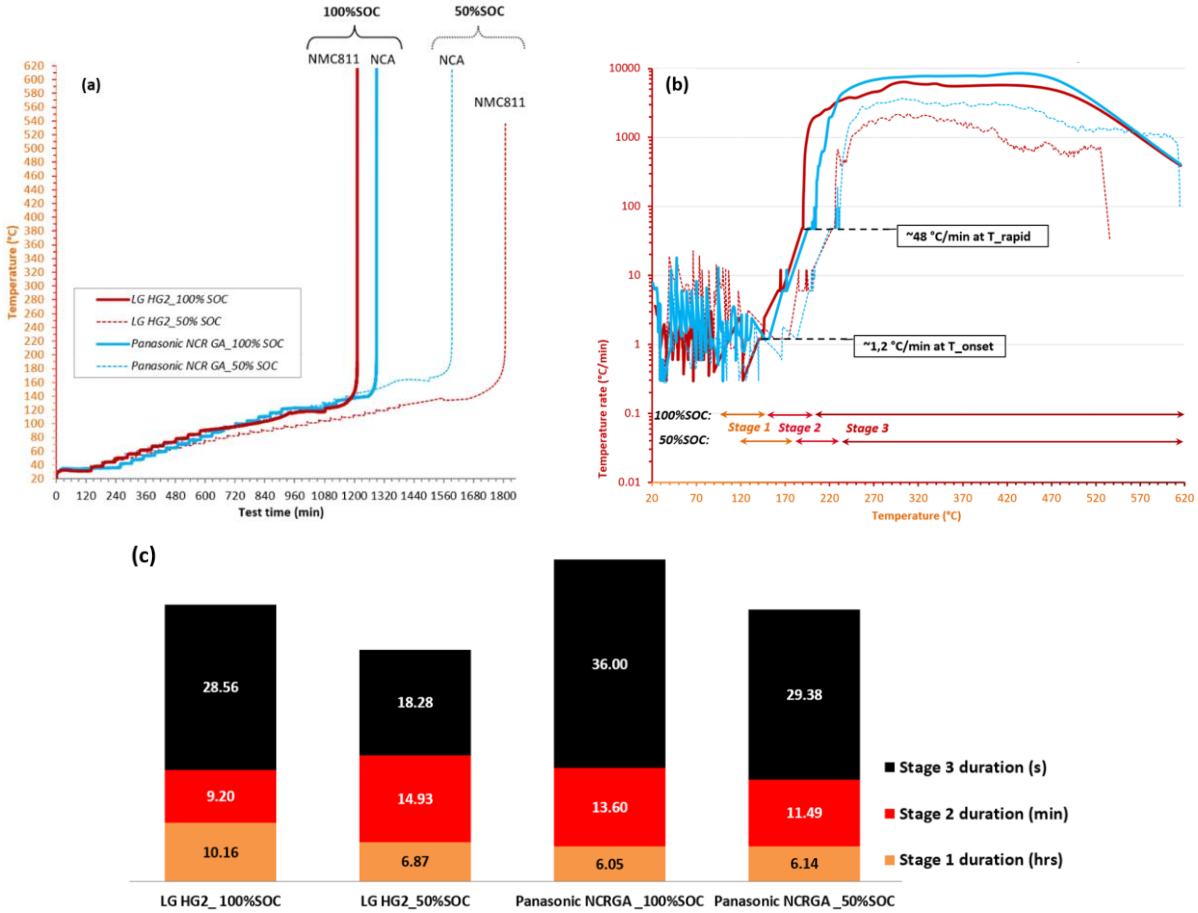


Figure10: (a) Temperature profiles versus test time and (b) temperature rate versus temperature of the 50 and 100% SOC LG HG2 and Panasonic NCR GA cells plot. (c) duration of the stages during thermal runaway of the tested cells.

Lower SOC level leads to ISCs delay and thereby, shifts the activation of stage 2 and stage 3 to higher temperature as shown in Fig. 9. Additionally, proven in Fig. 10, cells at 50%SOC require more time before undergoing thermal runaway and their thermal runaway processes can only be activated at higher temperature. Moreover, their stage 3 is less severe with lower temperature rate and shorter duration. Therefore, the pristine cells 50%SOC is less reactive. This SOC-dependency shift is stronger in case of LG HG2 technology, their maximum temperature is also reduced with reduced SOC (~615°C in case of

100%SOC and ~535°C in case of 50%SOC). However, all test cells exhibited clearly 3 stages with similar temperature rate thresholds (~1.2°C/min at *T_onset* and ~48°C/min at *T_rapid*), regardless of SOC and cell technology.

3.2.4 The impact of safety features and SOC to venting and component ejection mechanism

During thermal runaway process, venting events lead to gas release & cell component ejection. The remaining of cell after undergone thermal runaway (residuals) has been measured. As shown in Fig. 11, the mass loss of pristine cells 50 % SOC is lesser than that of pristine cells 100%SOC. This is additionally confirmed the lower reactivity of cells with reduced SOC.

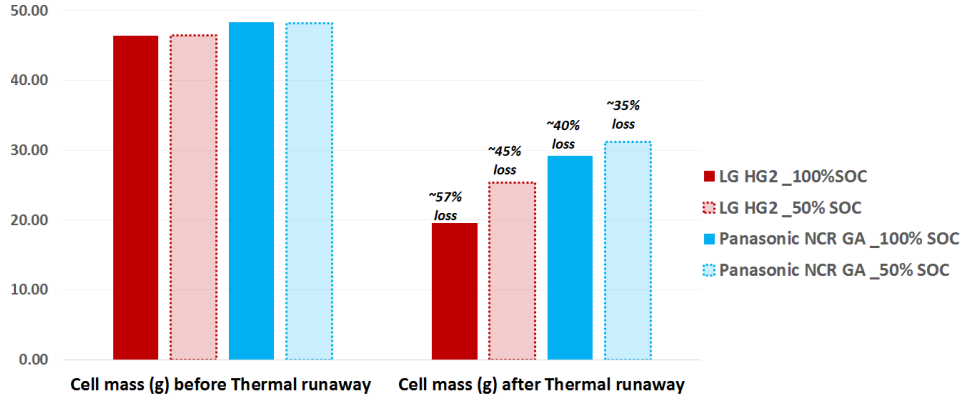


Figure11: Cell mass before and after thermal runaway

Investigating deeper into the impact of SOC to the venting mechanism, although these cells have different safety feature design, we observed in Fig. 12 that during final venting of all cells, the vent ports fully opened, however, the gasket seal only collappsed in case of cells with 50%SOC. This is also confirmed by the remaining of cells after the thermal runaway (presented in Table 2). This can be explained due to the delay of hard ISC in case of 50%SOC which shift the activation of final venting to higher temperature where more strong exothermic reactions occur with higher reaction rates and eventually, the pressure rate accelerated during stage 3 collapse the gasket seal. Therefore, the jelly roll of 50%SOC cells violently eject (Table 2). This also explain why the duration of stage 3 is shorter in case of 50%SOC.

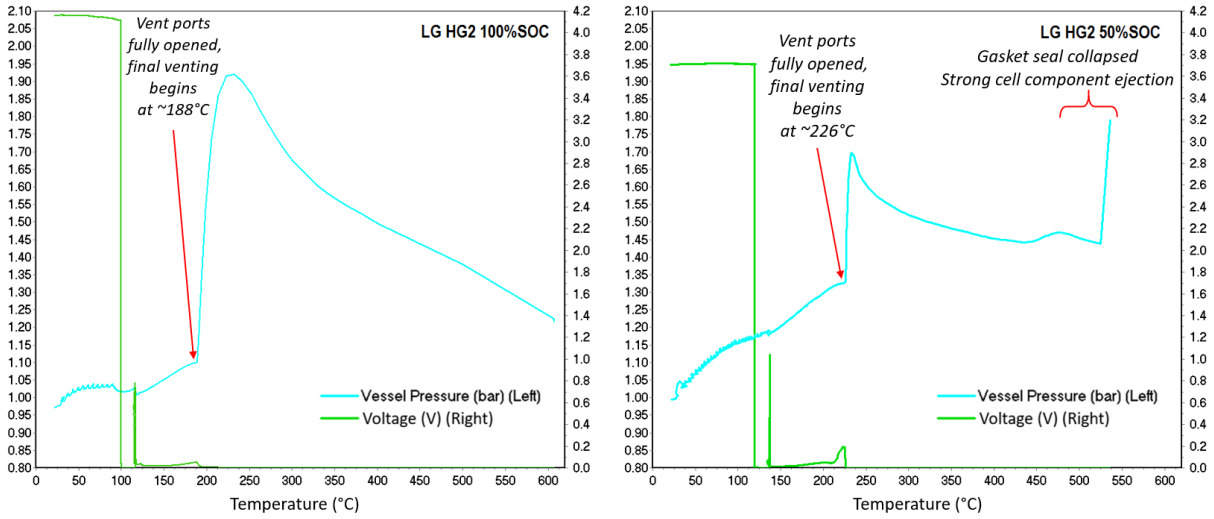
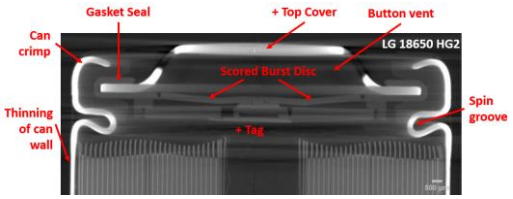
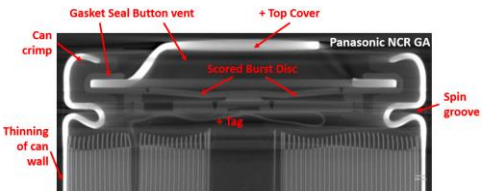
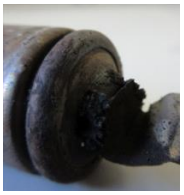
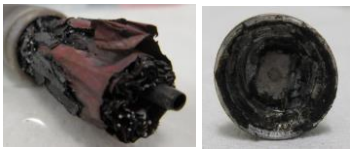


Figure12: Cell voltage and vessel pressure (bar) versus temperature of LG HG2 100%SOC and 50%SOC

The presence of a stiff center tube and a metal bar is clearly observed in the design of Panasonic cells and LG cells respectively (the open center core) in Table 2 to allow pressure equalization, preventing winding ejection during thermal runaway. However, it does not work effectively in case of cell at reduced SOC.

Table2: The impact of safety features and SOC to venting and component ejection mechanism through the pictures of LG HG2 and Panasonic NCR GA (50 and 100% SOC) after thermal runaway.

	LG 18650 HG2	Panasonic NCR GA
Cell internal tructure global view		
Original design of safety features near cell gasket		
100% SOC cells after thermal runaway		
50% SOC cells after thermal runaway		

4 Discussion and perspectives

The experimental methodology of exploring the effect of high energy Ni-rich LIBs on the thermal runaway and investigating the underpinning mechanisms and relationship between calendar & cycling aging and safety has been defined. As the critical first step for the rest of this research, this experimental works led us to a clearer understanding of the thermal runaway process influenced by both aging and cell technology.

The selected technologies studied are well confirmed that they are all very reactive technologies with high Ni content in positive electrode materials in combination with the Graphite-SiO_x composites negative electrode technologies. The safety-focused aging campaign is ongoing at IFPEN and the safety thermal tests in adiabatic condition (ARC) on pristine cells has also started at INERIS with the interesting presented results. The thermal safety tests on aged cells are in progress.

Future works deals with the calibration and validation purposes of the development of a consolidated thermal runaway 3D model in order to predict the behaviours of different Li-ion batteries nearby and during thermal runaway. This coupled multi-physics model will improve & extend the previous thermal runaway initial model built at IFPEN [7] by integrating the impact of Li Plating & SEI-driven cycling aging. The calibration of the model for different technologies will be based on the obtained results of experimental study. According to progress of modeling activities, complementary testing based on another thermal test such as Oven test, will be performed to serve modeling validation purposes. The models developed will be implemented to understand the electrical or thermal initiation of the phenomenon of thermal runaway and its propagation within a battery pack regarding its design. The experimental & simulation results will contribute to the finding of the keys factors that can improve the safety of these highly reactive Li-ion batteries. They will eventually be transposed into tools enabling the best design of the packs and avoidance of this undesirable phenomenon.

Acknowledgments

The authors gratefully acknowledge the supervision & the funding of IFPEN & INERIS and especially the guidance of LRCS for this work.

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