Safety Potentional Analysis of 18650 Lithium-Ion-Cells

Cell Performance Investigation by individual safety device in view of battery pack level

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***Abstract*—Safety behavior of battery systems is a major objective for all battery pack designs. 18650 cells with their small energetic masses and their safety devices such as CID or venting plate, show excellent safety behavior in single tests. Nevertheless this does not give a clear statement to the cell behavior in serial and parallel cell strings. This paper at hand examines the safety behavior of 18650 cells under battery pack level conditions. Therefore, three types of cells with different positive active materials (NCM, NCA, LFP) have been investigated. In various tests the cells were strained with voltages and currents that occure on battery module level. The thermal behavior as well as the electrical reaction of the CID has been investigated and an individual safety grid has been generated that displays, at which voltage and current values the cell goes into a hazardous behavior. The results reveal that the risk of a thermal runaway caused by a CID functionality failure increases with high current values in combination with high voltages. Generally there is a strong dependency on the current value that can drive NCA and NCM cells already at 20V to a thermal runaway. The LFP cells show generally a safer behavior with intensive smoke generation to higher voltages and currents. NCM cells show the most excessive thermal runaways.**

Keywords—Lithium-Ion, NCM, NCA, LFP, CID, safety. Battery module

I. INTRODUCTION

The Lithium-Ion-Technology reveals promising chances to establish electro mobility in society by increasing range and performance of electric vehicles. Nevertheless, Lithium-Ion Battery Systems (LIB) show fatal behavior if they are exposed to extreme temperatures, high voltages, or current values that are off the permitted operational window [1]. Single Lithium-Ion Cells (LIC) are equipped with several safety devices such as safety vents that shall ensure the system safety in case of Battery Management System failure. In multiple electrical, mechanical and environmental tests, the cells have to reveal their non-critical behavior [1] to ensure safe operation of the cells in the LIB.

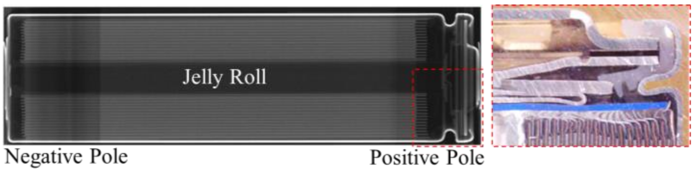
The safety behavior of single LIC itself however does not guarantee a safe operation of the battery system, since the boundary conditions are different – due to parallel and serial interconnection, the cells may be stressed with higher voltages and/or higher current loads due to a failure or rupture within the battery system. Since the safety device is not designed to cope stresses on system level (e.g. 48V battery module level), a crucial safety hazard is constituted. This elucidates the need of an investigation of single cells under system level stresses.

The paper at hand examines the safety behavior of three 18650 cells with different active materials (NCA, NMC, $LiFePO_4$) and contrasts the requirements for enforcing safety device failure in respect of cell capacity, current and voltage loads. The different active materials and capacities shall provide information about the individual hazardousness, if the cell goes into a thermal runaway.

A. Format of 18650 cells

Currently, there are the three applicable LIC designs *pouch*, *prismatic* and *cylindrical*. The paper at hand focuses on the cylindrical format 18650.

The cell design of an 18650 cylindrical battery cell (diameter 18mm, length 65mm) is a common format that is used in all Tesla Motors vehicles [2]. The coiled active materials (jelly roll) are inserted into a metal cup that provides a similar high stability as the prismatic design. The current drain is realized via the poles on the faces of the cylinder. The robust cup can resist an increasing internal pressure caused by chemical side reactions. Picture 1 shows a section of a 18650 LIC. The close-up of the LIC-head shows a complex arrangement of different safety elements that will be discussed in detail. In the axial direction, some cells feature an inner hollow tube, mainly because of cell production and jelly roll stability. The capacity of an 18650 cell is between 1 Ah and 3.4Ah, with other cylindrical sizes delivering up to 6.8Ah [4].



Picture 1:X-Ray scan of a 18650 cell with close up of cell head [5]

B. General safety aspects

A powerful lever for improving the safety behavior of single cells are the active materials masses and their chemistry.

By comparing the energy equivalent of an 18650 cell with a capacity of 2.2Ah to a large Coffee-Bag cell (40 Ah) the small cell shows only about 6% of the releasable and therefore potentially hazardous energy of the large cell [17]. Small cells with their little reactive masses generally can be considered safer compared with large cells. In different studies and simulations, scientists have shown that cascading failure of cells and therefore the hazard increase with cell dimensions [4].

The safety hazards of a cell is mainly influenced by its active materials, especially by its cathode [1]. $LiCoO_2$, as one of the first commercial used cathode materials shows distinct higher safety hazards due to exothermic reactions at the lowest temperatures. The mixed metal oxides NCA and NMC show a safe behavior to higher temperatures. $LiFePO_4$ and $LiMn_2O_4$ reveals the lowest heating rates and therefore the largest safety window. One reason is that the last two barely produce oxygen that acts as combustive agent [6].

Burda [4] summarizes various values for the cathode material description as follows:

Table 1: Safety Characteristic of active materials [4]

Positive Electrode	Stability	Safety
LCO $LiCoO_2$	→	↘
NCA $LiCo_xNi_yMn_zAl_wO_2$	↗	↘
NCM $LiCo_xNi_yMn_zO_1$	→	→
LFP $LiFePO_4$	↑	↑

Ketterer et al. outlines the safety performance regards actual cells with the corresponding negative electrode, see Table 2. Regarding the exothermic reactivity; he states that $LiFePO_4$ shows no exothermic reaction while $LiCoO_2$ reacts at the lowest temperatures followed by NCA. [7]

Table 2: Characterization of LIC [7]

Positive (negative) Electrode	Perfor-mance	Energy	Safety
$LiCoO_2$ (Graphite)	↗	↗	↘
$Li(Ni_{0,95}Co_{0,2}Al_{0,04})O_2$ (Graphite)	↗	↗	→
$LiFePO_4$ (Graphite)	↗	→	→
$Li(Ni_{1/3}Co_{1/3}Mn_{1/3})O_2$ (Graphite)	→	→ / ↗	→

Various safety rankings, based on investigations of different positive electrode materials, have been made. Nevertheless, they do not display a holistic view on the cell safety since aspects like anode reactivity, cell design aspects, or the probability of internal defects are not taken into account. [5]

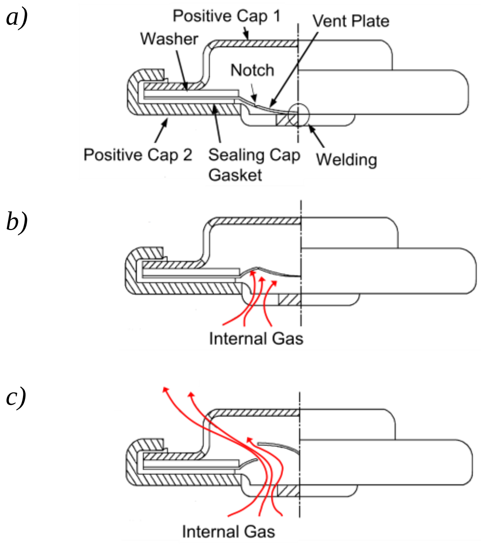
C. *Safety devices*

In addition to safety enhancement measures of the active material composition, the 18650 cells can be equipped with safety devices such as safety vents, positive temperature coefficient elements, shutdown separators and current interrupt devices [9]. Most of these devices are located at the positive pole of the cell.

The positive temperature coefficient device (PTC) is a self-resetting safety device located at the positive pole as a dividing layer that passively increases its electric resistance at high temperatures. At high current values, the current flow through the cell is stopped and therefore the joule heating (~RI^2) within the cell is reduced. After the PTC temperature decreases to a certain value, generally 100°C, it becomes conductive again. The PTC protects the cell of overheating and can operate several times, even though it resets not fully, which again can lead to higher ohmic resistances. [8]

The separator between the positive and negative electrode can be designed as “shutdown” which means that in case of an extreme temperature rise within the battery, the separator begins to melt closing the transfusing holes in the separator and interrupting the ionic conductivity. This effect stops the heating of the active materials due to interrupting the electric current flow and therefore the ohmic heating (~I^2R) of the cell. These shutdown separators can also close single punctures [5]. Due to thermal heating process before the shutdown, the temperatures may rise even after the melting processes causing the electrodes to come into direct contact. This “meltdown” can then cause a thermal runaway [8]. Most shutdown separators were designed for voltages under 20V and under 10A, which indicates that under higher values this safety device may not work properly [9]. Other separators with ceramic components offer a stability up to higher temperatures, but without the shutdown capability [5].

In case of an internal pressure rise within the cell (for example due to chemical reactions), the positive pole obtains two aligned safety devices called current interrupt device (CID) and gas release vent.



Picture 2: a) Positive pole design; b) Phase 1: CID opens; c) Vent opens [10]

In a first reaction to an increasing inner cell pressure, the CID can stop the charge/discharge current flow through the cell. If the internal pressure exceeds a certain value, the welding between the vent plate and cap breaks off. This pushes the vent plate up and disconnects the positive cap from the active material (Picture 2a). Hence, the current flow through the cell is stopped and the internal pressure might be limited to avoid gas release, leakage, or rupture. This mechanism is unrecoverable and usually takes action in case of overcharging [10]. In case of an ongoing gas generation, the second mechanism (gas release vent) is triggered. Usually this happens at inner pressures of 13.8 bar [5]. The pushed up vent plate will break at its designated notch to release the gas, see Picture 2c. This leads to a leakage of active materials but prevents the cell from rupture [5, 10]. This graceful venting of Panasonic cells occurs at pressures around 10 bar [10]. The APR18650M1 shall vent at pressures of 33.4 - 37.2 bar and burst at 55.2 – 58.5 bar [11].

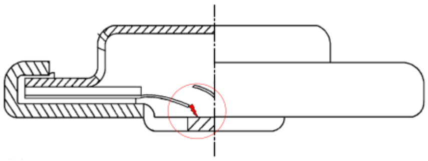
D. *Safety hazards of single cells in battery systems*

The mentioned safety devices show a safe reaction at single cell tests. However, the electrical conditions of the cell within a module differs disctintly. Depending on the actual LIB module design, a number of about 12 to 14 cells are connected in serial and over 30 cells can be aligned parallel.

Doughty et al. observed the harzardous behavior of shutdown separators within a small module [6]. The failure was investigated in a short circuit test of a 12-cell series-connected string of 5-Ah Li-ion rechargeable batteries. The shutdown separator was triggered at the temperature of about 130°C as supposed to and the cell became high ohmic resistive, the voltage drop within the cell reached -38V and within a short time, the cell caught fire. [6, 12] This high voltage exposure to single LIC indicates that the shutdown separator may become a hazard to the module safety. Beyond that, the voltage level is a potential risk for other safety features to fail.

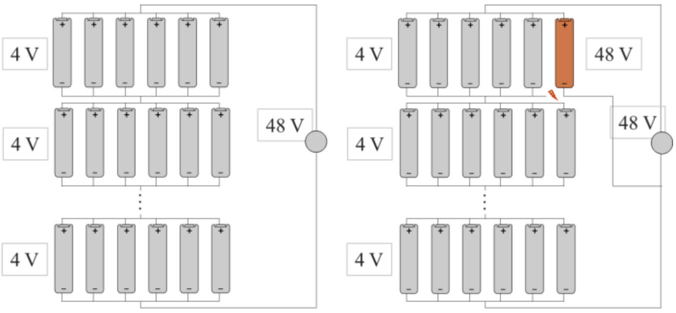
In different examinations, protection devices within the cell such as CID and PTC showed poor safety behavior. Especially while overcharging processes, the safety devices could not protect the cell from hazardous situations, since the separator was damaged before the PTC became high ohmic resistant and the CID reacted. [13] The CID may show a hazardous behavior if the cells are connected in series. Generally the CID in small cell arrays work properly. The probability of a non-simultaneous reaction of the CID however increases with the size of the cell array. Therefore, high currents may be applied to the cells with the slowest CID increasing the risk of a thermal runaway in systems. [13].

Another threat is an unclean rupture of the venting disk, see Picture 3. If the disk does not break at the notch, the space between the active material and the positive cab could be not wide enough. This entails the risk of a voltage flashover, so that the discharge/charge current flow is not interrupted. In case of higher voltages and/or high currents, this could lead to an unplanned spot welding. Due to the ongoing current flux and the extreme welding temperatures at the positive pole the cell will heat up rapidly. Furthermore, the current cannot be interrupted and may cause a thermal runaway of this cell.



Picture 3: Incorrect tear of vent disc with indicated spark-over

The situation of high voltage impact to single LIC may occur if the tabs of the cell gets in touch with the overall system ground potential, see Picture 4. The system voltage then is applied to the red marked cell. This may result by a tear-off of spot welded Hilumin ®-Tabs through vibrations, or by the ramifications of an impact on the battery system (puncture, crush, etc.)



Picture 4: Hazard of high voltages at a single cell in a battery system

Experimental Setup

E. *Samples*

For the investigation the following three types of 18650-LIC have been selected:

Table 3: Cell data, Source: Datasheet Cell manufacturer

Description	Manu-facturer	Positive Electrode	Nominal Capacity [mAh]	Nominal Voltage [V]	Max. cont. discharge [A]	Number of Samples
NCR 18650PD	Panasonic	NCA	2900	3.6	10	22
UR 18650AA	Panasonic	NCM	2250	3.7	8-10	14
APR18650M1	A123	LFP	1100	3.3	30	8

The types have different active materials (NCA, NCM, LFP) on the positive electrode. The capacity of the cells vary between 1.1 and 2.9 Ah since the cell types are high performance, hybrid and high energy.

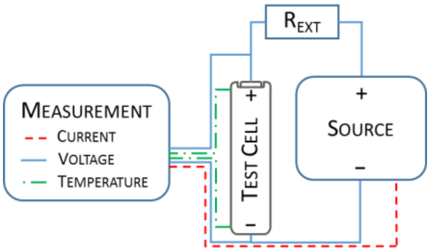
The APR18650M1 cells of A123 with its $LiFePO_4$ – chemistry at the positive electrode shall have an excellent abuse tolerance, due its nanoscaled electrochemical material particles [14]. A123 claims to have a more chemically stable cathode than metal oxide cathodes. In abusive conditions (high temperature, over voltage), there shall be less degradation (exothermic or heat-generating reactions) and therefore, less release of gaseous oxygen. Furthermore, cells with this Nanophosphate chemistry shall not show the excessive thermal runaway that metal oxide li-ion-cells usually show. [14]

The Panasonic cells shall have a superior safety behavior due to the HRL (Heat Resistance Layer) Technology. This layer is an insulating metal oxide on the electrode surface (Negative Material) that shall prevent an overheating even in case of a short-circuit. Panasonic also claims that due to its PSS (Panasonic Solid solution), the safety characteristics of cobalt based cells shall be increased to the safety standard of $LiMn_2O_4$- batteries [15].

F. *Apparatus*

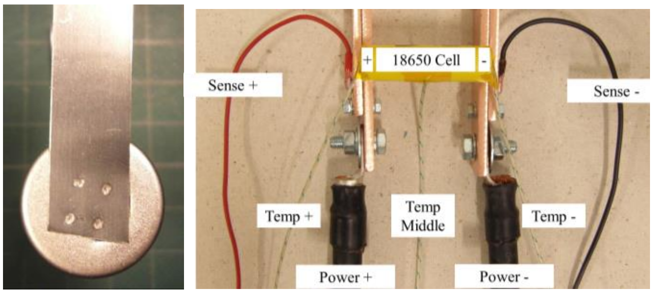
For the experimental process, the test cell has been set into a circuit of an external resistance R_{EXT}, a current source and a measurement unit, see picture 5.

The Source is capable to set the current between 20A and 800A. The voltage range is between 20V and 1000V. The external resistance has been set to 180 mOhm. The resistance of the 45mm² cables, the copper clamps and screw connectors can be set to additional 45 mOhm. For the electrical connection all cells have a Hilumin-Tab (~10 x 40 x 0,05 mm) spot welded on its positive and negative pole.



Picture 5: test bench setup

The cells was connected to the test bench via two copper clamps. Beside the Sense-wiring and the current measurement, three temperature sensors were placed to the positive, negative pole and in the middle of the jelly roll (approx. 30mm in axial direction from the negative pole).



Picture 6: a) LIC Spot welding b) Cell fixture, sensor positioning

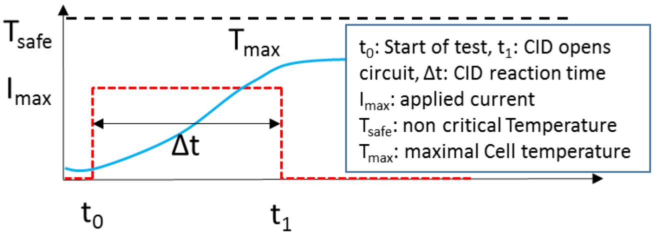
G. *Test conditions and process*

The investigations have been performed at room temperature 21°C. All cells have been charged to the maximum voltage: The NCM/NCA LIC to 4.2 +0.03 V, the LFP to 3.6V +0.03V. Through the test the parameters Cell Voltage, External Voltage, Current Value and Temperatures have been monitored at a frequency of 1000 Hz

The test cell was loaded with different current values between 20A and 120A at voltage levels between 20V and 140V by the source. Its thermal and electrical reaction was monitored via the measurement unit. The tests were recorded for visual verification. The applied electrical force was kept up until the cell reaches a steady states that did not show significant changes. To evaluate the individual behaviors of the cells, three categories were allocated:

1) *Case 1: Safe behavior (green)*

When the cell showed a benign reaction to the external penetration, the cell was marked as safe. In this case the CID interrupts the current flow at t_1, the cell temperature T_{max} does not exceed T_{safe}, see Picture 7. Eventually there was some smoke generation due to bursts of the venting plate, but no fire or thermal runaway occured.



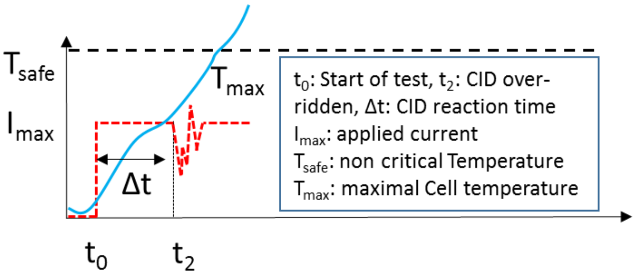
Picture 7: Safe behavior of a LIC

2) *Case 2: Undefined behavior (yellow)*

This case describes a potential hazardous, instable status of the cell. The CID opens, venting with smoke generation definitely occurs. Due to an ignition source caused by hot metal or an undefined spot welding, the cell gas catches fire causing a local hot spot. The LIC itself does not go into a thermal runaway.

3) *Case 3: Hazardous behavior (red)*

The third case describes the most hazardous reaction, called thermal runaway. The CID opens but gets overridden (t_2) and the cell goes into an excessive thermal runaway with fire, smoke and explosion ($T_{max} > T_{safe}$), see Picture 8. At the end, the cell body shows a glowing rod.



Picture 8: Hazardous behavior of a LIC

II. RESULTS AND DISCUSSION

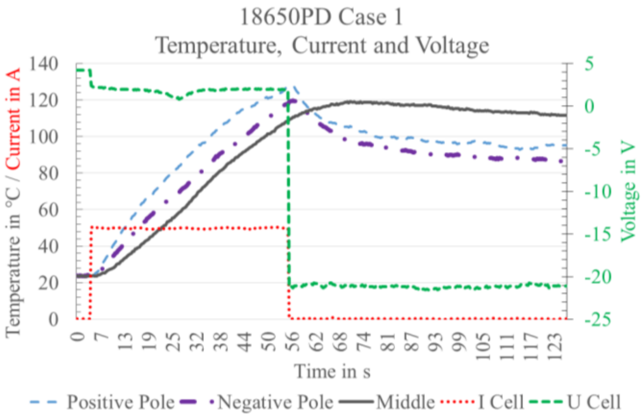
A. *NCR 18650PD*

1) *Thermal Reaction*

During the penetration, the cells of this type showed the following thermal reaction, see Picture 9. The reaction of the two other cells showed similar courses of the temperature curves with different absolute temperature values.

In the first phase of the penetration, the cell heated up to 120°C ± 5°C starting at temperatures between 21°C and 30°C. The temperatures at the positive and negative pole showed a significant faster heat up then the temperatures in the middle of the cell. This might be caused by the local heat generation at the welding spots. Furthermore the massive copper clamps with its high thermal masses might heat up these areas due to previous tests and cause a thermal impact on the positive and negative pole – this explained the different start temperatures of the individual tests.

In case 1 (CID opens, smoke) the temperatures did not exceed the 120°C on the cell surface. The gases from the inside nevertheless showed significant higher temperatures of about 250°C. Picture 9 shows a typical cell behavior of case 1; the temperatures of the positive and negative pole steadily rose until the CID interrupted the current flow after 49 sec. The sensor in the middle detected a delayed temperature profile due to the thermal inertia of the jelly roll. These temperatures did not trigger any further hazardous situations. The voltage profile displayed a voltage drop to U = 2V beginning from the current flow and a negative voltage of U = -20V at the cell due to the CID reaction. In case 2 (CID opens, smoke, fire) the temperatures were within the same range, but up to 140 – 160 °C. In case 3 (thermal runaway) the temperatures easily exceeded the 500°C.



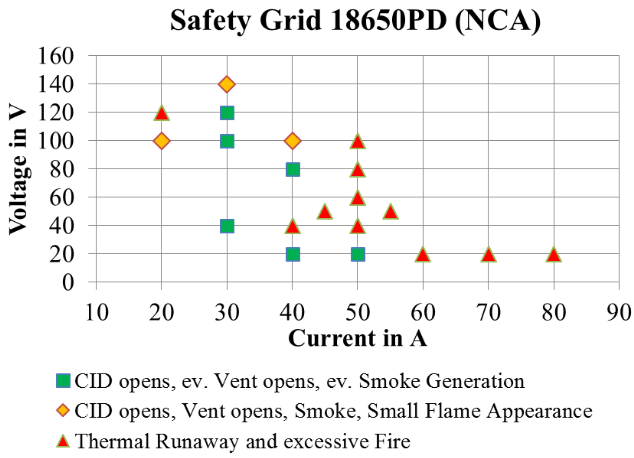
Picture 9: Case 1, benign thermal Reaction, LIC 18650PD, 50A / 20V

2) Safety grid of 18650PD

The test of the 21 specimens showed different reactions according to the voltage and current levels that the test LIC were confronted with. As shown in Picture 10, the cells showed benign and hazardous reactions depending on the penetration. The examination revealed, that current values over 40A seemed to cause a Thermal runaway more likely than values below 40A. To cause a triggering of the safety mechanisms, the current load required beside its value a certain time period for overriding. This can be explained by the gas production processes within the cell. The current flow heats up the active materials which then react chemically at certain temperatures after a certain amount of time. As soon as the inner pressure exceeds a threshold, the CID/vent tripping could be recorded.

The dependency of the voltage however was not significant. The test results showed no direct connection between voltage level and cell reaction. Even voltage levels up to 120-140V could not cause a hazardous reaction.

The safety mechanism of the CID and the vent showed a reliable triggering at voltages below 100V in combination with current values below 40A. Assigning these values to a LIC array design, a number of 23 serial cells might not cause a thermal runaway if the current flow can be limited to less than 40A for a time span of less than 100s.

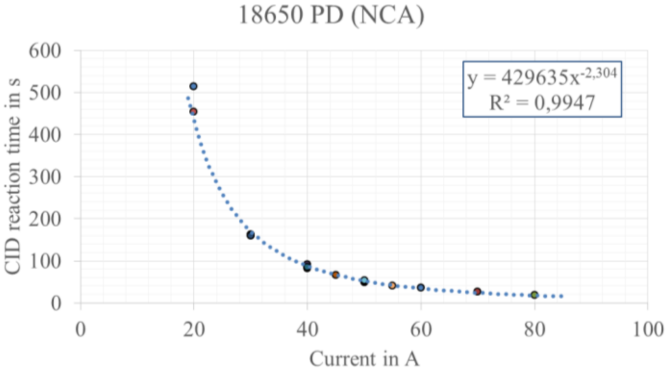


Picture 10: Safety Grid of LIC 18650 PD

The cells showed excessive fire and heat generation in the Thermal Runaway. In a first stage, the fire sparks out of the positive pole since the venting plate burst there in the first place. Secondly, the cell sparked out of the negative pole that has a notch in the casing. After a few seconds the LIC remained as a glowing rod, see Picture 11.

Picture 11: Thermal Runaway of LIC 18650 PD

The experimental data revealed a strong dependency between the response of the CID and the current load, the cell is confronted with. Picture 12 shows the reaction time Δt of the CID which is the timespan between the start of the current load until the first current interruption through the CID (either t_1 or t_2). Depending on the penetration, the cell went afterwards in a safe, potential hazardous or hazardous status.



Picture 12: CID response time of LIC 18650PD, 22 data points

At current values of 40A or higher, the reaction time was shorter than 100s and was even limited to less than 15s at 80A. All data points were located on an exponential curve, see formula in Picture 12. The accuracy of the fit was over 99 %. Only at low current values (<40A) the dependency was not that distinct. This may be explained by the slow activation of gas production processes which may vary in their response time.

B. NCM 18650AA

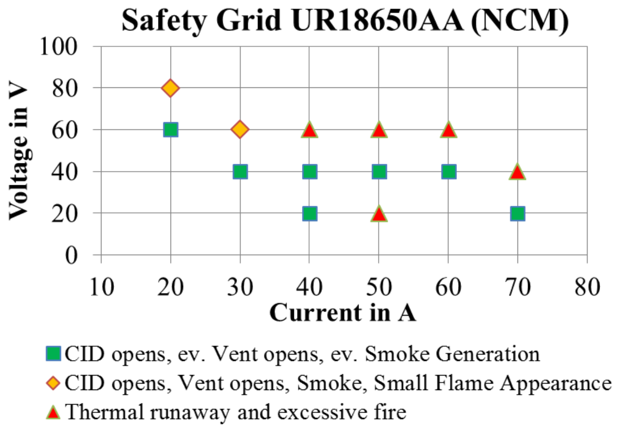
1) Thermal Reaction

The cells generally showed a similar behavior to the latter LIC but the temperatures rose generally up to 100°C until the CID reacts. The temperatures of the negative pole rose significantly faster to the 100°C; at this temperature, the value to the ones at the middle or the positive pole reached a lower temperature with a difference of 20°C to 42°C less.

In case 1 (CID opens, smoke) the temperatures generally did not exceed the 100°C on the cell surface. The gases from the inside nevertheless, showed significant higher temperatures of about 200°C. These temperatures did not trigger any further hazardous situations. In case 2 (CID opens, smoke, fire) the temperature range was about 120 - 140°C. In case 3 the temperatures easily exceeded the 500°C.

2) Safety grid of 18650AA

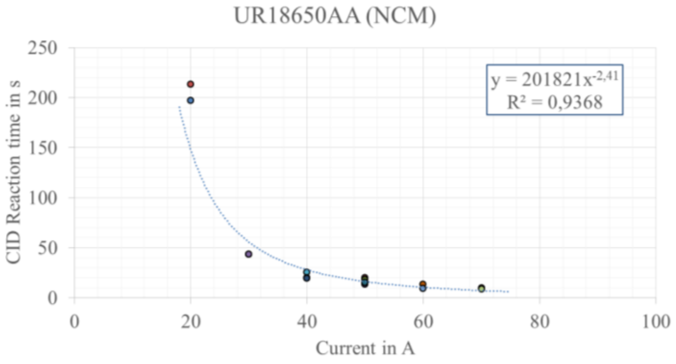
The 14 specimens showed all three reaction types depending on the voltage and current levels that the LIC were confronted with. Pictures 13 displays a dependency of the LIC reaction to the voltage level. If the voltage level was increased, the thermal runaway was likely even at low current values of 20A. Voltages below 50V showed a benign reaction up to 70A. Nevertheless at a load of 50A at 20V, a LIC went into a Thermal Runaway which counteracted a stringent relation between safety and voltage level. The dependency of the cell behavior to the current was not fully given.



Picture 13: Safety Grid of LIC 18650 AA

The response time of the CID showed similar dependencies to the current value as the results with the LIC 18650PD. As can be seen in Pictures 14, the response time Δt of the CID had an exponential trend that fits with an accuracy of 93% to the experimental data.

The tripping time of the hybrid 18650AA specimen was significantly shorter compared to the high energy cell 18650PD. At current values of 60 to 70 A, the CID response time was between 8.4 and 13.2 seconds. The CID in the 18650PD reacts after 36s (60A), 26s (70A) respectively 18.6s at currents of 80A.



Picture 14: CID response time of LIC 18650AA, 14 data points

C. LFP APR18650M1

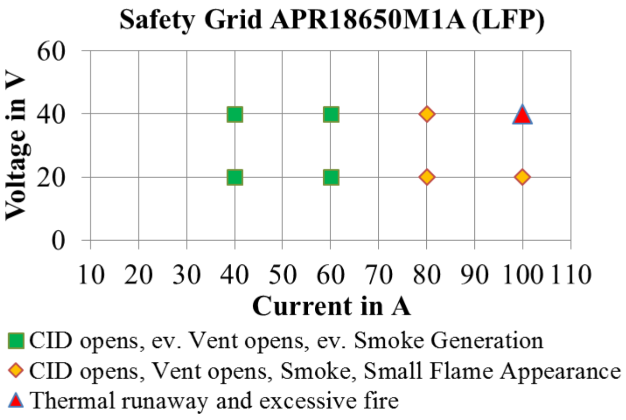
1) Thermal Reaction

The LFP LIC as a high power cell showed a different reaction to the high energy or hybrid LIC. In the phase until the CID reacted, the specimens heated up to different values between 90°C and 169°C until the CID trips. The temperatures in the middle was significantly lower than at the position of positive and negative pole. This may be caused by the distinct higher current values of 60 to 100A that caused a higher ohmic loss in the welding spots and therefore higher local heating. The temperature of the negative pole generally hit the 120°C in the first place while the temperature of the positive pole and the LIC can (middle) was about 10°C to 17°C lower. In the test, significant higher values were applied to cells since they explicit higher capability of high currents up to 3 times more (30A). The LFP-cells generally showed higher, darker smoke generation in case 1 and 2 compared to the other cells.

In case 1 (benign reaction of the LIC), the temperatures usually did not exceed the 120°C on the cell surface. The gases from the inside nevertheless showed significant higher temperatures of about 250°C. These temperatures did not trigger any further hazardous situations. In case 2 (undefined reaction) the temperature rose up to 250°C on the cell surface, caused by the little fire at the positive pole. Nevertheless the cells did not show a hazardous reaction. In case 3 (Thermal runaway) the temperatures exceeded the 400°C.

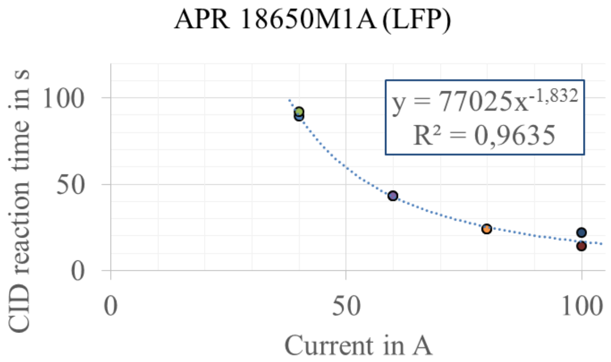
2) Safety grid of APR1650M1

The LIC in this test series generally showed less severe reactions compared to the LIC 18650PD and 18650AA. There has been neither such excessive Thermal Runaways nor intensive fire sparks. Nevertheless the measurement data gave indications of the CID reaction, so that all three cases could have been allocated to the specimens. Pictures 15 shows the dependency of the cell reaction to the current level. If the current was increased, the thermal runaway was more likely. The generally higher values of the current can be explained with the design, since the LIC with its LFP active material had the ability to provide continuous discharge currents of 30A. A significant dependency of the cell behavior to the voltage could not be found in the data set.



Picture 15: Safety Grid of LIC 18650M1

The response time of the CID showed stringent dependencies to the current value as well. As can be seen in picture 16, the response time of the CID has an exponential trend that fits with an accuracy of 96% to the experimental data. The CID response time however with over 42s at 60A and 24s at 80A was the highest of the three LIC types, which may also be explained by the high current design of this cell type.



Picture 16: CID response time of LIC 18650M1, 8 data points.

III. CONCLUSION

Within the 44 cell tests, different reactions of the CID were monitored. Generally it can be stated, that the CID works properly if the LIC is confronted with low voltages and low current values. However, if at least one of these parameters is increased, the CID may get overridden and the cell might go into a thermal runaway.

The severity of the thermal runaway of the high energy and hybrid cell show a higher degree of damage. Compared to the high power cell, the NCR18650PD cell with the highest capacity of 2.9Ah shows excessive fire and rupture. This can be explained with the reactive masses, since these are equivalent to the cell capacity. Furthermore the evaluation of A123, that the APR18650M1 cell does not show equally excessive thermal runaway behavior such as metal oxide LIC, can be confirmed.

In addition to this, the APR18650M1 show significantly higher smoke emission in case of a benign or undefined behavior. Compared to both other cells, the smoke has a more opaque color.

The test results of all cell types show that the CID reaction is significantly dependent on the current value. The high performance LIC ARP18650M1 shows a hazardous behavior only at current values over 80A whereas the high energy LIC NCR18650PD and the hybrid LIC UR186500AA show a hazardous behavior already at 20A, if the voltage level exceeds 60V.

Regarding the interpretation of the test results of single cells on a battery system level, an interconnection of cells in an array may lead under certain circumstances to hazardous situations. Already a serial string of 5 battery cells (Maximum string voltage U_{max}=21V) may cause a potential hazardous reaction if one of the cells is confronted with the system voltage and the current flow is achieved.

With NCR18650PD, a current value of 60A for about 36 seconds respectively 50A for about 13.2 seconds for the UR18650AA cell lead to a thermal runaway. Therefore already a small quantity of parallel cells providing the necessary current flow can become critical.

To avoid a hazardous module design for the single LIC by excluding potentially dangerous framework conditions on the electrical level, the number of serial LIC would have to be limited to 5. The parallel cell interconnection also would have to limit the maximum current flow through one cell to less than 50A over several seconds. These requirements would restrict the field of applications drastically since module voltage level usually exceed the 24V and 48V level.

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