



CELL-INTEGRATED SENSING FUNCTIONALITIES FOR SMART BATTERY SYSTEMS
WITH IMPROVED PERFORMANCE AND SAFETY

GA 957273

D3.3 – REPORT ON PROTOTYPING 1AH CELLS
WITH INTEGRATED LEVEL 1 SENSORS



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Summary

The deliverable D3.3 “Report on prototyping 1Ah cells with integrated level 1 sensors” summarizes the activities related to the Task 3.3 of work package 3 of SENSIBAT project.

The main objective of this task is to integrate level 1 sensor into novel prototype pouch cells. SENSIBAT level 1 sensors measure battery cell internal temperature and pressure through an integrated matrix that allows the read out with spatial resolution. Special care has been taken on integration of the sensor without changing the electrochemical behaviour of the prototype cells compared to the baseline cells (without sensors). In addition to the electrochemical behaviour, focus has been paid to the sealing, where the sensor exits the cell, to avoid leakage at this position. To sum up the main aspects of the sensor integration process are:

- Manual integration of level 1 sensors in 1Ah pouch cells
- Use of state-of-the-art standard electrodes (graphite anode and NMC622 cathode)
- Integration of sensors without changing the electrochemical behaviour of the prototype cells
- Ensure a sealing that avoids leakage

All the pouch cells are fabricated with stacked design i.e., anode/ separator/cathode layers will be stacked as many times as needed and connected in parallel to reach the overall electrode area and target cell capacity. Thus, fabricated cells will undergo a formation cycle at 0.5C in room temperature 25 °C. The idea of this task is to prove that there is no performance influence on the 1 Ah cells due to the integration of the sensor. In order to fulfil the objective of this tasks, the integration process of level 1 sensors has been divided into two different processes, due to the lack of functional sensors and with the objective of advancing with the task, first dummy sensors were implemented. This allowed to verify some parts of the integration process without the need of real sensor devices. Once the sensor elements were available, those were manually integrated into the pouch cells.

The changes in the cell manufacturing matrix proposed in Annex I of the Grant Agreement entail the need of some changes in the Tasks and Deliverables of WP3, modifying the lead of “Task 3.3 Integration studies of level 1 sensing functionalities in 1Ah prototype cells” from AIT to ABEE and the lead of Deliverable “D3.3 Report on prototyping 1Ah cells with integrated level 1 sensors” in the same way.

In addition, some delays in the development of level 1 sensors caused the delay of the activities to be carried out in the Task 3.3 and consequently an accumulated delay of 4 months in the present deliverable. The delay was related to the observed instability of the sensor in the electrolyte environment. This issue had to be solved, to allow the developed sensors to operate as foreseen. The necessary steps to ensure this operational functionality are described in this document. The resulting delay was carefully evaluated and is unlikely to lead to further delays in the SENSIBAT project.

Finally, this document also serves as a demonstration of the achievement of Milestone 3 of the SENSIBAT project “MS3 Proof of concept cells (1Ah) with integrated level 1 sensors”.



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Abbreviations

Symbol / Abbreviation	
MS	<i>Milestones</i>
NMC622	<i>LiNi_{0.6}Mn_{0.2}Co_{0.2}O₂</i>
PI	<i>Polyimide</i>
WP	<i>Work Package</i>



1 Introduction

The present deliverable D3.3 “Report on prototyping 1Ah cells with integrated level 1 sensors” provides information on the production of novel pouch cells with integrated SENSIBAT level 1 sensors. It describes the work performed in the frame of Task 3.3 regarding the sensor implementation and incorporates figures of the sensor level 1 integration itself. This report is accomplished by the addition of first electrochemical data and the direct comparison of this electrochemical data to baseline cells. In addition, the cells fabricated in this task will serve as a second baseline/reference cells for comparing with the cells integrated with level 2 sensors.

Since the work in the connected Task 3.1 displayed difficulties with the sensor encapsulation and its reaction with the electrolyte, some measures had to be undertaken. The present report describes these corrective actions to ensure the proper cell and sensor behaviour in detail. In this context, the integration process of level 1 sensors had been divided into two different processes:

- (i) In a first step dummy sensors were implemented, allowing the verification of some parts of the integration process.
- (ii) In a second step, the sensor elements with final layout were integrated and electrochemically characterized.



2 Cell Design and Setup

2.1 Cell Formats and Components

To be comparable to the baseline cells previously reported in the frame of D3.2 “Report prototyping baseline pouch battery cells”, the selected cell components and cell format was equal to the baseline cells. The data for the used NMC 622 cathode and graphite anode, as well as other baseline cell specifications can be found in the D3.2. The variation in the cell format between the partners ABEE and VAR (fabrication instruments available within the partners) should be mentioned at this place and will be considered in the evaluation. These differences were also reported within D3.2.

In line with the integration of a sensor component to the cell, additional cell components are added to the list of cell components used for baseline cells. Figure 1 shows the developed level 1 sensor, which will be placed on top of the stacked electrodes in the 1Ah cell. This level-1 sensor was developed in task 3.1 and is described in detail within the connected D3.1 “Report on adaptation of Level 1 sensors for incorporation into battery cells”.

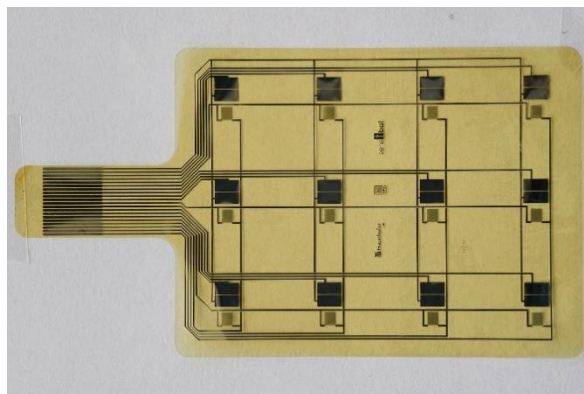


Figure 1: SENSIBAT Level 1 Sensor matrix



3 Sensor Integration

In the following section the measures undertaken to achieve a successful Level 1 sensor integration will be reported. Needless to mention, that the integration of a new cell component always results in an (at least slight) change of the cells operation behaviour. The aim of the presented activities was to reduce this influence on a minimum. A sufficient integration without changing the electrochemical behaviour of these cells compared to the baseline cells can be considered the fundamental condition for the success of the integration of such sensor devices.

3.1 Sensor positioning

In a first step, the project partners decided on the optimal position of the sensor. It was agreed that the preferable position for the feedthrough is between the feedthrough of the negative and positive electrode tabs. There are different reasons for the chosen position: The main reason for this positioning is related to the cell manufacturing, as the welding on this side of the cell must be done for the negative and positive electrode tab. Accordingly a third welding at this side for the sensor feedthrough is preferable, while the other sides of the cell remain unmodified. One must always keep in mind that such a welding area represents a weakness to leakage for the cell. As the lithium-ion cell environment is very sensitive to humidity and leakages (not only for safety but also for performance reasons) an accurate leak proofness must be ensured. Figure 2 displays a schema of the foreseen sensor (and sensor feedthrough) position.

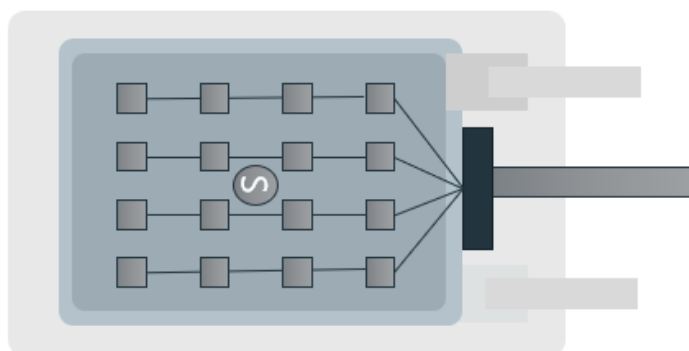


Figure 2: Drawing of 1 Ah cell with positioned level 1 Sensor

3.2 Providing spare volume for sensor related components

As indicated by the area with the black colour in Figure 2, the positioning between the negative and positive electrode tab, also provides some additional spare volume. This spare volume can be used for sensor related components. Of course, with respect to the final application of such sensors in commercial cells, the positioning and any demanded additional spare volume must be reconsidered individually.

3.3 Optimization of the feed through

As mentioned before, feedthroughs across the cell's outside containment as necessary for positive and negative electrode tab – and a sensor respectively – must always be considered a potential weakness area of the cell.

Such feedthroughs clearly increase the risk of potential leakages. And due to the humidity and leakage sensibility of every known lithium-ion cell with traditional liquid electrolyte, the absence of any leakages must be ensured.



ABEE and VAR performed various measurements comprising the tests of various PI-samples with different thicknesses and width. The target was to ensure accurate welding behaviour of the pouch-bag cells and to provide valuable information to project partners. The partners performed various measurements, indicated by Figure 3.



Figure 3: Welding test of PI-samples with different thicknesses and widths in combination with pouch bag foil (left), Leakage test of feed throughs by applying vacuum to pouch bag foils with integrated PI-samples (right)

The results of these tests were further distributed between the partners ABEE, VAR and AIT (responsible to produce sensor integrated 5 Ah cells), to give them indications how to handle the preparation of the feed through. Dependent on the companies individual sealing technology, a possible integration of an additional sealing tape is possible and could be recommendable.

Furthermore, these results were integrated in the layout design, performed by FHG-IISB in the frame of Task 3.1, with respect to the optimal width and thickness of the PI material. In the following and to verify the sensor feedthrough at an early stage, the described work eventuated in the processing of “Level 1 sensor dummies”.

Under the name of “Level 1 sensor dummies”, metallic dummy structures on PI, with an encapsulation of Parylene F-VT4 and an electrically connected via crimp, can be understood. These sensor structures are displayed in Figure 4.

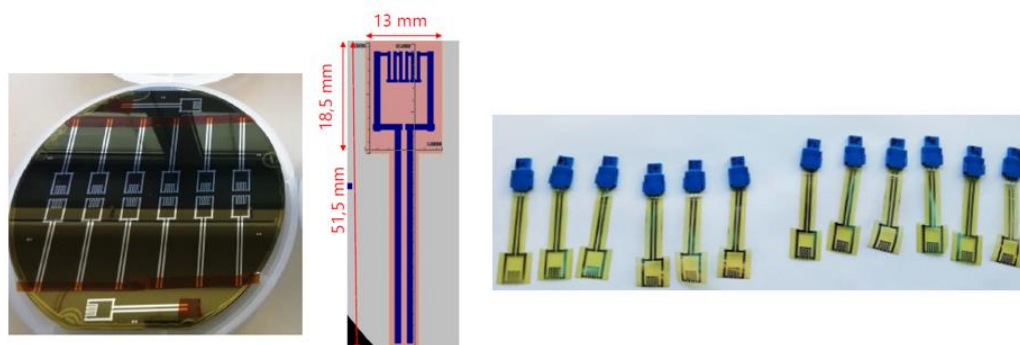


Figure 4: Level 1 Sensor dummies on waver during processing (left), their dimensions (middle) and before cell integration for feed through tests (right)

ABEEE and VAR used these sensor dummies and integrated them in test cells which are filled with electrolyte (VAR) and/or investigated the sensor stability in an excess of electrolyte (VAR and ABEE). Both partners detected comparable results displayed in Figure 5 and Figure 6.

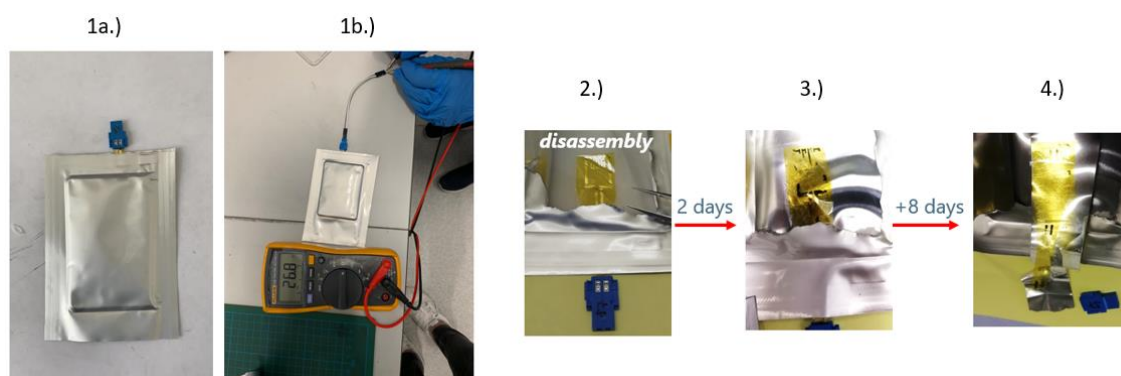


Figure 5: Pouch bag cells with separator and sensor dummy during measurement (1a., 1b.); the intact sensor dummy after direct disassembly (2.), the damaged sensor dummy when disassembled after 2 days (3.) and 10 days (4.) (VAR)

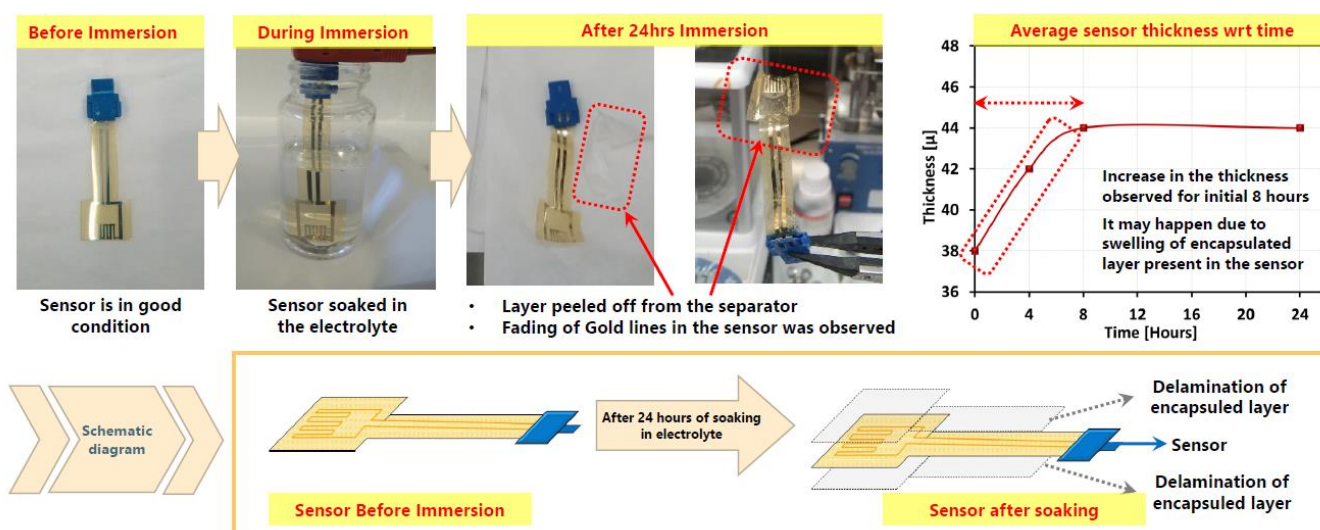


Figure 6: Delamination of the sensor structure with prolonged storage time in electrolyte (ABEE)

While the sealing quality in the feedthrough region of the cell displayed adequate stability, the measurements performed at the two involved partners revealed the dissolution of the aluminium through the electrolyte,



because of the delamination of the encapsulation layer. This critical finding will be described in more detail within the next chapter.

3.4 Sensor encapsulation of the sensors

Adequate encapsulation of sensitive (metallic) structures of the sensor against the aggressive electrolyte environment is essential to ensure sensor stability and functionality. The measurements revealed that the encapsulation originally foreseen does not meet the requested demands in such an aggressive medium. This was further indicated by the findings in Figure 7, where the ohmic resistance of such sensor dummies was observed over 1 month. Even the visual assessment displayed the delamination of the encapsulation layer, which is therefore no longer able to protect the sensor structures against the electrolyte.

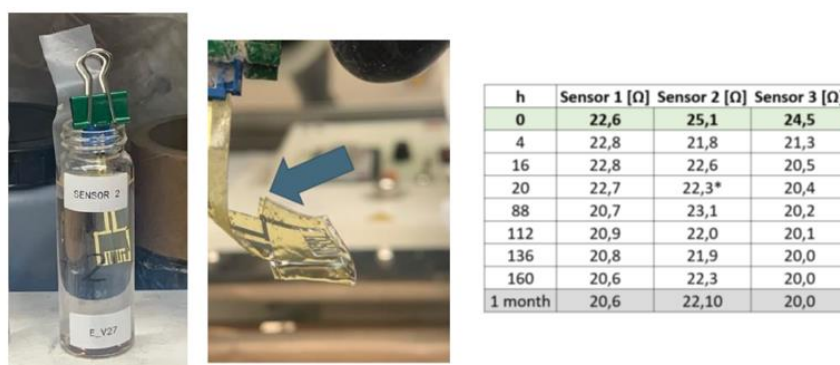


Figure 7: Sensor dummy stability test performed at VAR (left), displaying delamination (middle) and changes of the ohmic resistance over time (right)

As the observed instability of the sensor in the electrolyte environment must be solved to allow the application of sensors in the foreseen application, the partners undertook different approaches to solve this issue.

In a first step an additional tempering step at elevated temperatures was initiated. As comparable measurements to Figure 7 revealed an improvement, no satisfying result could be achieved. Consequently, different material combinations together with a tempering step were investigated. The results of these tests revealed that the application of Parylene C instead of Parylene significantly improves the sensor stability (indicated within Figure 8).



Figure 8: Out of the investigated alternatives, Parylene C allows superior stability in the electrolyte environment



Nevertheless, the sufficient encapsulation could be solved, the integration of a tempering step and the usage of a different encapsulation material had implications on the sensor layout. This is described in more detail in the frame of D3.1 and is mainly connected to a necessary change of the pressure sensor layout due to the tempering step. Connected to these necessary changes, the project partners requested to delay the D3.3, as well as the MS3 to month 19 of the project.



4 Cell production with level 1 sensor dummies and electrochemical assessment

To proceed with the investigations and to save time lost due to the implications stated before, the partners continued to manufacture cells with level 1 dummy sensors in a first step. It must be noted that these sensors have the same material setup as the final level 1 sensors and allow to investigate the sensor stability in the cell environment even though the layout is not final.

Accordingly, 1Ah cells with these level 1 sensor dummies were produced by ABEE and VAR. The Figure 9 below displays the examples of this cell production

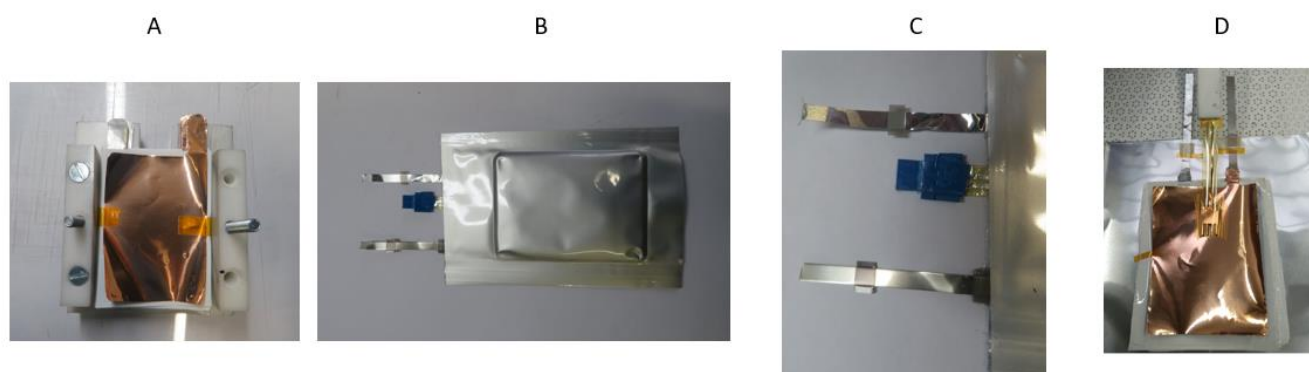


Figure 9: Electrode stack during construction at VAR (A), Finished cell with embedded sensor dummy (B), feedthrough area (C), Electrode stack during construction at ABEE (D)

These cells were electrochemically evaluated after construction. In this context, they were compared to baseline cells, to assess differences in the cell potential during charging/discharging. This is displayed in Figure 10, where in red the potential (and applied current according to test protocol) values of 3 cells with level 1 dummy sensors and in blue 3 baseline cells are displayed. A major influence of the sensor on the cell, or a significant destruction of the sensor devices would result in significant changes of the cell potential, which is not visible. Of course, there are slight differences of the cell's performance due to prototype like nature of the cells, but in general no influences of the embedded sensor devices are visually detectable.

This assessment is further supported by the comparison of measurements of a check-up cycle according to the defined test regime in D1.2. In Figure 11 this comparison is displayed. The shift in the cell capacity is due to electrolyte distribution well known for prototype cells and does not indicate problems. More severely would be a different potential pattern or "spikes" in the potentials, which are not observable in the measurements.

This is also supported by the up to now available results of the cycling studies displayed in Figure 12. Also, these measurements currently reveal no influence of the sensor on the cell performance. Due to the long time these measurements take, it also provides some first estimations regarding the sensor long time stability in electrolyte contact. In this context, the improvement of the sensor regarding the measures undertaken to ensure sufficient encapsulation can be estimated.

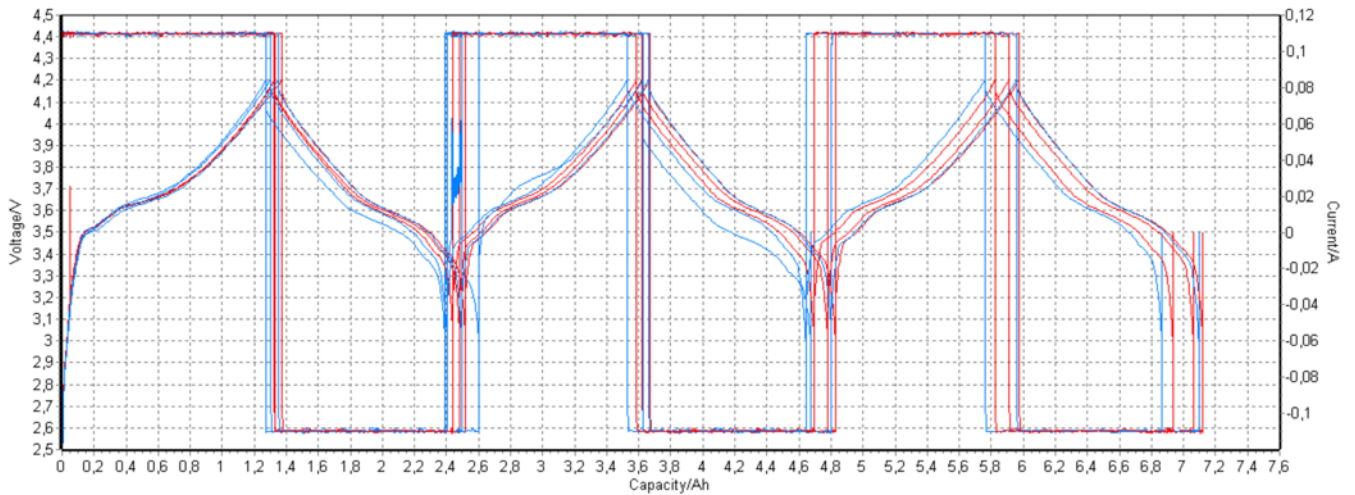


Figure 10: Formation data from VAR of 1Ah cells with dummy sensors compared to baseline cells; Potential and Current values of 3x 1 Ah cells with sensor dummies (red) compared to 3x 1Ah baseline cells without sensor (blue)

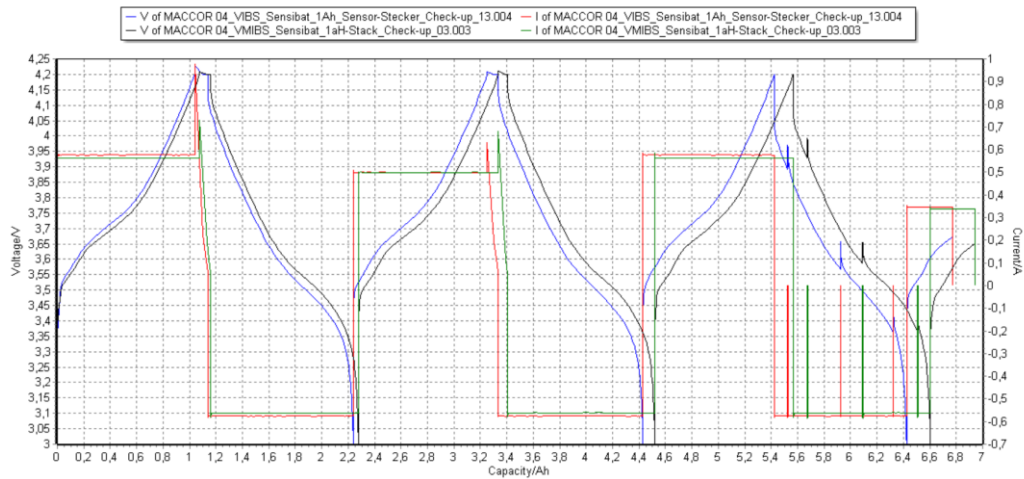


Figure 11: Comparison Check-Up cycling routine of 1 Ah baseline cell (voltage: black; current: green) and 1Ah cell with integrated sensor dummy (voltage: blue; current: red)

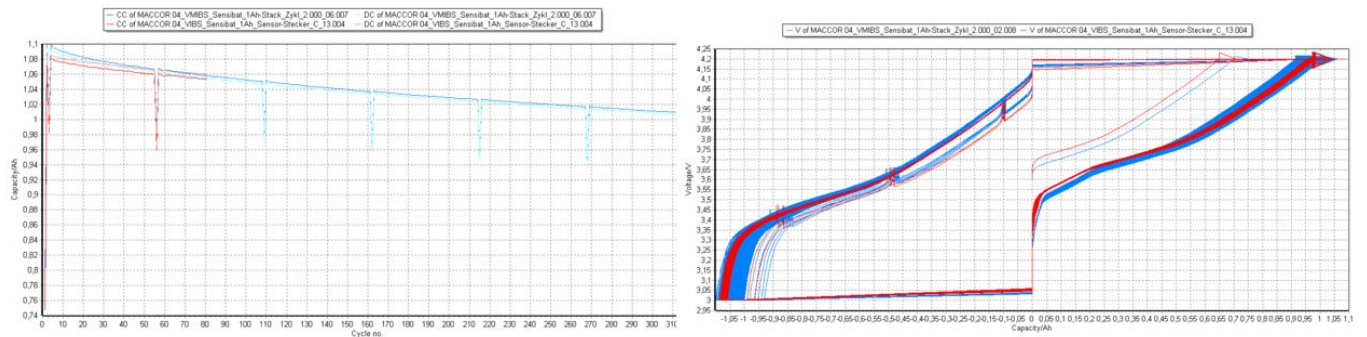


Figure 12: Comparison Cycle life testing – Achievable capacity per cycle (left; 1 Ah baseline cell: blue; 1Ah cell with integrated sensor dummy: red) and normalized potential vs. capacity values plot (right; 1 Ah baseline cell: blue; 1Ah cell with level 1 sensor)



5 Cell production with level 1 sensors (final layout) and electrochemical assessment

While test with sensor dummies on 1 Ah cells prolonged, the necessary adjustment on the final layout (see description above and D3.1) were finished. As a result, the partners were able to start the integration studies of level 1 sensors close to the finalisation of the present report.

Insights on the production process of cells with level 1 sensor at VAR are displayed in Figure 13 and insights on the production at ABEE is visible in Figure 14 and Figure 15 (Before starting the test, fabricated cells were sandwiched on the top and bottom aluminum pressure plates to hold the cells in the right place during cycling).

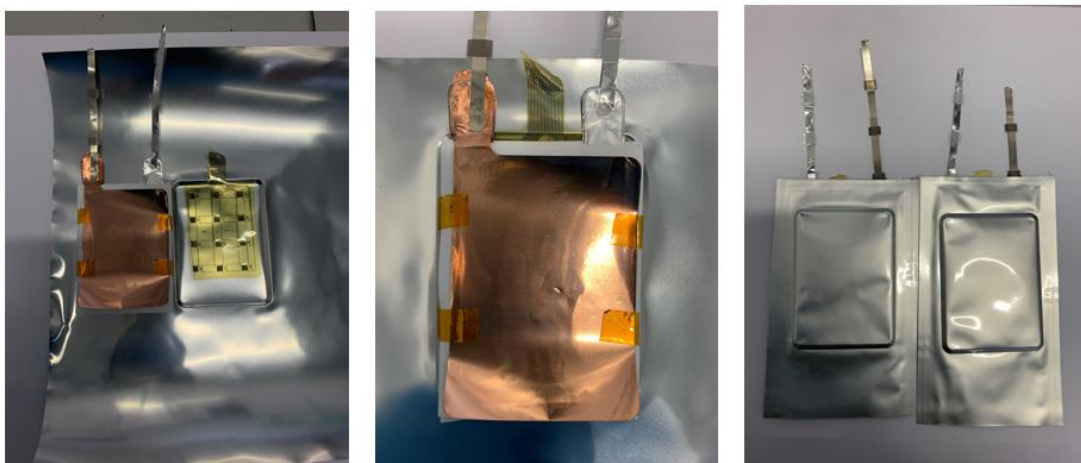


Figure 13: SENSIBAT 1Ah pouch cells with level 1 sensor produced at VAR

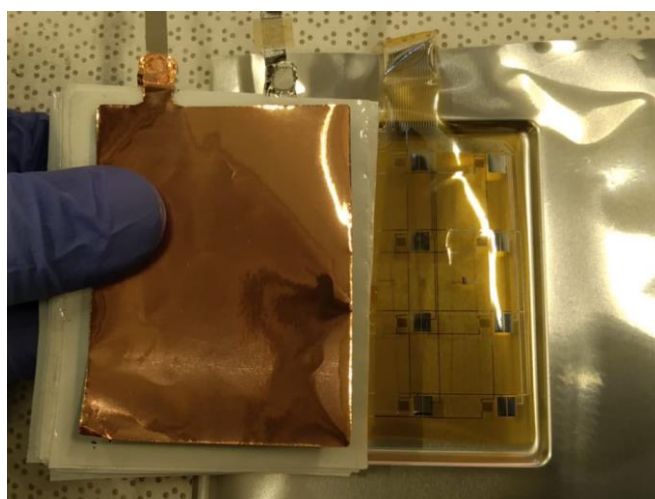


Figure 14: SENSIBAT 1Ah pouch cells with Level 1 sensor produced at ABEE

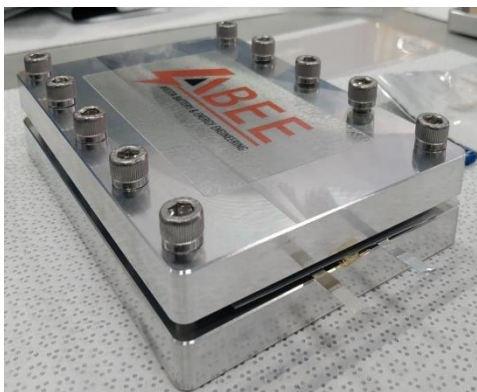


Figure 15: SENSIBAT 1Ah pouch cells with Level 1 sensor produced at ABEE with pressure jig

5.1 Electrochemical Results

Initially, the cells were preconditioned using the defined formation procedure in D1.2. The voltage profiles displayed below, show a good comparability of the produced cells to 1 Ah baseline cells, indicating no influence of sensor on the performance of the cell.

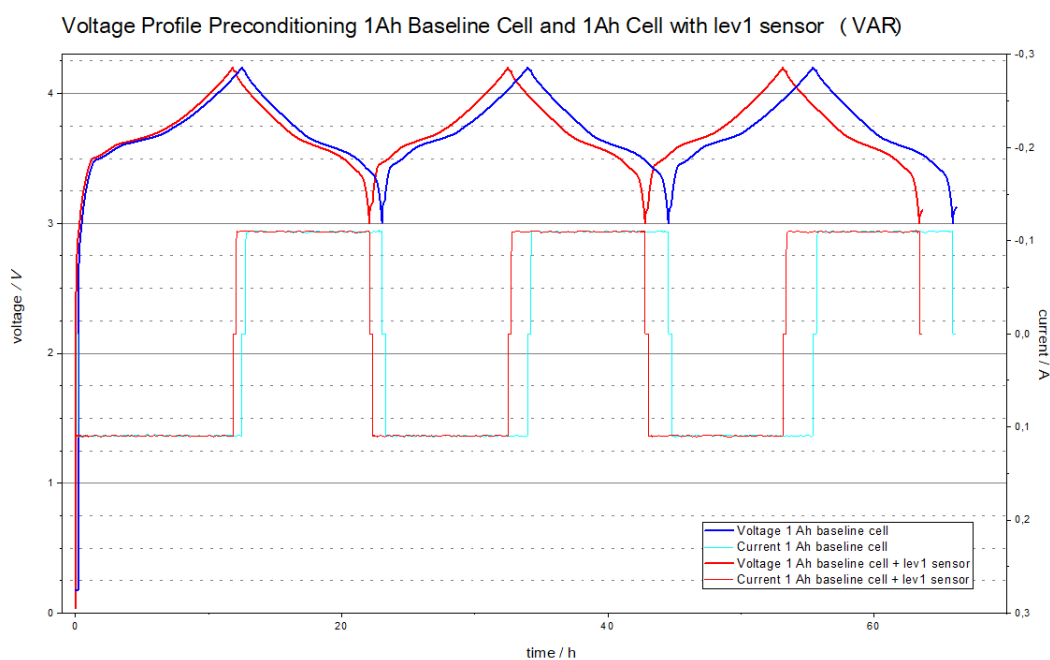


Figure 16: Comparison of Formation Cycle Voltage Profiles of Baseline cell and Cell with integrated level-1 Sensor

The comparison of the formation data (Figure 16) discloses again a timely shift of the potentials, attributable to slight differences in the electrode batches and electrolyte distribution. Important is the absence of any potential differences, as they would indicate major influences of the sensor on the cell's performance. In this context, also a performed check-up measurement (displayed in Figure 17) revealed the high comparability of baseline cells and cells with integrated level 1 sensor devices.

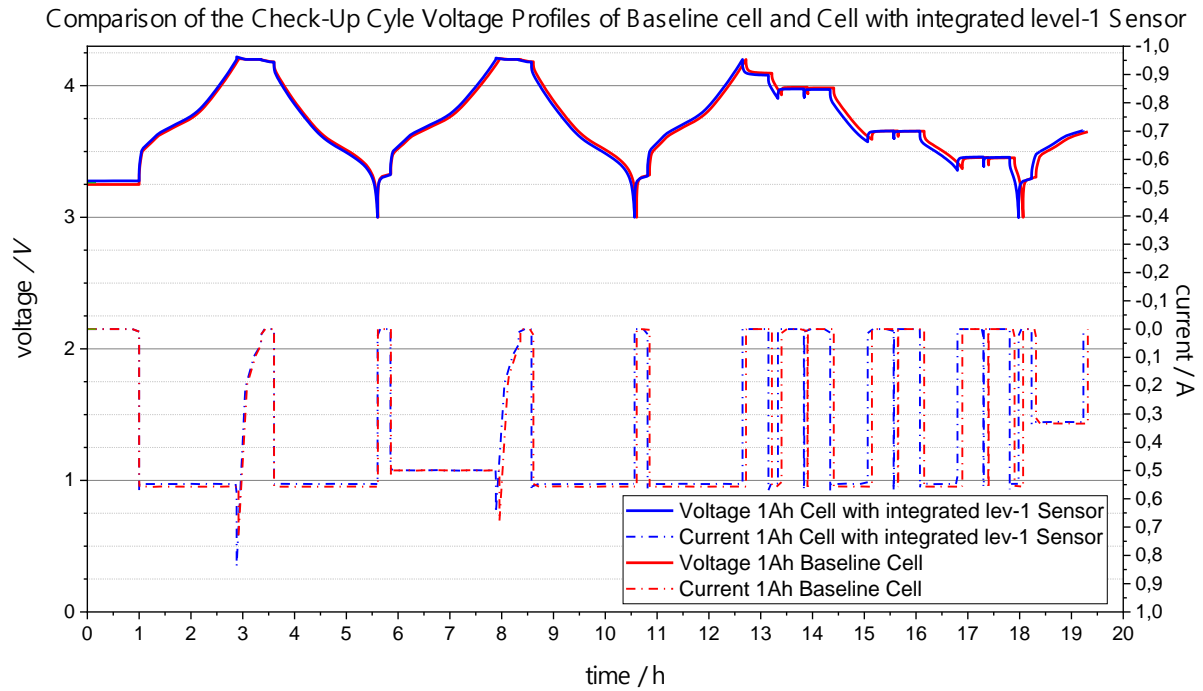


Figure 17: Comparison of the Check-Up Cycle Voltage Profiles of Baseline cell and Cell with integrated level-1 Sensor

Figure 18 displays the formation data of 1 Ah cells with integrated level 1 sensors prepared at ABEE, there were no voltage off shoots or other issues arising from the cell with the integration of level 1 sensors. The measurement is still ongoing and will be further updated.

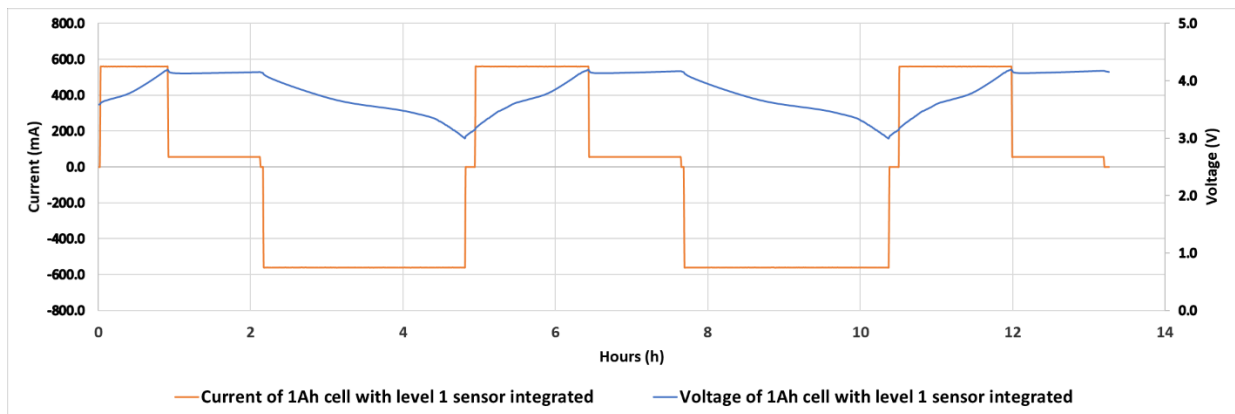


Figure 18: Voltage profile of the SENSIBAT 1Ah pouch cell with sensor produced at ABEE with applied jig



6 Discussion & Conclusion

The results presented in this document show that the objective of Task 3.3 and MS3, which required the integration of an internal temperature and pressure sensor within a novel li-ion cell in pouch format were achieved.

The results shown from the testing of cells with the level 1 sensors proves that there is no influence of the integrated level 1 sensors on the initial electrical performance of the constructed cells. The present deliverable describes this by comparing the first results of produced cells with integrated level 1 sensor with the results of tests previously carried out on identical cells without sensors, baseline cells. In the following months this comparison will be analysed in greater detail by means of different tests in WP5, among which prolonged degradation tests are included.

The deliverable also describes the progress to achieve a satisfactory integration of the level 1 sensor in 1Ah cells, as well as the measures undertaken due to the detected issue regarding sensor encapsulation stability. The difficulty of the sensor encapsulation, necessary due to the exposure of the sensor to the electrolyte, was overcome adopting material choice and integrating a tempering step. In addition, it has also been explained how, to avoid a greater delay, advances have been made in sensor integration techniques using dummy sensors.

As a consequence, the schedule of the deliverable and the linked milestone had to be postponed by 4 months, but it was carefully evaluated and can be stated that this will not further impact the projects progress.



7 Risks

Risk No.	What is the risk	Probability ¹	Effect ²	Solutions to overcome the risk
1	Sensors cannot withstand adverse environment in battery cell (e.g. may react with electrolyte to produce by-products) and lose sensitivity	3	1	The probability of R1 seems to be reduced by successful embedding of the Sensors observed by encapsulated sensor dummies. This has been proved in this deliverable.
2	Feedthrough of measurement contacts from the inside to the outside of the cell without leakage is not possible	3	1	R2 was successfully minimized from cell manufacturing point of view Priority can be considered "low". This will be monitored during ageing studies.
3	Incompatibility of the sensors with the pouch cell assembly process	2	3	Eliminated through the present deliverable and Deliverable 3.1. This risk can be considered completely eliminated
4	Integration of sensors without changing the electrochemical behaviour of the battery cell (e.g the transport or transfer of lithium ions between anode and cathode electrodes) is not possible	2	1	Risk has been minimized but will be monitored as cycling prolongs
5	The EIS with internal auxiliary electrodes (level 2) is too complex and expensive (both in extra hardware required and modelling) to be implemented	2	1	Risk has been minimized but will be monitored as cycling prolongs
9	Integration effort of the sensors higher than expected	2	1	Risk has been minimized
17	Pouch bag cells are typically operated between two plates (braced together) – attached sensor may result in additional mechanical stress	2	2	Risk has been minimized

¹ Probability risk will occur: 1 = high, 2 = medium, 3 = Low

² Effect when risk occurs: 1 = high, 2 = medium, 3 = Low



8 Acknowledgement

The author(s) would like to thank the partners in the project for their valuable comments on previous drafts and for performing the review.

Project partners

#	PARTICIPANT SHORT NAME	PARTNER ORGANISATION NAME	COUNTRY
1	IKE	IKERLAN S. COOP.	Spain
2	BDM	BEDIMENSIONAL SPA	Italy
3	POL	POLITECNICO DI TORINO	Italy
4	FHG	FRAUNHOFER GESELLSCHAFT ZUR FOERDERUNG DER ANGEWANDTEN FORSCHUNG E.V.	Germany
5	FM	FLANDERS MAKE VZW	Belgium
6	TUE	TECHNISCHE UNIVERSITEIT EINDHOVEN	The Netherlands
7	NXP NL	NXP SEMICONDUCTORS NETHERLANDS BV	The Netherlands
8	NXP FR	NXP SEMICONDUCTORS FRANCE SAS	France
9	ABEE	AVESTA BATTERY & ENERGY ENGINEERING	Belgium
10	VAR	VARTA MICRO INNOVATION GMBH	Germany
11	AIT	AIT AUSTRIAN INSTITUTE OF TECHNOLOGY GMBH	Austria
12	UNR	UNIRESEARCH BV	The Netherlands

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