



Li-S Cells for Space Applications (LiSSA)

ESA Contract No. 4000117343/16/NL/HK

Executive Summary Report

Project Details

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Document Revision

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1	18/01/18	1 st Draft	Ashley Cooke
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1. Introduction

The Lithium-Sulfur Cells for Space Applications (LiSSA) project is a project funded by the European Space Agency (ESA) which was conducted between 15th June 2016 and May 2018. The project proposal was submitted in response to ESA's invitation to tender (ITT, reference: AO/1-8425/15/NL/HK). In collaboration with CEA and Airbus Defence & Space (ADS/Airbus), the LiSSA project was aimed at developing OXIS Energy's lithium-sulfur (Li-S) technology for high energy density batteries for geostationary earth orbit (GEO) satellites. OXIS's Ultra Light cells at the start of the project were already capable of achieving 300-400 Wh/kg for at least 40 cycles at 100% DoD. An energy density of 400 Wh/kg and GEO cycle life of 1350 cycles until 80% beginning-of-life (BoL) was targeted for the LiSSA Project. Such a cell would offer substantial weight advantages to GEO satellites developed by Airbus and launched by ESA. The weight savings would allow for increased satellite payloads and/or significant cost reduction with reduced fuel usage.

2. Background

Common energy storage devices based on lithium-ion technology used in the space industry have reached a plateau. However the space industry still requires more energy to power the future missions as weight saving implies cost saving. Indeed, the electrical power subsystems designed today for space applications, are required to cope with new trends driven by the fast-growing business: more powerful payloads meaning more power stored on-board, global increase of mission duration, ever more constraining safety requirements and lower satellite masses for the battery subsystem. The particular characteristic that all battery engineers and researchers strive to optimise is the specific energy density.

Next generation Li-S technology is able to tackle the specific energy density weakness of the lithium-ion batteries. Indeed Li-S battery technology offers one of the highest theoretical energy densities of any rechargeable battery system known; with a theoretical gravimetric energy density of 2700 Wh/kg of active material (5 times that of currently used lithium-ion). Such a Li-S battery, developed by OXIS Energy, has shown impressive promises, particularly in terms of specific energy performances. The state of progress of Li-S batteries justifies that this is the next breakthrough technology for space batteries and that it will enable the specific energy to double again when compared to the current lithium-ion batteries.

The research on the Li-S technology has been pursued by various projects, which were concentrated on terrestrial applications, while a dedicated focus towards space applications is still missing. Given its potential, the Li-S technology can become a winning horse in the global competition for the most competitive space systems. Its potential as the next game changer technology for space is showcased in the listing of the main benefits below:

- Battery mass reduction of spacecraft and launchers
- Global increase of mission duration
- More powerful satellites payloads
- Less maintenance on ground
- Reduced costs

OXIS is the European leader in the development of Li-S technology. OXIS has already demonstrated over 1000 cycles and is in the commercialisation stage. Batteries with 300-400 Wh/kg have been successfully assembled and tested over the last two years.

OXIS received assistance from CEA, who possess extensive experience in Li-ion and Li-S battery technology, whilst Airbus is the European leader for space applications and provided their expertise to OXIS in order to assess the feasibility of OXIS's newly-developed Li-S cells for application in geostationary satellites.

3. Development Strategy and Project Outline

Following a comprehensive survey of the state-of-the-art in Li-S energy storage technology by CEA (TN1), a coherent development strategy was outlined which included research areas in materials development and optimisation of cell cycling conditions (TN2). After experimental assessment of each of the research strategies by OXIS, a preliminary cell design (designated Iteration 1) was proposed for testing. 22 cells of the same composition were produced by OXIS's Cell Production department based on this design and a variety of tests were conducted in order to understand the effects of various cycling conditions (voltage windows, C-rates, temperatures and rest periods) on performance, and to optimise conditions for maximised cycle life. These results were summarised in TN3. Following this work, a full-scale demonstrator cell design was prepared (TN4) and tested under a test plan devised by Airbus, CEA and OXIS (TN5). A demonstrator cell manufacturing report detailing the production development and scale-up was reported in TN6. Airbus conducted testing at their site which included GEO cycling as well as rate, temperature and calendar tests, summarised in TN7. A full stand-alone report of all activities conducted within the LiSSA project is provided here, documenting the development process from literature survey to full-scale evaluation of newly-designed OXIS Li-S cells for application in geostationary satellites. The broad strategy adopted within the project is summarised in Figure 1.

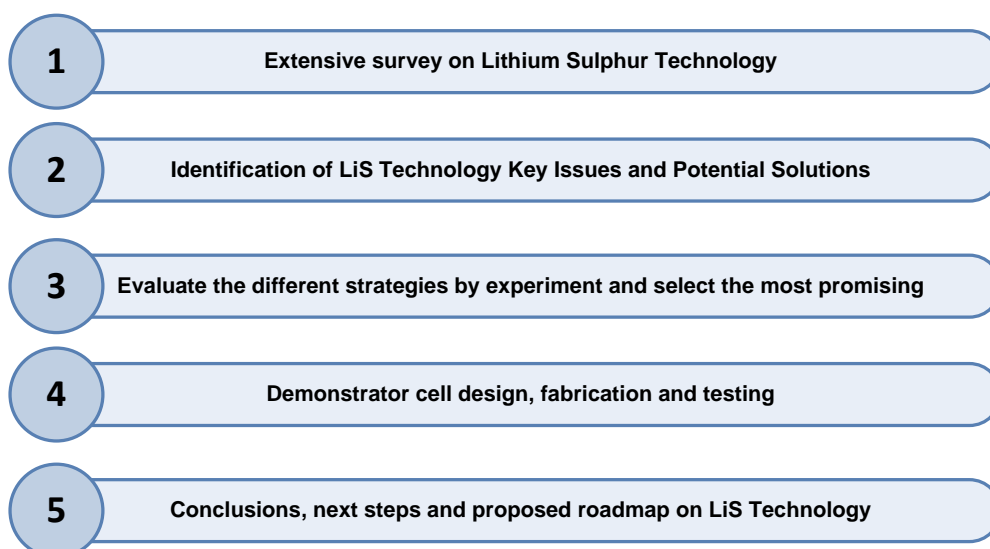


Figure 1: Broad strategy for the LiSSA Project to develop high energy Li-S cells for GEO applications.

4. Objectives

The main technical objectives as stipulated in the ITT statement of work (SoW) was to perform to following:

- An extensive survey of recent publications and patents on the Li-S state of the art.
- Identification of the key issues of Li-S technology for both space and terrestrial applications and a proposition of the most promising ways to solve these issues.
- To produce cells according to the requirements as per the SoW and to evaluate the developed Li-S technology by test.

5. Results

5.1. Materials Development

Various new materials in the cathode, electrolyte and separator were tested by OXIS during the development period, taking into account many of the most promising strategies outlined in the survey of Li-S technology by CEA. A novel cathode stabilisation method was optimised and showed greatly improved adhesion and integrity, which is understood to be particularly important so that high areal capacity cathodes are able to survive the large volume changes taking place during charging and discharging (Figure 2). Electrochemical testing demonstrated a three-fold improvement to cycle life with pre-treated cathodes under 70% depth-of-discharge cycling (Figure 3).

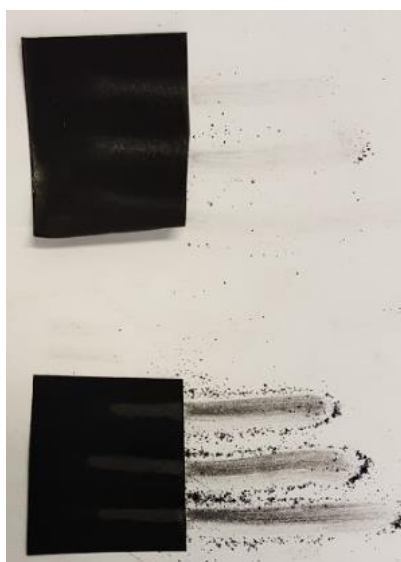


Figure 2: Cathode scraped without pre-treatment (left), cathode scraped after pre-treatment (right).

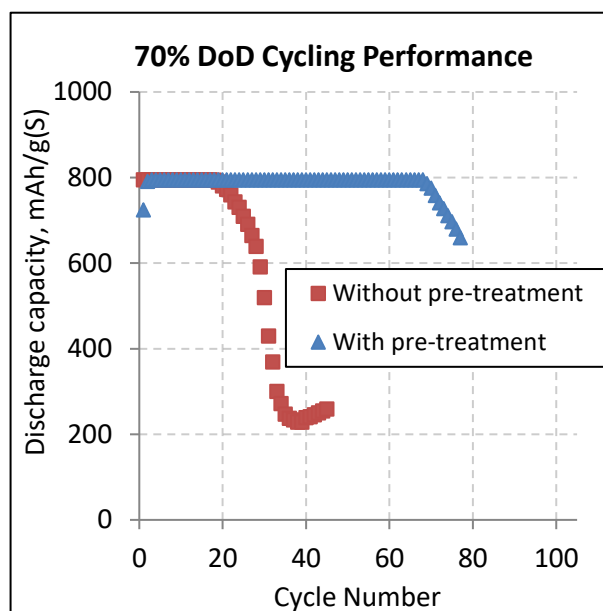


Figure 3: Pouch cells using cathodes with and without pre-treatment, cycled at 70% DoD, C/5 D/1.5, 1.90-2.45 V, 30°C.

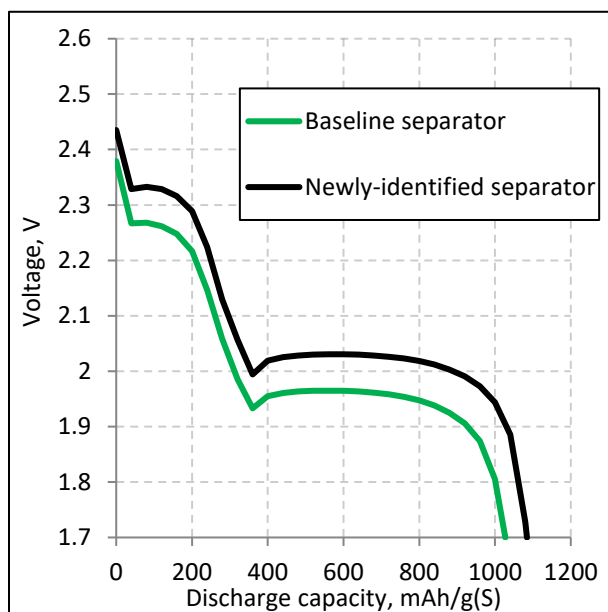


Figure 4: Pouch cells using different commercial separators, discharged at 1C, 1.70-2.60 V, 30°C.

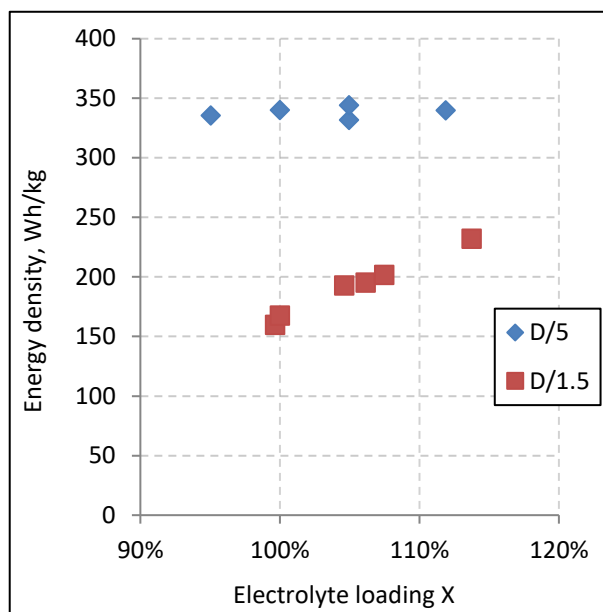


Figure 5: Energy density vs. electrolyte loading at D/5 discharge from fully charged to 1.9V (blue), and D/1.5 discharge 2.45-1.9V (red), 20°C.

Other advances included selection of electrolyte additives and tailoring of the electrolyte formulation for high energy density and cycle life. A new commercial separator was also identified from tests with nine different commercial separators that was able to deliver improved capacity at higher voltage (therefore higher energy) under a relatively high discharge rate of 1C (Figure 4).

Given the short timeframe for materials development during the project, only one new strategy for development of the lithium anode component was tested, namely, 3D micro-patterning of lithium foil.^{reference} By use of a nickel roller equipped with custom-designed patterns, lithium foils were pressed to endow the surface with various 3D micro-structures (grooves, pyramidal arrays, etc.). Whilst there was some evidence of reduced dendrite formation during the first plating of lithium, no improvement to cycle life was observed when micro-structured lithium was implemented in pouch cells.

5.2. Final Cell Design

Following the materials optimisation step, OXIS undertook a Production Development process to scale-up the technologies which had been previously demonstrated at small-scale pouch cell level. The manufacturing of materials (electrolyte, cathode, anode, separator, pouch etc.) at larger scale was tested and optimised, as were new assembly procedures and protocols for the scaled-up LiSSA cell design. During this process, cells were also discharged at various C-rates with different loadings of electrolyte. The energy density achieved on Production Development cells showed strong correlation with increasing electrolyte loading (Figure 5). This was due to the sensitivity of the second discharge plateau to electrolyte loading (and hence electrolyte viscosity) which is exacerbated at high discharge rates due to kinetic limitations. From this work, a high final electrolyte loading was selected, which was also anticipated to deliver improved cycle life as electrolyte depletion is a leading cause of capacity fade in Li-S cells. A total of 20 cells were manufactured and delivered to Airbus for testing. Figure 6 provides a schematic of the final cell design.

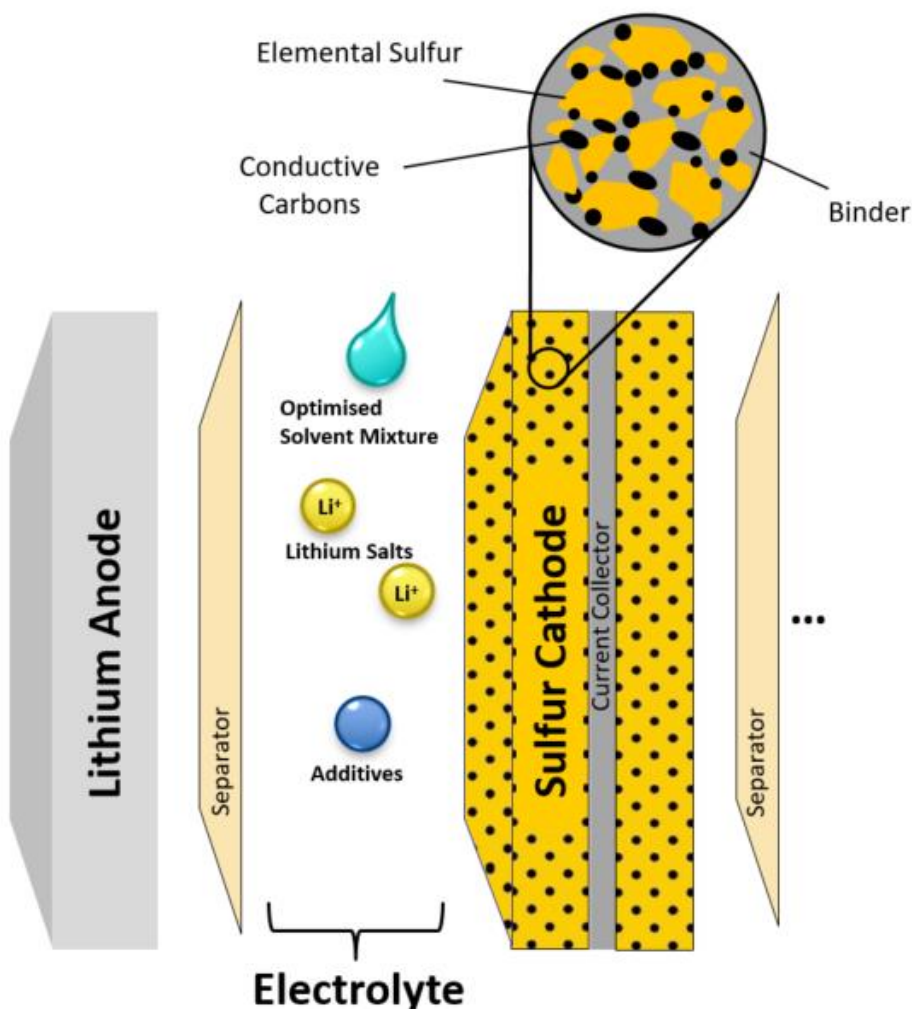


Figure 6: Design schematic of the final cell design. One repeat unit shown.

5.3. Final Cell Testing and Characterisation

The final LiSSA cells were validated by Airbus under different accelerated GEO cycling profiles, calendar tests, C-rate tests and temperature tests. Three cells were used for each test to ensure experimental reliability. The energy density was shown to be highly dependent on discharge rate and temperature; a very high energy density of 425 Wh/kg was achieved at a relaxed discharge rate of C/20 (30°C), although this was reduced to around 270 Wh/kg at C/2.5 (20°C).

Three GEO cycling profiles were tested, varying in charge rate and discharge power. A charge rate of C/5 was found to give improved cycle life compared to C/10, whilst a lower discharge power was found to favour high cycle life. Ultimately, a maximum cycle life of 4 seasons was achieved using C/5 charge and 9.8 W discharge. Whilst this is well short of the targeted cycle life, it represents a large increase compared to previous tests of Li-S cells for the application. Additionally, there were no signs of cell swelling or unsafe temperature increase observed for any cells.

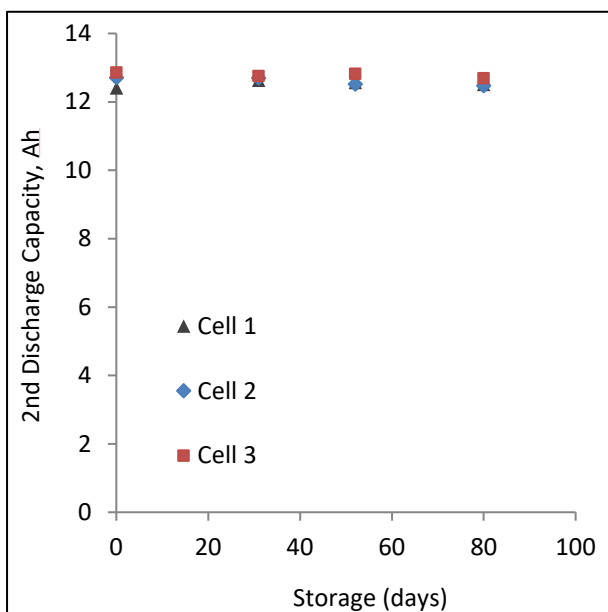


Figure 7: Reusable capacity during In-orbit storage conditions at 50% state-of-charge and 20°C. Reference capacities collected by discharging twice at C/2.5 and 20°C.

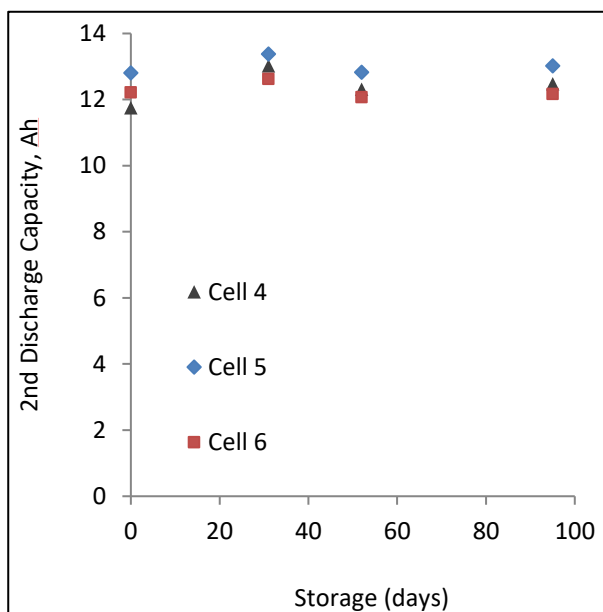


Figure 8: Reusable capacity during AIT storage conditions at 0% state-of-charge and 5°C. Reference capacities collected by discharging twice at C/2.5 and 20°C.

The cells performed well under two types of calendar ageing situations evaluated over a 3-month period. Firstly, cells were shown to have an average of -0.8% capacity fade over the 3-month test period when stored under in-orbit conditions at 50% state-of-charge (20°C). Secondly, cells were shown to have an average of +2.5% capacity fade over 3 months under Assembly, Integration and Test (AIT) storage conditions set at open circuit and 0% state-of-charge (5°C).

Temperature performance testing showed cells to give roughly the same energy density at 20°C and 30°C, with a 40% loss of capacity when discharged at 5°C.

Testing under different C-rates performance (at 20°C) revealed a large drop of 50% in energy density from C/5 to 1C discharging.

6. Conclusions and Roadmap

A high capacity cell was designed and manufactured after integrating technologies developed during a rapid period of materials research by OXIS, with strategies derived in part from a comprehensive survey conducted by CEA on state-of-the-art lithium-sulfur research (Figure 9). 20 cells were tested by Airbus under different GEO profiles, demonstrating a maximum cycle life of 4 seasons. A very high energy density of 425 Wh/kg was obtained at relaxed discharge rates (C/20) owing to the high areal capacity of the cathodes and newly-developed electrolyte formulation. Although this decreases to around 270 Wh/kg under the faster discharge rates required for GEO cycling (C/1.5), it still represents a markedly improved energy density over the incumbent cell technology. Cells showed high sensitivity to low temperature, retaining only 50% capacity at 5°C compared to nominal 20°C performance. Storage tests under in-orbit and AIT conditions displayed no significant loss of capacity over the test period.

6.1. Electrochemistry level – remaining challenges

Based on the results described above, the remaining challenges can be briefly summarized as follows:

- It is highly challenging to get high energy density along with high cyclability
- The cycling conditions are crucial to maximize the cell performances, and need to be optimized for each cell design, as the best conditions strongly depend on the cell chemistry and the application targets.
- Current best performances and high energy density target are obtained at medium C-rates (up to C/5), while higher current densities would be highly desirable.
- Calendar ageing is severe and the use of rest periods (or even OCV periods) is detrimental for the cell performances.
- The use of low charge rates (e.g. C/10) is also detrimental for the cell performances, while being highly desirable for the GEO application.

The following table summarizes how the proposed future perspectives could allow addressing the previously described challenges at a materials level. Each proposed strategy is also accompanied with its own risks and drawbacks, as well as differing timescales.

High energy density along with high cyclability	<ul style="list-style-type: none"> - Alternative sulfur/porous carbons composites - Development of functional binders - Development of 3D current collectors - Solvent-in-salt electrolytes - Polymer electrolytes - All-solid-state electrolytes - Lithium texturing - Polymer-based protections of lithium - Inorganic protections of lithium - Multilayered coatings of lithium
Reduced calendar ageing	<ul style="list-style-type: none"> - Sulfur confined into micropores - Development of functional binders - Solvent-in-salt electrolytes - Polymer electrolytes - Polymer-based protections of lithium - Inorganic protections of lithium - Multilayered coatings of lithium
Higher discharge rate	<ul style="list-style-type: none"> - Alternative sulfur/porous carbons composites - Sulfur confined into micropores - Development of 3D current collectors
Lower charge rate	<ul style="list-style-type: none"> - Sulfur confined into micropores - Solvent-in-salt electrolytes - Polymer electrolytes - All-solid-state electrolytes

6.2. Systems level – remaining challenges

From a systems-level perspective the following key criteria must be met to allow implementation into GEO satellites at high technology readiness level:

- Again, cycle life is crucial and must achieve >1350 cycles with <20% capacity loss in order to be used in GEO satellites. Robustness to different charge-discharge rates is also of high importance, allowing for easy integration into satellite power subsystems without loss of energy density or cycle life. In particular, ability to charge at very low rates allows the spacecraft to have an optimized solar array size.
- High energy densities must be met at GEO rates to show significant benefit/advantage over the incumbent lithium ion technology.
- The calendar tests show no degradation after 3 months, in the frame of the study. However, the duration is not sufficient to confirm the trend on several years of storage in open circuit or in floating. It is mandatory to know the degradation trend to embed Li-S on the GEO platform.
- Mechanical and thermal accommodation of the cells in a battery will have to be assessed if the previous criteria are met. The manufacturer would be required to demonstrate that the Li-S cell is compliant with space environment. If the present pouch cell design is not compliant, the cell housing shall be modified. In case of cells modification, the previous criteria must be demonstrated again with the new design.
- The safety of Li-S has to be certified with usual standards, to guarantee safe use in AIT and on-board (compliance with satellite passivation mandatory in the future).
- The last point of this roadmap is to demonstrate that the non-recurring and recurring costs for a new battery design and production could be competitive compared with Li-ion batteries.



Figure 9: Representative OXIS pouch cell