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Graphene, MXene and ionic liquid-based sustainable supercapacitor

GREENCAP - Deliverable report

D4.3. – Cylindrical Prototype Cells

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Author(s)	Paul Ionescu (SKL)	2025-06-16
Checked by	Paul Ionescu (SKL)	2025-06-25
Reviewed by (if applicable)	Tobias Burton (SOLV), Alix Ladam (SOLV) Alberto Morenghi (BED), Ahmad Bagheri (BED)	2025-06-21 2025-06-24
Approved by	Francesco Bonaccorso (BED) - Project coordinator	2025-06-30
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V1.1			
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Project summary

GREENCAP focuses on developing high-performance supercapacitors in an industrial scale and format that have significantly improved energy density and power density compared to existing products on the market by using graphene and MXenes as electrode materials and ionic liquids for high-voltage electrolytes. By using tailored materials to maximise specific surface area, ion accessibility, and charge storage, improved device properties are expected.

The GREENCAP consortium brings together academic and industrial partners from six EU member states, as well as the UK and Ukraine. Focused on advancing the energy storage sector, GREENCAP aligns with the EU's climate neutrality objectives and the Critical Raw Materials Action Plan. The project aims to validate supercapacitor technology at industrial scale (TRL 6) and develop a management system to enhance its integration into premium applications and support circular economy principles.

Publishable summary

Skeleton Technologies has manufactured electrochemical-double-layer-capacitors (EDLCs) prototype based on a novel configuration, which exploits ionic liquid (IL)-based electrolytes and curved graphene (CG). Double-side carbon coated aluminium electrodes were fabricated using an industrial production line, then assembled into a cylindrical D60 cell format. The cells were filled with novel IL-based electrolyte and are currently undergoing rigorous electronic testing to validate their properties.

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Abbreviations & Definitions

Abbreviation	Explanation
CG	Curved Graphene
DoA	Description of Actions
EDLC	Electrochemical double layer capacitor
EM	Electrode material
ESR	Electrical Series Resistance
IL	Ionic liquid
WP	Work Package

Item	Definition

1 Introduction

This document presents the design, fabrication, and preliminary evaluation of a cylindrical supercapacitor prototype developed using novel electrode materials (EMs) and ionic liquids (ILs)-based electrolytes. Supercapacitors, known for their high-power density and rapid charge-discharge capabilities, are increasingly vital in bridging the gap between conventional capacitors and batteries. Specifically, the prototypes are based on curved graphene (CG), fluorine-free and water-based binder, as described in D3.1, and IL-based electrolytes, exploiting $PyR_{13}BF_4$ and $Pyr_{11}BF_4$. The synergistic behaviour between the selected materials allowed to enhance the electrochemical performance and sustainability of energy storage devices. The prototype assembly process, material characterization, and performance metrics are detailed herein, laying the groundwork for future optimization and scalability, which will be described in D4.7 in M36.

2 Fabrication of Prototype

2.1 Electrode manufacture

The overall description of the entire prototype manufacturing is shown in Figure 2-1.

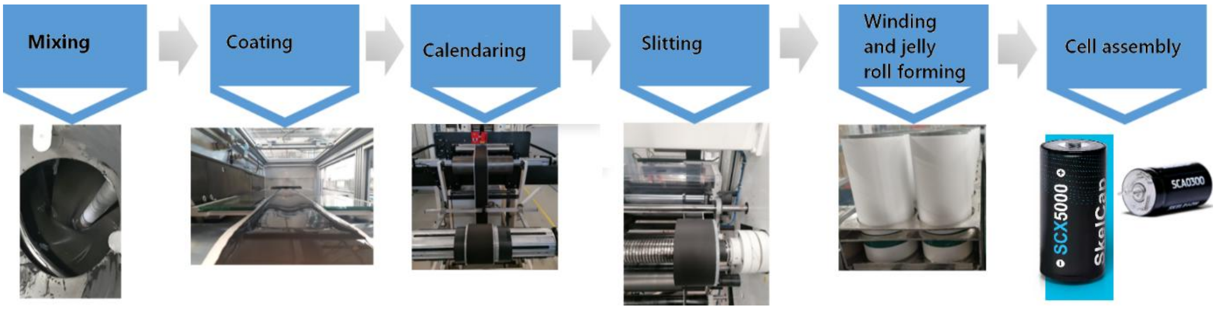

Figure 2-1 The fabrication process going from raw materials through to the coating of electrodes, calendaring, slitting, winding and finally cell assembly.

Specifically, the description of the electrode manufacture through mixing and coating can be found in detail in report *D4.2 – Industrial Chain Manufacturing*. The electrode is coated flawlessly, and its properties are listed below and are within the required parameters expected (Table 1 Measured properties of the electrode). The density has been slightly increased, while reducing the thickness by a calendaring process. In detail, the raise of the electrode density to 0.7-0.8 g/cm^3 allowed to maintain high electrical conductivity and to enhance the energy densities, both volumetric and areal, without compromising the rate performances.
The electrode coating width has been increased from 45 mm (as stated in the DoA) to 115 mm (Figure 2-2 a) for aligning with the supercapacitor standard format of Skeleton Technologies. Thus, the milestone M9 are therefore achieved.

Table 1 Measured properties of the electrode

Electrode loading	6 – 8 mg/cm²
Electrode density	0.7 – 0.8 g/cm²
Thickness of one-sided coating	100 – 150 µm

2.2 Electrolyte Filling

The electrodes are rolled up into a "jelly roll" configuration (Figure 2-2 a), inserted into a D60 cell format (60.2 mm diameter, 138 mm height). Subsequently, the cells underwent to the drying process, which is performed overnight in dynamic vacuum and 100 °C inside of a glovebox antechamber. Moreover, in order to ensure the total removal of trapped air/water, the system is repeatedly evacuated and filled with argon. The cans are vacuumed one final time prior to filling with electrolyte, as the low pressure aids in the efficient filling.

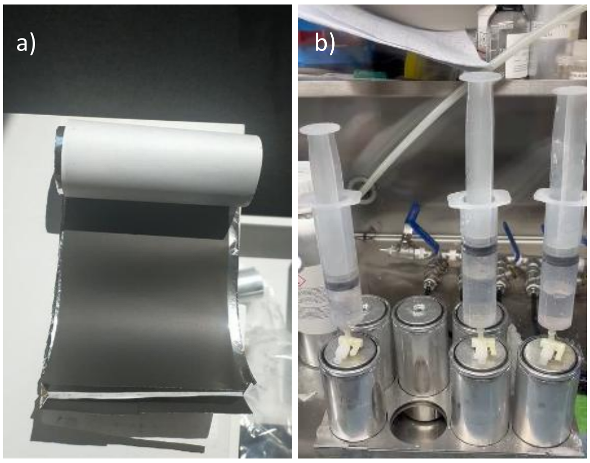

Figure 2-2 a) Electrode roll with a coating width of 115 mm; b)
Supercapacitors filling process with the IL-based electrolytes.

Two different prototype pairs were produced based on the electrolytes selected within the other WPs (WP1, WP2 and WP3), i.e. 1.2 M $Pyr_{11}BF_4$/ACN and 1 M $Pyr_{13}BF_4$/ACN, which are developed and provided by SOLV. Some issues arose during the filling process, as gas evolution was observed with both electrolytes, suggesting a contamination of the electrolyte (**Error! Reference source not found.**). Further investigations, like water and oxygen content evaluation are currently undergoing. The presence of oxygen/water contaminations may negatively affect the cell lifetime and, consequentially, the accelerated lifetime test. Therefore, a less stressful electrical testing approach has been adopted, decreasing the operating voltage of the cells from 3 V to 2.85 V, whilst maintaining the test temperature unchanged.

2.3 *MS 9 - The Finished Prototypes*

The prototypes, which are carefully sealed and welded following the procedure described in D4.2, satisfy the Milestone 9 target, even if the electrode parameters slightly differ. In fact, the cell parameters described in the "2.1 Electrode manufacture" section improved with respect the Milestone M9, determining a higher volumetric energy density. Thus, the Milestone M9 "Realization of a cylindrical SC cell with electrode loading of 4-6 mg/cm^2, density of 0.59-0.65 g/cm^3, thickness of 150-200 um and flawless coating width of 45 mm" is reached. An image of the prototypes is seen in **Error! Reference source not found.**.

Figure 2-3 The assembled prototypes ready for testing

3 Results & Discussion

5-cycle method capacitance and ESR

First, the cell capacitance has been evaluated performing 5 charging/discharging cycles at 0.1 A/g, in 0 – 2.85 V window, taking the average measurement of the last three of the five cycles. The gravimetric capacitance is calculated, which then is used to calculate the capacitance of the secondary cell based on the mass of the electrodes. Based on this measurement, a secondary cell is used in the 5-cycle method and charged to 2.85 V and discharged to 1.425 V with a constant 10 mA/F current. The discharge characteristics are then used to determine the capacitance and the ESR_{10ms} and ESR_{1s}.

Accelerated lifetime test

After the 5-cycle test, the cell is put in an oven and heated to 65 °C, at which point it is kept in a fully charged state at 2.85 V and recording the time.

The cell is then allowed to cool intermittently after pre-determined time intervals (120 h) and its properties are evaluated with the 5-cycle method. The cell testing is stopped once the cell capacitance drops below 80% of its initial value.

3.1 Results

Initially a 1 M N_{1113}FSI/ACN electrolyte was tested previously in a CG supercapacitor configuration. It was found that this electrolyte was not compatible with CG material, as unexpectedly low capacitance was recorded for this combination. As a result, this electrolyte was no longer pursued in the development of the prototype.

Subsequently, Greencap consortium selected the 1.2 M $Pyr_{11}BF_4$/ACN and 1 M $Pyr_{13}BF_4$/ACN as the prototype electrolytes. The supercapacitors were initially tested using a 5 cycles method, for capacitance at half-rated voltage and ESR (Figure 3-1).

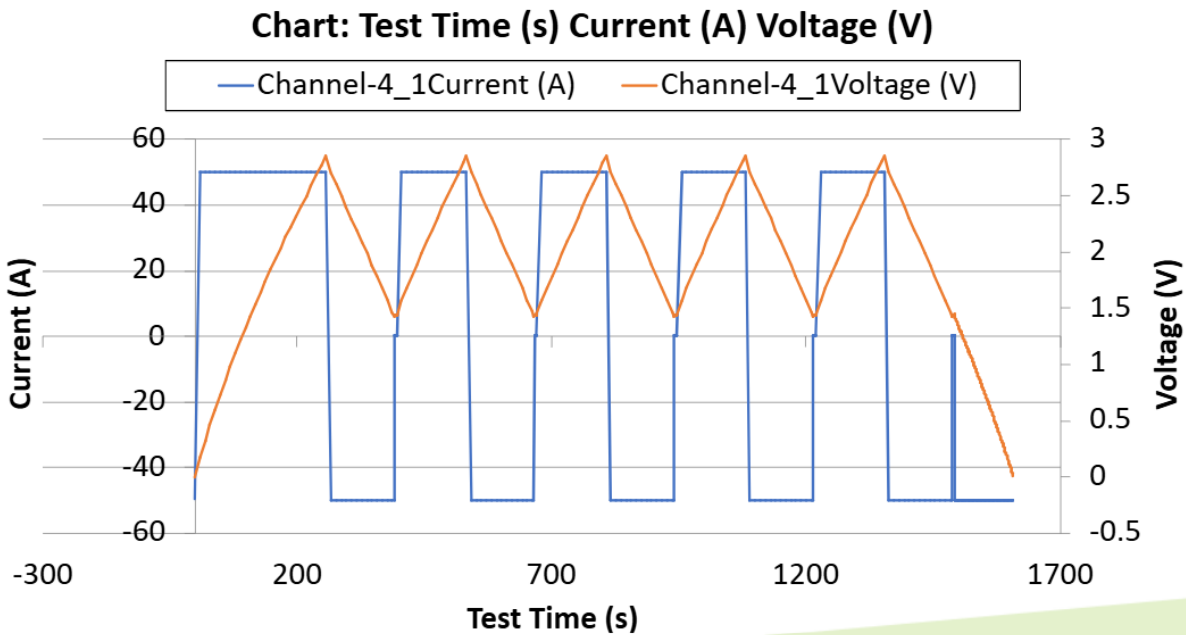

Figure 3-1 5 cycle test method using pulsed current on a $Pyr_{11}BF_4$/ACN cell up to 2.85 V. The behaviour of the device is as expected.

The cell behaves as expected. The data from the cycling tests was used to determine the 1s ESR and capacitance of the two different prototypes and compared with a Skeleton Technologies manufactured cell, Figure 3-2.

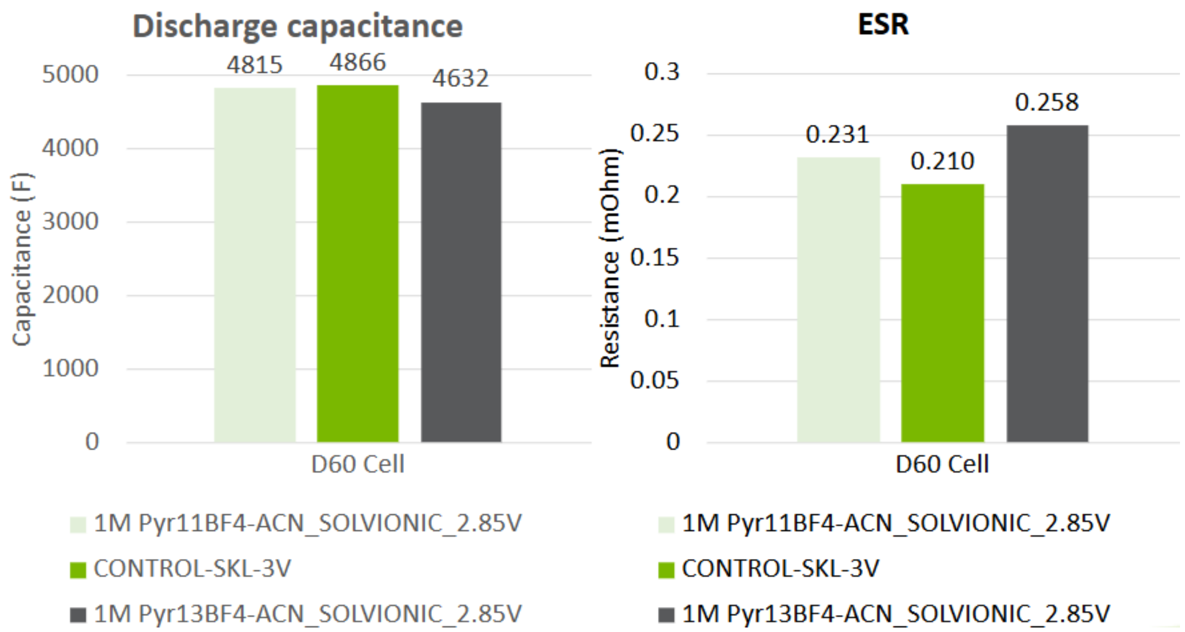

Figure 3-2 Discharge capacitance (left) and ESR (right) of the two prototypes manufactured compared with a control cell from the Skeleton Technologies production line.

It can be seen that the performance of both devices is quite similar to the control cell, however both performed worse for ESR measurements. This consequently will decrease the peak current that the devices can supply, as well as increase the thermal footprint of the device. $Pyr_{11}BF_4$ performed better in total capacitance than $Pyr_{13}BF_4$. The control cell is a reference which is fully optimized through iterative internal development outside of the GreenCap project.

Lifecycle testing and floating tests are currently being performed, however no data has been collected as of this point in time. They will be presented in the following reports.

3.2 Contribution to project (linked) Objectives

This deliverable shows the prototype manufactured in D4.3, and directly leads to the development of a mature, comprehensive supercapacitor management system as required by D4.4 and D4.8. Furthermore, the experience gained here will lead to completion of D4.6 industrial chain manufacturing, D4.7 Updated Cylindrical Prototype cells.

3.3 Contribution to major project exploitable result

Given a superior performance to commonly available supercapacitors, these prototypes stand to have a significant impact on the market, by leading with a strong edge over other manufacturers.

4 Conclusion and Recommendation

The prototypes have been manufactured and milestone M9 has been satisfactorily achieved. The supercapacitors must now be rigorously tested using floating and lifetime tests so their performance can be validated, benchmarked and evaluated in comparison to other devices on the market. Furthermore, the prototypes will be tested on capacitance, working voltage, lifetime, cycle life, working temperature and finally, a cost estimate will be created.

The Greencap project has evaluated multiple material combinations for a novel supercapacitor prototype and continues to pursue leads for improved properties. The new prototypes will be validated using the same testing as described in this report.

5 Risks and interconnections

5.1 Risks/problems encountered

Risk No.	***What is the risk***	***Probability of risk occurrence[1]***	***Effect of risk[1]***	***Solutions to overcome the risk***
WP4.3	*Electrolyte impurity leads to shortened lifetimes*	*Medium*	*Medium*	*A procedure to purify the electrolytes prior to the use in cells, as well as a secured transportation recommended.*
	cells do not significantly outperform existing ones on the market	*Medium*	*Low*	*Rigorous testing and validation of the devices, iterative development*

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

6 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:

#	Partner short name	Partner Full Name
1	BED	BEDIMENSIONAL SPA
2	SOLV	SOLVIONIC
3	FSU	FRIEDRICH-SCHILLER-UNIVERSITAT JENA
4	SKL	SKELETON TECHNOLOGIES OU
5	TCD	THE PROVOST, FELLOWS, FOUNDATION SCHOLARS & THE OTHER MEMBERS OF BOARD, OF THE COLLEGE OF THE HOLY & UNDIVIDED TRINITY OF QUEEN ELIZABETH NEAR DUBLIN
6	TUD	TECHNISCHE UNIVERSITAET DRESDEN
7	UNISTRA	UNIVERSITE DE STRASBOURG
8	SM	SKELETON MATERIALS GMBH
9	UNR	UNIRESEARCH BV
10	CNR	CONSIGLIO NAZIONALE DELLE RICERCHE
11	UCAM	THE CHANCELLOR MASTERS AND SCHOLARS OF THE UNIVERSITY OF CAMBRIDGE
12	CU	Y CARBON LLC

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7 Appendix A - Quality Assurance Review Form

The following questions should be answered by all reviewers (WP Leader, reviewer, Project Coordinator) as part of the Quality Assurance procedure. Questions answered with NO should be motivated. The deliverable author will update the draft based on the comments. When all reviewers have answered all questions with YES, only then can the Deliverable be submitted to the EC.

NOTE: This Quality Assurance form will be removed from Deliverables with dissemination level "Public" before publication.

Question	WP Leader	Reviewer	Project Coordinator
	Paul Ionescu (SKL)	Tobias Burton (SOLV)	Francesco Bonaccorso (BED)
1. *Do you accept this Deliverable as it is?*	Yes	Yes	Yes
2. *Is the Deliverable complete?* - *All required chapters?* - *Use of relevant templates?*	Yes	Yes	Yes
3. *Does the Deliverable correspond to the DoA?* - *All relevant actions preformed and reported?*	Yes	Yes	Yes
4. *Is the Deliverable in line with the GREENCAP objectives?* - *WP objectives* - *Task Objectives*	Yes	Yes	Yes
5. *Is the technical quality sufficient?* - *Inputs and assumptions correct/clear?* - *Data, calculations, and motivations correct/clear?* - *Outputs and conclusions correct/clear?*	Yes	Yes	Yes
6. *Is created and potential IP identified and are protection measures in place?*	Yes	Yes	Yes
7. *Is the Risk Procedure followed and reported?*	Yes	Yes	Yes
8. *Is the reporting quality sufficient?* - *Clear language* - *Clear argumentation* - *Consistency* - *Structure*	Yes	Yes	Yes