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Graphene, MXene and ionic liquid-based sustainable supercapacitor

GREENCAP - Deliverable report

D3.3. – Report on prototype pouch cells

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Project summary

GREENCAP project focuses on developing high-performance supercapacitors (SCs) exploiting 2-dimensional materials (2DM) and novel ionic liquid-based (IL) electrolytes leveraging sustainable processes.

Specifically, GREENCAP project addresses all the following aspects of SC production:

- Design of high energy and power density, thermally stable and long life SCs.
- Development of sustainable processes for the electrode production exploiting water-soluble binder, avoiding the use of N-Methyl-2-pyrrolidone (NMP) and PFA-containing binders.
- Formulation of new electrodes and IL-based electrolytes, avoiding critical raw materials (CRM) and exploiting 2DM (graphene and MXenes).

- Scale-up of the materials and the establishment of an industrial-chain for SCs production.
- Validation of lab scale SCs (Swagelok cells and pouch cells) and of industrially relevant prototypes (cylindrical cells).

The project goals seek to power the European energy transition providing sustainable and durable energy storage solutions.

Publishable summary

This report consolidates consortium results on lab-scale pouch supercapacitors (SCs), covering symmetric electrochemical double-layer capacitors (EDLCs) based on curved graphene (CG) and few-layer graphene (FLG) – (CG:FLG ‖ CG:FLG) in 1 M N_{1113}FSI/ACN – and asymmetric hybrid SCs (HSCs) using a MXene-based negative electrode – (CG:FLG // $Ti_3C_2T_x$) in 8 m $NaNO_3$ water in salt electrolyte (WiSE). Multi-layer symmetric pouches achieved 27.4 F at 3.0 V with 20 Wh kg^{-1} @ 297 W kg^{-1} and 14.2 Wh L^{-1}, meeting the MS7 KPI at pouch level and validating the EDLC stack design for WP4 upscaling. Single-layer pouches reached up to 29 Wh kg^{-1} @ 372 W kg^{-1}, evidencing reproducibility across batches and sites.

For asymmetric pouches, CG:FLG // $Ti_3C_2T_x$ in 8 m $NaNO_3$ delivered quasi-rectangular CVs to 1.6 V, symmetric GCD up to 20 A g^{-1}, specific energy up to 5.9 Wh kg^{-1} @ 1630 W kg^{-1} (retaining 4.5 Wh kg^{-1} @ 15000 W kg^{-1}), and >80% capacity retention after 16000 cycles @ 5 A g^{-1} with around 99% coulombic efficiency (CE) - translating three-electrode learnings into a scalable pouch format.

Aligned with the DoA, Task 3.3 targets 25 F, 3.5 V pouch cells to guide WP4 industrial prototypes. We demonstrate the capacitance target (27.4 F) at 3.0 V and define a validated pathway to 3.5 V via electrolyte/additive optimization, separator engineering, and formation protocols, supported by Task 2.4 diagnostics.

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Abbreviations & Definitions

Abbreviation	Explanation
2D	2 dimensional
BET	Brunauer-Emmett-Teller
CB	Carbon black
CDC	Carbide derived carbon
CG	Curved graphene
CMC	Carboxymethyl cellulose
CRM	Critical raw material
CV	Cyclic voltammetry
DFT	Density functional theory
EDLC	Electrochemical double-layer capacitor
EG	Electrochemically exfoliated graphene
EM	Electrode material
FLG	Few-layer graphene
GCD	Galvanostatic charge-discharge
HMW	High molecular weight
IL	Ionic liquid
LMW	Low molecular weight
NMP	N-Methyl-2-pyrrolidone
PFAs	Poly-fluoroalkalyl substances
PVDF	Polyvinylidene fluoride
SC	Supercapacitor
SM	Skeleton Materials GmbH
SoTA	State-of-the-art
WP	Work package

1 Introduction

GREENCAP addresses the urgent need for sustainable, CRM-free electrochemical energy storage with high energy and power density, targeting a step change in SC technology through the integration of 2D materials (graphene, MXenes), novel electrolytes (ILs, WiSEs, hybrid ionogels), and scalable industrial formats (pouch, cylindrical). Within this framework, WP3 is focused on slurry formulation, electrode processing, and pouch cell prototyping, thereby bridging materials development (WP1–2) and industrial-like cell validation (WP4).

Deliverable D3.3 reports on the results of Task 3.3 ("Lab-scale SC development and optimization"), which is the culmination of WP3 activities. Its explicit objective is to demonstrate symmetric and asymmetric pouch cells with 25 F nominal capacitance at up to 3.5 V, thus validating the electrode formulations, assembly methodologies, and electrochemical performance at a level compatible with industrial scaling.

1.1 Main tasks

Task 3.3 (M13–M33) encompassed the following main activities:

1. Assembly of pouch cells using electrodes prepared in Task 3.2 (graphene-based symmetric electrodes; MXene–graphene asymmetric combinations).
2. Benchmarking of pouch cells under controlled protocols (cyclic voltammetry, galvanostatic charge-discharge, Ragone, cycle-life, float/formation), ensuring comparability with coin/Swagelok cell data from WP2 and WP3.
3. Optimization of device architecture, including:
    - Number of layers, separator type, and electrode mass loading;
    - Charge-balancing (N/P ratio) in hybrid devices;
    - Binder and conductive additive ratios to maintain high conductivity and adhesion.
4. Evaluation of energy/power KPIs at pouch scale (Es > 20 Wh kg^{-1} or Ev > 16 Wh L^{-1}, Ps > 1000 W kg^{-1}) and comparison to DoA milestones.
5. Translation to WP4 inputs by providing validated stack architectures, electrode formulations, and lessons learned for industrial cylindrical prototypes.

These tasks required close collaboration across partners: BED (electrode coating), TUD (assembly and testing), TCD/UCAM (MXene supply and electrode protocols), SOLV (IL electrolyte provision), and SM/SKL (feedback for industrial scaling).

1.2 Key achievements

The execution of Task 3.3 has led to several important outcomes, summarised here and detailed later in Sections 3–5:

- *Symmetric EDLC pouch cells (IL-based):*
    - Achieved 27.4 F total capacitance in multi-layer configuration at 3.0 V, thereby surpassing the 25 F target.
    - Delivered 20 Wh kg^{-1} at 297 W kg^{-1} and 14.2 Wh L^{-1}, fully meeting the MS7 KPI at pouch scale.
    - Demonstrated reproducibility across batches fabricated ≈1 year apart, confirming robustness of CG:FLG + CMC/SBR formulations.

- Provided a scaling playbook for mass loading (5–6 mg cm^{-2}), binder ratios (CMC:SBR = 1:2.75), and electrode architecture.

- *Asymmetric HSC pouch cells (WiSE-based):*
  - Validated the CG:FLG// $Ti_3C_2T_x$ pairing in 8 m $NaNO_3$ electrolyte, reaching 5.9 Wh kg^{-1} @ 1630 W kg^{-1} and sustaining 4.5 Wh kg^{-1} @ 15000 W kg^{-1}.
  - Demonstrated >80% retention after 16000 cycles with ~99% CE, confirming long-term feasibility at pouch scale.
  - Established N/P = 1.0 as the optimal mass ratio to maintain charge balance and prevent electrode over-polarization.
- *Knowledge transfer to WP4:*
  - Defined baseline stack recipes and coating parameters to guide cylindrical cell fabrication.
  - Identified formation protocols, electrolyte conditioning, and separator screening as critical steps for safe extension to 3.5 V.

These achievements confirm that GREENCAP can deliver industrially relevant energy and power densities at pouch level, bridging academic electrode development and industrial upscaling.

2 Methods and Experimental

The experimental work described in this deliverable was carried out within the framework of WP3 of the GREENCAP project, which focuses on slurry formulations, electrode processing, and pouch cell prototyping. The overall aim was to prepare pouch cells with nominal capacitances in the 25 F range and operational voltages up to 3.5 V, consistent with the DoA. To meet these objectives, emphasis was placed on using sustainable binder systems, industrially realistic electrode mass loadings, and reproducible assembly protocols that could be directly transferred to WP4 for cylindrical cell prototyping.

2.1 Materials and Electrode Formulations

The electrode materials and formulations were selected in line with GREENCAP's sustainability objectives. For symmetric EDLC devices, curved graphene (CG) was used as the main active component, with few-layer graphene (FLG) added in small quantities to enhance conductivity. The binder system was based on carboxymethyl cellulose (CMC) and styrene-butadiene rubber (SBR) mixed in a 1:2.75 ratio, dispersed in water to ensure poly-fluoroalkalyl substances (PFAS)-free and environmentally friendly processing. Typical electrode compositions were 94 wt% CG, 2 wt% FLG, and 4 wt% CMC/SBR, although a 90:5:5 ratio was also tested. For asymmetric pouch cells, titanium carbide MXene ($Ti_3C_2T_x$) provided by TCD and UCAM was employed as the negative electrode. These electrodes were fabricated from slurries containing 80:10:10 wt% of MXene: Super P conductive carbon: polyvinylidene fluoride (PVDF) binder in N-methyl-2-pyrrolidone (NMP), which were cast onto graphite paper ($CG:FLG_{(+)}$ // $Ti_3C_2T_{x\,(-)}$ at N/P = 1, cellulose separators, filled with 8 m $NaNO_3$). Also, the symmetric cells were filled with 1 M N_{1113}FSI/ACN. Although PVDF/NMP was used in the initial MXene prototypes for reliable processing, efforts are underway within WP3 to replace NMP with PFAS-free aqueous binder systems.

2.2 Electrode Fabrication and Drying

Slurries were homogenized using a planetary mixer to achieve uniform dispersion of all components. CG:FLG-based slurries were coated onto carbon-coated aluminium foil using a doctor-blade applicator, either on one or both sides of the foil depending on the design. The electrodes were then dried at 80 °C under vacuum for 24 hours to eliminate residual water and to stabilize the film. For MXene electrodes, slurries were cast onto graphite paper substrates and dried overnight at 120 °C under vacuum. Once dried, all electrodes were stored in a vacuum oven at 120 °C until assembly to minimize moisture uptake.

2.3 Electrode Sizing and Tab Welding

Before assembly, electrodes were cut into rectangular sheets measuring approximately 4.5 × 5.5 cm^2, corresponding to an active area designed to yield capacitance values in the 25 F range. To ensure reliable external connections, aluminium tabs were welded to each electrode, providing a robust current pathway during cycling. For symmetric EDLCs, two identical CG:FLG electrodes were used, while asymmetric devices paired a CG:FLG cathode with a $Ti_3C_2T_x$ negative electrode. Electrode mass loadings were controlled in the range of 5–6 mg cm^{-2} for EDLCs, while MXene electrodes were tailored to balance charge with the positive electrode in HSC devices.

2.4 Pouch Cell Assembly

The electrode-separator stacks were assembled inside laminated aluminium pouch foils, which act as hermetic housings and provide protection against ambient oxygen and moisture. A cellulose-based separator was placed between the positive and negative electrodes to ensure ionic conduction while preventing electrical shorting (**Figure 1**). All assembly operations beyond electrode drying and cutting were carried out in an argon-filled glovebox (O_2 and H_2O < 0.1 ppm). Inside the glovebox, the pouch foils were heat-sealed along two sides, leaving one edge open for electrolyte injection.

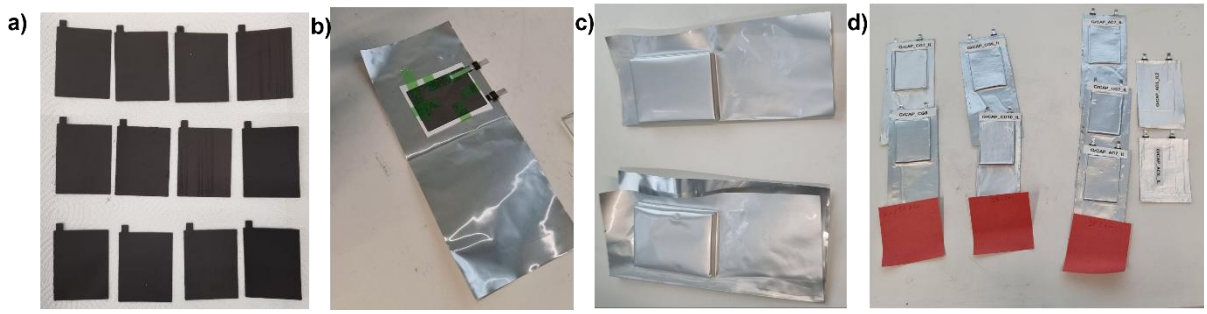

***Figure 1**. Pouch cell type design. a) Fabricated electrodes b, c) assembly of multilayer pouch cell before electrolyte injection d) final prototypes after electrolyte injection.*

Electrolytes were introduced through the unsealed side: 1 M N_{1113}FSI/ACN was used for symmetric EDLC devices, while 8 m $NaNO_3$ aqueous solution was selected for HSCs. The injected volume was adjusted according to electrode mass to guarantee complete impregnation of the porous electrode structures without excess liquid. After filling, the half-sealed pouches were allowed to rest in the glovebox for several hours to enable initial penetration of the electrolyte into the separator and electrodes. Vacuum sealing was then performed to eliminate trapped gases and to consolidate the pouch structure. Fully sealed devices were subsequently rested in the glovebox for 24 hours under open-circuit conditions. This wetting step was essential for full electrolyte saturation of the electrodes and separator, and ensured stable, reproducible electrochemical behavior during testing.

2.5 Electrochemical Testing Protocols

Electrochemical characterization of the pouch cells was carried out at 25 °C using a Bio-Logic VMP3 potentiostat/galvanostat. Cyclic voltammetry was performed at scan rates ranging from 5 to 1000 mV s^{-1}, with a set of ten pre-conditioning cycles at 50 mV s^{-1} before data collection. Galvanostatic charge-discharge measurements were carried out across a wide current range from 0.1 to 20 A g^{-1}. The voltage window was set to 3.0 V for symmetric IL-based EDLC devices and to 1.6 V for asymmetric HSC devices in 8 m $NaNO_3$.

The specific capacitance (C_g, F g^{-1}) of the SC electrodes was calculated from the GCD profiles using the equation :

$$C_g = 2 \times \frac{|i| \times t_d}{m \times \Delta V}$$

where m is the mass loading of the electrodes (excluding the current collector, mg), i is the charging/discharging current (mA), t_d is the discharge time of the GCD profile (s). The specific capacities (Cs, mAh g^{-1}) of the electrodes were calculated using the following equation:

$$C_s = 2 \times \frac{|\mathrm{i}| \times \mathrm{t_d}}{\mathrm{m} \times 3.6}$$

The specific energy (Es, Wh kg^{-1}) and specific power (Ps, W kg^{-1}) of the devices were calculated using the following equations:

Es (Wh kg^{-1}) = $\frac{\mathrm{i_d}}{2\times3.6} \int \mathrm{Vdt}$

Ps (W kg^{-1}) = $3600 \times \frac{Es}{\mathrm{t_d}}$

where ∫Vdt is the area under the galvanostatic discharge curve, i_d is the specific current (A g^{-1}), and Es is the discharge specific energy. Furthermore, considering the Es, Volumetric energy density (Ev, Wh L^{-1}) calculated accordingly;

Ev (Wh L^{-1}) = $\frac{Es}{\mathrm{d}}$

where d is the volume of the prepared cell. For asymmetric pouch cells, electrode mass ratios were pre-optimized in three-electrode configurations to ensure charge balance, with the full device window defined to avoid over-polarization of the MXene anode.

2.6 Data Processing and Analysis

All results were processed using the unified GREENCAP benchmarking protocol to ensure comparability across device formats and partners. Specific capacitances were normalized to the active material mass excluding current collectors. Energy and power densities were reported both gravimetrically and volumetrically, while Ragone plots were generated to illustrate the balance between energy and power. Long-term cycling tests were analyzed in terms of capacitance retention and coulombic efficiency over extended cycling, and pouch-level results were compared with data from coin and Swagelok cells to evaluate scale-up effects.

3 Results and Discussion

3.1 Symmetric EDLC Pouch Cells in 1 M N_{1113}FSI/ACN (CG:FLG || CG:FLG)

3.1.1 Single-layer pouches: Baseline and reproducibility

The development of symmetric pouch cells began with single-layer prototypes to establish baseline performance and assess reproducibility across batches and partner sites. These cells were constructed using CG combined with FLG, bound with an aqueous CMC/SBR system, and coated on carbon-coated aluminium foil. The electrodes, prepared at mass loadings between 2.5 and 5.5 mg cm^{-2}, were paired with cellulose-based separators and filled with 1 M N_{1113}FSI/ACN. As depicted in **Figure 2**, electrochemical characterization revealed that these single-layer pouch cells could deliver specific energies up to 29 Wh kg^{-1} at power densities around 372 W kg^{-1}. Importantly, devices fabricated from batches separated by almost one year and prepared at different partner laboratories produced overlapping Ragone plots, confirming excellent reproducibility of the electrode formulations, coating procedures, and assembly protocols. The triangular GCD profiles and quasi-rectangular CV curves underscored the dominance of electric double-layer behavior, with minimal hysteresis and low internal resistance. These results provided strong validation of the material and process choices, showing that sustainable water-based binders and graphene-based electrodes could yield pouch-scale devices with both high energy and reliable repeatability. Having demonstrated reproducibility and strong baseline performance in single-layer devices, the next logical step was to scale the architecture to multi-layer stacks in order to meet the target total capacitance of 25 F at the pouch level.

3.1.2 Multi-layer architecture: 27.4 F device at 3.0 V

After establishing reproducibility with single-layer prototypes, the work advanced to multi-layer pouch assemblies in order to reach the project's target capacitance of 25 F per device. Multi-layer stacks were constructed using five double-sided CG:FLG electrodes separated by cellulose membranes and assembled within laminated aluminium pouches. Electrochemical testing demonstrated that these devices achieved a total capacitance of 27.4 F, surpassing the DoA requirement. At the electrode level, capacitances reached 80.5 F g^{-1} at 0.5 A g^{-1}, while the cell-level capacitance was measured at 20.1 F g^{-1}. These multi-layer pouch cells delivered an energy density of 20 Wh kg^{-1} at a power density of 297 W kg^{-1}, corresponding to a volumetric energy density of 14.2 Wh L^{-1}. The galvanostatic charge–discharge curves remained nearly triangular across a current density range of 0.2 to 7 A g^{-1}, with only modest increases in IR drop at higher currents. The electrochemical performance of the fabricated pouch cell is summarized in **Table 1.**

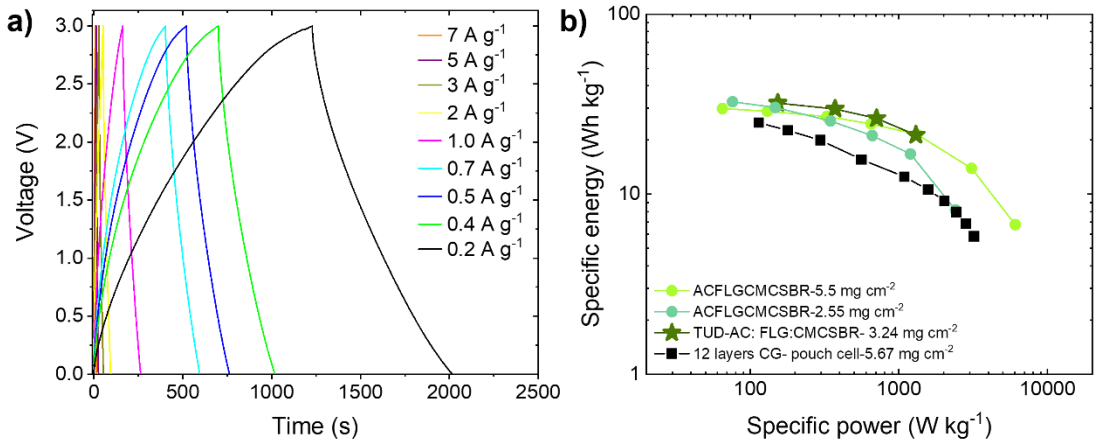

Figure 2. *(a Galvanostatic charge-discharge curves of a multi-layer pouch cell supercapacitor employing CG:FLG:CMC/SBR (94:2:4) electrodes tested in 1 M N_{1113}FSI/ACN electrolyte at various specific currents (0.2–7 A g^{-1}) up to 3 V. b) Ragone plot (energy vs. power density) comparing single-layer and multi-layer pouch cell supercapacitors with various electrode formulations and mass loadings.*

Table 1: *The electrochemical performance of the multilayer pouch cell prototype.*

Electrode mass loading (mg/cm^2)	$C_{electrode}$ (F/g)	C_{cell} (F/g)	Capacitance (F)	E_s (wh/kg)	Total energy (wh)	$E_{v,ALL}$ (wh/L)
5.67	80.53 @ 0.5 A/g	20.1	27.4	20 @297 W/kg	0.027	14.2

These results confirm that the electrode formulation, separator selection, and stacking approach are robust enough to maintain capacitive behavior in thicker pouch cell architectures while meeting the MS7 milestone KPI. While the total capacitance target was successfully exceeded, the scaling from single- to multi-layer also highlighted trade-offs in specific energy and power, which are discussed in the following subsection.

3.1.3 From single- to multi-layer: scaling rules, trade-offs, and KPIs

The comparison of single- and multi-layer pouch cells highlights important scaling rules and trade-offs. While single-layer devices achieved higher specific energies of nearly 29 Wh kg^{-1}, their total capacitance remained below the 25 F target. Multi-layer devices, by contrast, achieved the required capacitance of 27.4 F but with somewhat lower specific energy and power performance due to increased ionic resistance in the thicker stack and reduced electrolyte accessibility. These observations emphasize the need to carefully balance areal loading, separator porosity, and stack compression in order to scale from coin cells to pouch cells without sacrificing high-rate capability. Nevertheless, the multi-layer devices clearly demonstrate that the GREENCAP electrode technology can be engineered to satisfy DoA requirements at the pouch scale. The results also highlight the critical role of reproducibility across partners: the ability to achieve nearly identical Ragone characteristics from electrodes fabricated at different sites and times is a strong indication that the slurry formulation and coating processes are industrially transferrable.

3.2 Asymmetric Pouch Cells: CG:FLG // $Ti_3C_2T_x$ in 8 m $NaNO_3$

3.2.1 Rationale and three-electrode insights

Parallel to the symmetric IL-based EDLCs, asymmetric pouch cells were developed using CG:FLG as the positive electrode and $Ti_3C_2T_x$ MXene as the negative electrode. Extensive three-electrode studies had shown that while MXene achieves its highest capacitances in acidic electrolytes such as 3 M H_2SO_4, overall device performance is superior in concentrated neutral electrolytes due to their wider electrochemical stability window. In 8 m $NaNO_3$, the water activity is significantly reduced, suppressing parasitic reactions such as hydrogen evolution and allowing devices to operate up to 1.6 V without rapid degradation. These considerations guided the choice of 8 m $NaNO_3$ as the electrolyte for hybrid pouch cells, as it represents the best compromise between MXene capacitance, extended device voltage, and long-term cycling stability. Having established the electrolyte rationale, the next step was to compare performance of symmetric MXene pouches with hybrid HSCs to determine the most promising architecture for scale-up.

3.2.2 Symmetric MXene pouches (IL) and pivot to HSC

Early attempts to fabricate symmetric MXene pouch cells in ionic liquids revealed significant performance limitations. Although MXene is a highly pseudocapacitive material, its operation in IL-based symmetric configurations suffered from high interfacial resistance, non-ideal capacitance retention, and poor rate handling. These shortcomings prompted a strategic pivot to asymmetric configurations pairing MXene with CG:FLG in neutral WiSE electrolytes. By leveraging the strengths of each material–MXene for fast, reversible redox and intercalation processes, and CG:FLG for stable double-layer storage–the hybrid configuration overcame the limitations observed in purely MXene-based pouch devices. With this pivot, the project moved to detailed electrochemical characterization of CGFLG//MXene pouch cells in 8 m $NaNO_3$, ensuring careful charge balance to maximize performance.

3.2.3 Pouch-level electrochemistry (WiSE HSC, N/P = 1)

Hybrid pouch cells were assembled with balanced electrode masses to achieve charge neutrality (N/P = 1), ensuring that both electrodes could operate within their stable potential ranges (**Figure 3**). The cyclic voltammograms of these pouch cells were quasi-rectangular up to 500 mV s^{-1} across the 1.6 V window, with only slight distortions at the highest scan rates, reflecting the increasing contribution of resistive processes. The profiles of GCD cycles were nearly triangular across a current density range of 1 to 20 A g^{-1}, with relatively small IR drops, especially given the aqueous electrolyte and multi-layer assembly. The devices delivered a maximum specific energy of 5.9 Wh kg^{-1} at a power density of 1630 W kg^{-1}, while still retaining 4.5 Wh kg^{-1} at 15000 W kg^{-1}. Long-term cycling confirmed the durability of this configuration, with more than 80% of the initial capacitance retained after 16000 cycles at 5 A g^{-1} and CE consistently around 99%. These results validate the asymmetric approach at the pouch scale and confirm that the advantages of neutral WiSE electrolytes observed in three-electrode experiments (performed by BED) translate effectively to practical device architectures. Having confirmed the electrochemical performance, it was essential to reflect on design implications and sustainability considerations for the hybrid pouch approach.

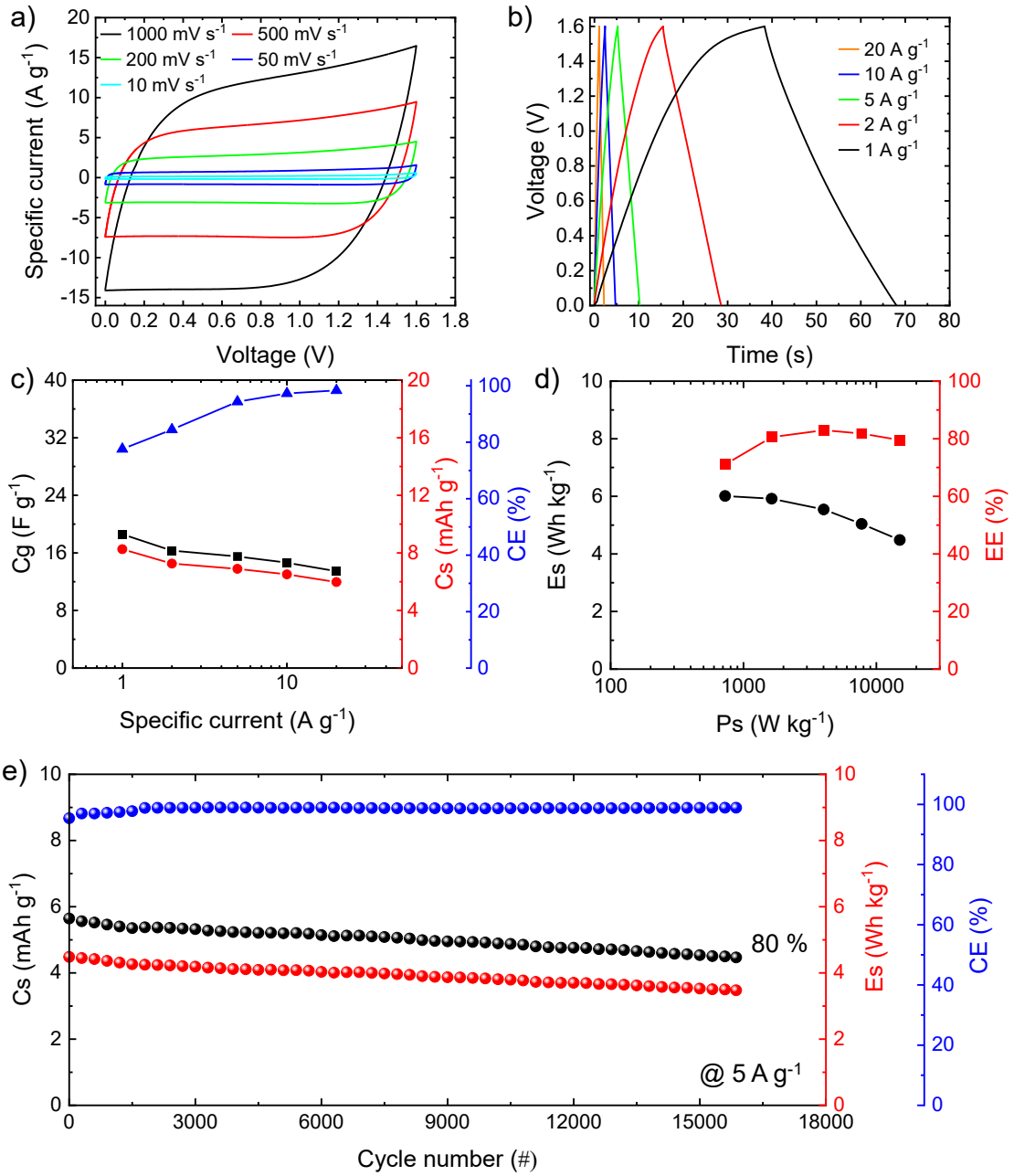

***Figure 3**. Electrochemical performance of the CG:FLG // MXene pouch cell in 8 m $NaNO_3$ (N/P =1). a) CV curves from 10 to 1000 mV s^{-1} within 1.6 V . b) GCD profiles at specific currents from 1 to 20 A g^{-1}, c) Rate performance plots showing Cg, Cs, and CE as a function of specific current, d) Ragone plot Es vs. Ps, e) long-term cycling at 5 A g^{-1}.*

3.2.4 Design implications and sustainability

The success of the CG:FLG//MXene hybrid pouch cells provides several important design lessons. First, careful charge balancing between the two electrodes is essential to avoid over-polarization, which would otherwise trigger parasitic reactions and compromise durability. Second, electrolyte selection plays a defining role: although MXene is more capacitive in acid, the wider working potential window achievable in concentrated neutral electrolytes leads to higher energy at the device level. Third, while the present prototypes relied on PVDF/NMP for MXene electrode preparation, efforts are ongoing to transition to water-based, PFAS-free binders and to explore ionogel-based systems for improved mechanical and electrochemical compatibility. These adaptations will ensure that future

pouch cells not only deliver high performance, but also align fully with the sustainability goals of GREENCAP and of the European Community. With both symmetric and asymmetric pouch prototypes validated, the results were then assessed against the project's key performance indicators and the remaining gap to 3.5 V operation.

3.3 Alignment with DoA Targets and Future Directions

The pouch cell results presented here demonstrate that the GREENCAP project has already fulfilled one of its critical objectives: achieving 25 F pouch cells with energy densities meeting or exceeding 20 Wh kg^{-1}. This was accomplished with symmetric CG:FLG devices operated at 3.0 V in IL electrolyte, surpassing the MS7 milestone KPI. Although the full 3.5 V operational window has not yet been achieved, the pathway is clearly defined through electrolyte optimization, separator development, and formation/float testing. The asymmetric pouch cells, while operating at a lower voltage window of 1.6 V, provided valuable proof of concept that hybrid devices can combine EDLC-type and pseudocapacitive electrodes at pouch scale with excellent cycling stability and high-power delivery. These results not only validate the materials choices and assembly methods but also provide practical design rules—particularly in terms of mass balancing and electrolyte selection—that will guide the next stage of development.

Together, the symmetric and asymmetric pouch cell demonstrations represent a significant step toward industrially relevant supercapacitor prototypes. The knowledge generated in Task 3.3 establishes both the baseline architecture and the scale-up practices required to transfer the GREENCAP technology into WP4 and beyond.

3.4 Contribution to project (linked) Objectives

BED: FLG production, CG:FLG slurry development (water-based), electrode coating; single- & multi-layer pouch inputs.

TUD: Pouch assembly/testing; metrics, scaling to multi-layer; D3.3 coordination.

TCD/UCAM: MXene powders and slurry know-how; three-electrode benchmarks for N/P setting.

SOLV: IL formulation (1 M $N_{1113}FSI$/ACN) and electrolyte guidance.

WP2/Task 2.4: Interface diagnostics to de-risk high-V operation (operando GC-MS, in-situ Raman) and inform additives/formation.

WP4 (**SKL, BED, BM, SOLV**): Uptake of stack recipe, mass-loading windows, and QA metrics for near-industrial formats and SMS development.

3.5 Contribution to major project exploitable result

Validated EDLC unit stack (CG:FLG + $N_{1113}FSI$/ACN) with 27.4 F @ 3.0 V and 20 Wh kg^{-1} / 14.2 Wh L^{-1}, a directly transferrable recipe for WP4 cylindrical prototypes and SMS modelling.

Validated HSC architecture (CG:FLG//$Ti_3C_2T_x$ + 8 m $NaNO_3$) demonstrating high-power pouch operation with strong cycling and well-defined N/P mass-balance rules for hybrid stacks.

4 Conclusion and Recommendation

EDLC IL-based pouches: Multi-layer CG:FLG achieves 27.4 F @ 3.0 V, 20 Wh kg^{-1}, 14.2 Wh L^{-1}, and robust rate response, 25 F target met with clear, near-term route to 3.5 V.

HSC WiSE pouches: CG:FLG//$Ti_3C_2T_x$ confirms that neutral WiSE can deliver pouch-level energy/power and lifetime if N/P = 1 and WPW are controlled.

Path to 3.5 V: IL co-anion tuning (FSI/TFSI), separator upgrades, rigorous dryness, stepwise formation + float with operando gas/spectroscopy to validate stability margins.

Sustainability: Transition MXene to PFAS-free, water-based binders and explore ionogels to align with project objectives while preserving kinetics.

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
WP3	Electrode with >200F/g and >150F/cm^3 performance in IL-based electrolytes	2	2	Optimization of electrolyte formulation (ratio IL/solvent); functionalization of 2D materials to improve the hybridization/intercalation of ILs with/in 2D materials to produce IGs

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

5.2 Interconnections with other deliverables

- With D3.1 and D3.2

- Electrode formulations and slurry developments (e.g., CG:FLG with CMC/SBR) form the direct basis of pouch cell assemblies in D3.3.
- Reproducibility across partner sites validates these earlier deliverables.

- With Task 2.4

- Diagnostic insights (gas analysis, spectroscopy) guide electrolyte and separator improvements.
- Essential for extending voltage window to 3.5 V.

- With WP4 deliverables

- Pouch cell recipes (mass loading, binder ratios, separator type, collector choice) directly inform cylindrical prototypes.
- Demonstrated reproducibility across BED and TUD confirms inter-site compatibility for scale-up.

6 Deviations from Annex 1

While the main objectives of Task 3.3 have been successfully addressed, several deviations from the original Annex 1 plan require explicit mention. The most significant deviation concerns the voltage target. Current pouch prototypes have been operated stably at 3.0 V in the case of symmetric EDLCs with ionic liquid electrolytes and at 1.6 V for asymmetric HSCs in WiSE. However, sustained operation at 3.5 V has not yet been achieved, primarily due to limitations in electrolyte oxidative stability and the onset of gassing at higher potentials. To address this, corrective actions have already been initiated, including electrolyte optimization through co-anion blending and eutectic formulations, reinforcement of separator materials to improve high-voltage tolerance, and the implementation of stepwise formation protocols informed by Task 2.4 diagnostics.

Another deviation was observed in the early trials of symmetric MXene-based pouch cells in ionic liquids. These devices underperformed as a result of interfacial resistance growth and kinetic constraints, which prevented them from reaching acceptable levels of performance. In response, the focus of the work was shifted toward the development of asymmetric CGFLG//MXene pouch cells in concentrated neutral electrolytes, which have successfully demonstrated the required energy, power, and cycling stability metrics.

A further deviation concerns binder sustainability in the MXene electrodes. For early proof-of-concept devices, MXene slurries were formulated with PVDF dissolved in NMP to facilitate electrode processing. While this allowed for rapid prototyping, it does not align with GREENCAP's sustainability goals. Ongoing activities in WP2 and WP3 are therefore dedicated to replacing PVDF/NMP with PFAS-free, water-based binders and, in parallel, to exploring ionogel formulations that offer improved electrochemical compatibility and environmental performance.

Finally, some multi-layer pouch assemblies exhibited slight discharge time variations compared to coin cell counterparts, which could be attributed to non-uniform stack pressure during lamination. This issue is being addressed through the development of optimized compression and lamination protocols, which ensure reproducibility and minimize heterogeneity in multi-layer pouch stacks.

Despite these deviations, the core capacitance target of 25 F has been fully achieved and, in fact, surpassed with 27.4 F in multi-layer EDLC pouches. Moreover, clear technical pathways toward stable 3.5 V operation and full industrial readiness have been defined, ensuring that GREENCAP remains firmly aligned with its project objectives and KPIs.

7 References

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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
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