

Deliverable D4.6

Delivery of Pouch Cells



Feasible Recovery of critical raw materials through a new circular Ecosystem for a Li-Ion Battery cross-value chain in Europe

WP4 - Scaling and validation of feasible technologies

D4.6 - Delivery of Pouch Cells

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List of Abbreviations

ACRONYMS	DESCRIPTION
Ah	Ampere-hour
C-Rate	Charge/Discharge Rate
CV	Cyclic Voltammetry
DEC	Diethyl Carbonate
DMC	Dimethyl Carbonate
EC	Ethylene Carbonate
EOL	End-of-Life
LFP	Lithium Iron Phosphate
LIB	Lithium-Ion Battery
LiPF ₆	Lithium Hexafluorophosphate
LMNO	Lithium Manganese Nickel Oxide
mAh/g	Milliampere-hour per gram
Ni	Nickel
NMC	Nickel Manganese Cobalt Oxide
OCV	Open Circuit Voltage
PEIS	Potential Electrochemical Impedance Spectroscopy
PVDF	Poly(vinylidene fluoride)
SEI	Solid Electrolyte Interphase
SOC	State of Charge
SOH	State of Health
Super P	Conductive Carbon Additive
XRD	X-Ray Diffraction

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1. Executive summary

This deliverable reports the production, optimization, and electrochemical validation of lithium-ion pouch cells fabricated using recycled cathode materials within the scope of the FREE4LiB project. The main objective was to demonstrate the feasibility of transferring recycled NMC622-based cathode materials from coin-cell validation to pouch-cell production and to assess their suitability for next-generation battery applications.

The work focused on FRNC-41, a recycled cathode-based formulation derived from recycled NMC622 material. Initial half-cell studies showed that the as-received recycled electrode required further optimization due to relatively high charge-transfer resistance and limited electronic conductivity. Therefore, several optimization strategies were applied, including slurry formulation adjustment, increased conductive carbon content, blending with commercial NMC622, and calendering. These steps significantly improved the electrode conductivity, reduced charge-transfer resistance, and enhanced electrochemical stability before pouch-cell scale-up.

Following the coin-cell optimization phase, pouch cells were fabricated using the optimized FRNC-41 formulation. The production process included vacuum mixing, doctor blade casting, drying, hot calendering, electrode stamping, semi-automatic stacking, housing, electrolyte filling, vacuum sealing, formation, aging, and electrochemical testing. Twelve pouch cells were produced to assess manufacturing reproducibility and reliability, as well as their suitability for module and battery pack integration. The cells were delivered to project partners for further validation in a real-life demonstrator, including integration into an electric bicycle system.

The optimized pouch cells demonstrated promising electrochemical performance. At C/10, the cells delivered an average specific discharge capacity of approximately 150 mAh g⁻¹, while rate capability tests showed stable behavior from C/10 to 2C. Long-term cycling at 1C over 250 cycles confirmed stable charge-discharge behavior, moderate polarization growth, and high coulombic efficiency. The maximum pouch cell capacity reached 4.09 Ah, corresponding to a nominal energy of 14.9 Wh and a gravimetric energy density of approximately 215 Wh kg⁻¹.

Overall, the results demonstrate that recycled NMC622-based cathode materials can be successfully optimized and integrated into pouch-cell configurations. This deliverable provides an important demonstration of material re-use in practical cell formats and supports the broader FREE4LiB objective of developing sustainable, circular, and scalable battery manufacturing routes based on recovered materials.

2. Introduction

The increasing demand for lithium-ion batteries in electric mobility, stationary energy storage, and portable electronic applications has intensified the need for sustainable battery manufacturing and recycling strategies. Conventional battery production relies heavily on critical raw materials such as lithium, nickel, cobalt, manganese, and graphite, many of which are associated with supply risks, environmental impact, and geopolitical dependency. In this context, the recovery and re-use of active materials from end-of-life batteries has become a key priority for the European battery value chain.

The FREE4LiB project addresses this challenge by developing and validating technologies for the recovery, reprocessing, and re-use of battery materials. Beyond recovering valuable elements, it is essential to demonstrate that recycled materials can be reintegrated into new battery cells with acceptable electrochemical performance, safety, and manufacturability. Therefore, the transition from material-level recovery to functional cell production represents a critical step toward validating circular battery concepts.

This deliverable focuses on the production and validation of pouch cells using recycled cathode materials, with particular attention to the FRNC-41 recycled NMC622-based cathode formulation. Before pouch-cell production, the recycled cathode material was evaluated and optimized at coin-cell level in order to reduce scale-up risks. Initial studies revealed that the recycled electrode required further improvement in electronic conductivity and charge-transfer resistance. For this reason, optimization strategies such as slurry formulation modification, conductive carbon adjustment, blending with commercial NMC622, and calendaring were applied.

After identifying the optimized electrode formulation and processing parameters, the work was transferred to pouch-cell production. Pouch cells represent a more application-relevant format than coin cells, as they require controlled electrode fabrication, stacking accuracy, electrolyte filling, gas management, sealing, formation, and aging. Demonstrating stable performance in this format is therefore an important milestone for assessing the practical potential of recycled battery materials.

The deliverable presents the full workflow from recycled cathode optimization to pouch-cell fabrication and electrochemical testing. The results provide evidence that recycled NMC622-based cathodes can be processed into pouch cells with promising capacity, rate capability, cycling stability, and energy density. This work contributes directly to the FREE4LiB objective of validating material re-use routes for next-generation lithium-ion batteries and supports the development of circular, resource-efficient battery technologies.

In addition to electrochemical validation, the produced pouch cells were intended for subsequent module and battery pack integration activities within the project. The manufactured cells were delivered to project partners to support demonstrator-level validation under realistic operating conditions, including integration into an electric bicycle system. This approach enables the project to assess not only material-level feasibility, but also the practical applicability of recycled-material-based lithium-ion batteries in real-life energy storage applications.

The FREE4LiB project plays a crucial role in addressing the challenges associated with LIB end-of-life management by evaluating the electrochemical performance of used battery cells, assessing the feasibility of second-life applications, and validating the use of recycled materials in new battery production. The results obtained will contribute to the optimization of recycling methodologies and the development of high-performance LIBs with sustainable material sourcing.

By demonstrating the viability of reusing spent batteries and incorporating recycled materials into newly assembled cells, this project supports the EU's strategic objectives in promoting circular economy practices and reducing reliance on critical raw materials. The findings will not only facilitate advancements in battery recycling technologies but also provide valuable insights for industry stakeholders involved in battery manufacturing, energy storage solutions, and policy development.

3. Recycled Cathodes (FRNC 41) Final Optimization Studies for Final Pouch Cell Production

To evaluate the electrochemical performance of the recycled NMC622 cathode material supplied by the TORRECID, cyclic charge-discharge (CCD) tests were conducted at the coin cell level using a half-cell configuration. The measurements were performed within a voltage window of 2.8-4.2 V at a C/10 rate (0.32 mA). These tests were designed to assess the specific capacity, cycling stability, coulombic efficiency, and polarization behavior of the recycled material. The obtained charge-discharge profiles provide insight into the electrochemical reversibility and internal resistance characteristics of the electrode under low-rate operating conditions before pouch cell production.



Table 3.1 FRNC-41 Electrode specifications (For the half-cell performances)

Parameter	Value	Notes
Electrode diameter	16 mm	Disc type
Electrode area (A)	2.01 cm ²	Calculated as πr^2
Active material mass	18 mg	= 0.018 g
Electrode thickness	100 μ m	= 0.01 cm
Areal loading	8.95 mg cm ⁻²	Based on active material
Active-based volumetric value	0.89 g cm ⁻³	(loading / thickness)
Areal Capacity (mAh/cm ²)	1.61 mAh cm ⁻²	= Design Capacity 180 mAh g ⁻¹

To evaluate the electrochemical performance of the recycled FRNC-41 sample, galvanostatic cycling results were analysed. The FRNC-41 electrode exhibited both a relatively low initial discharge capacity and a gradual capacity decay over prolonged cycling, which were attributed to increased internal resistance.

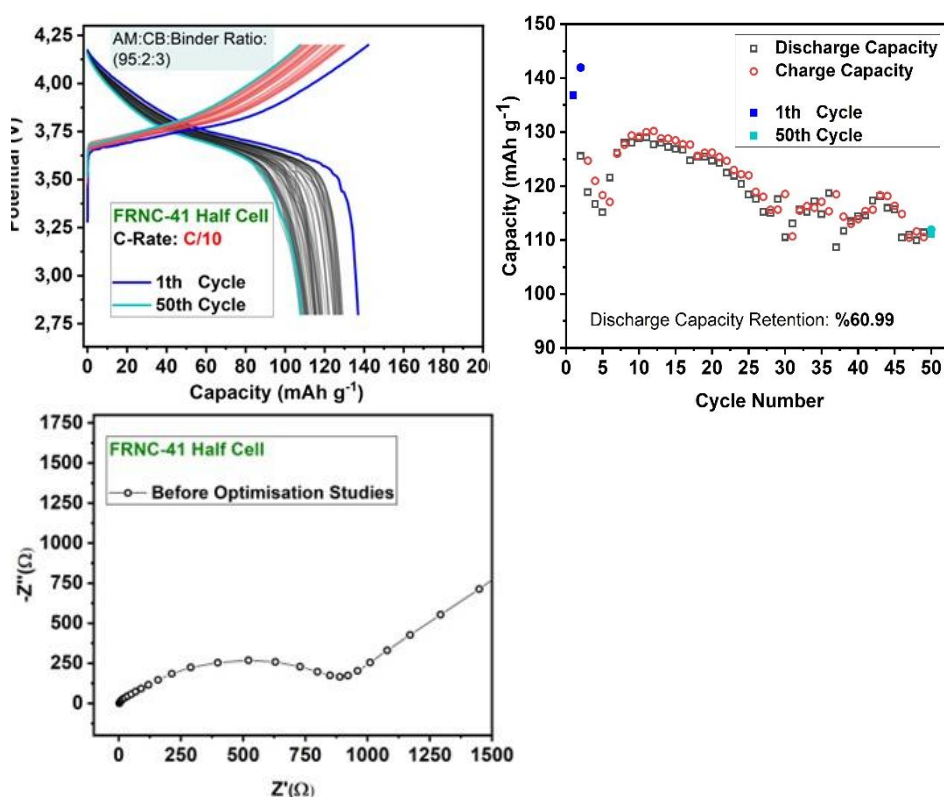


Figure 3.1 FRNC-41 half-cell electrochemical performances.

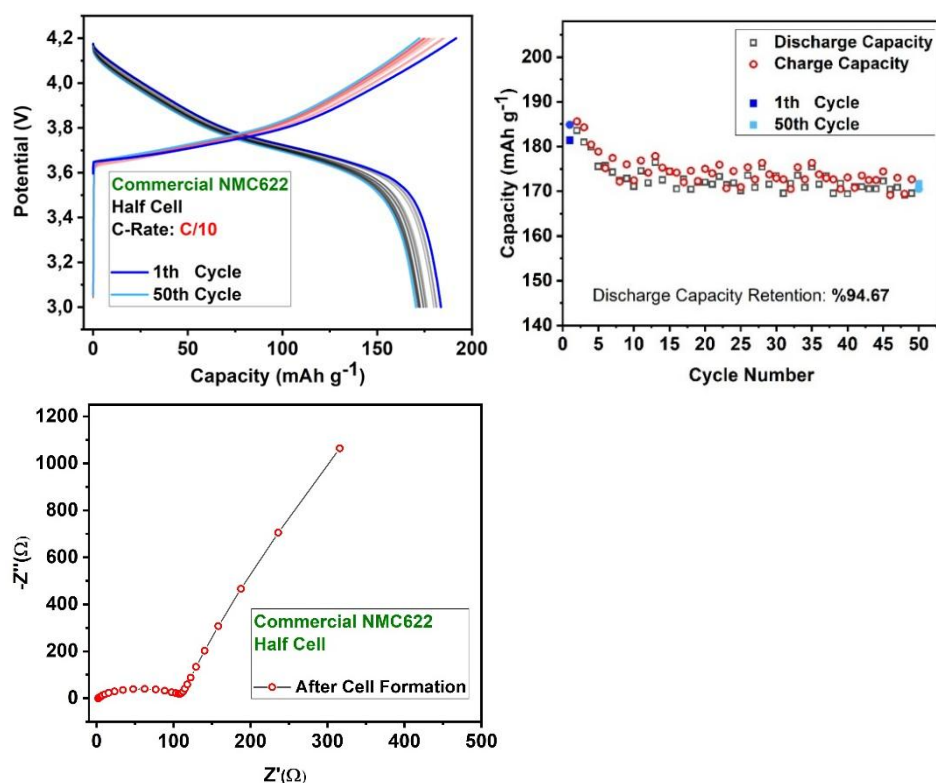


Figure 3.2 Commercial NMC622 half-cell electrochemical performances for comparison.

This interpretation was further supported by electrochemical impedance spectroscopy (EIS): compared to the commercial NMC622 electrode ($\approx 105 \Omega$), FRNC-41 showed a substantially higher charge-transfer resistance ($\approx 800 \Omega$). Therefore, the electronic transport properties of the electrodes were assessed using four-probe electrical conductivity measurements, and the results are summarized in Table 2. Overall, additional improvements were considered necessary to optimize the recycled material suitable for pouch cell manufacturing.

Table 3.2 Lucas Labs Pro4 - 4-point probe measurement (Keithley 2400 source meter)

	Applied Current	1 mA
	Substrate Area	2.5 mm x 2.5 mm
	Sample Thickness	90-100 μm
	Substrate	Si Wafer
	Temp.	25 $^{\circ}\text{C}$

Four-probe measurements revealed an electrical conductivity of 0.424 S/cm for the commercial NMC622 electrode, whereas FRNC-41 exhibited a much lower value of 0.01 S/cm. As a first improvement attempt, the electrode formulation was adjusted by increasing the conductive carbon content. Accordingly, the AM:CB:Binder ratio, previously optimized for maximizing energy density in the commercial NMC622 formulation (95:2:3), was revised to (92:4:4) for FRNC-41. Although a measurable improvement was observed, the conductivity still remained below the commonly accepted range reported in the literature and targeted by industry (0.1-1 S/cm) (Pantea et al., 2001; Yang et al., 2024)

Table 3.3 Four-probe electrical conductivity measurements

				Sample ID	Thickness (μm)	V/I (Ohm)	Areal Resistivity R_s , (Ohm/sq)	Resistivity ρ , (Ohm·m)	Intrinsic electrical conductivity σ , (S/m)	Intrinsic electrical conductivity σ , (S/cm)
Active M. Binder Carbon Ratios										
		COMMER.		NMC622 (95 3 2)	100	196	888,272	8,88E-02	1,13E+01	0,113
	OPT.	COMMER.		NMC622 (92 4 4)	100	52	235,664	2,36E-02	4,24E+01	0,424
Active M. Binder Carbon Ratios										
				FRNC-41 (95 3 2)	100	1345	6095,54	6,10E-01	1,64E+00	0,016
	OPT.			FRNC-41 (92 4 4)	100	921	4173,972	4,17E-01	2,40E+00	0,024
Commercial NMC622 Additive			(92 4 4)	FRNC-41 %10 Comm.	100	433	1962,356	1,96E-01	5,10E+00	0,051
			(92 4 4)	FRNC-41 %30 Comm.	100	214	969,848	9,70E-02	1,03E+01	0,103
	OPT.		(92 4 4)	FRNC-41 %50 Comm.	100	91	412,412	4,12E-02	2,42E+01	0,242
Calendering		FRNC-41 %50 Comm.	(92 4 4)	FRNC41 %20 Calend.	80	121	548,372	0,04386976	22,79474517	0,228
	OPT.	FRNC-41 %50 Comm	(92 4 4)	FRNC41 %30 Calend.	70	85	385,22	0,0269654	37,08456021	0,371
		FRNC-41 %50 Comm	(92 4 4)	FRNC41 %35 Calend.	65	134	607,288	0,03947372	25,33331036	0,253

While the root cause of the low conductivity in recycled materials should be elucidated via advanced characterization techniques, a rapid and practical mitigation strategy was also explored by blending FRNC-41 with commercial NMC622 at different ratios. A stepwise improvement in conductivity was

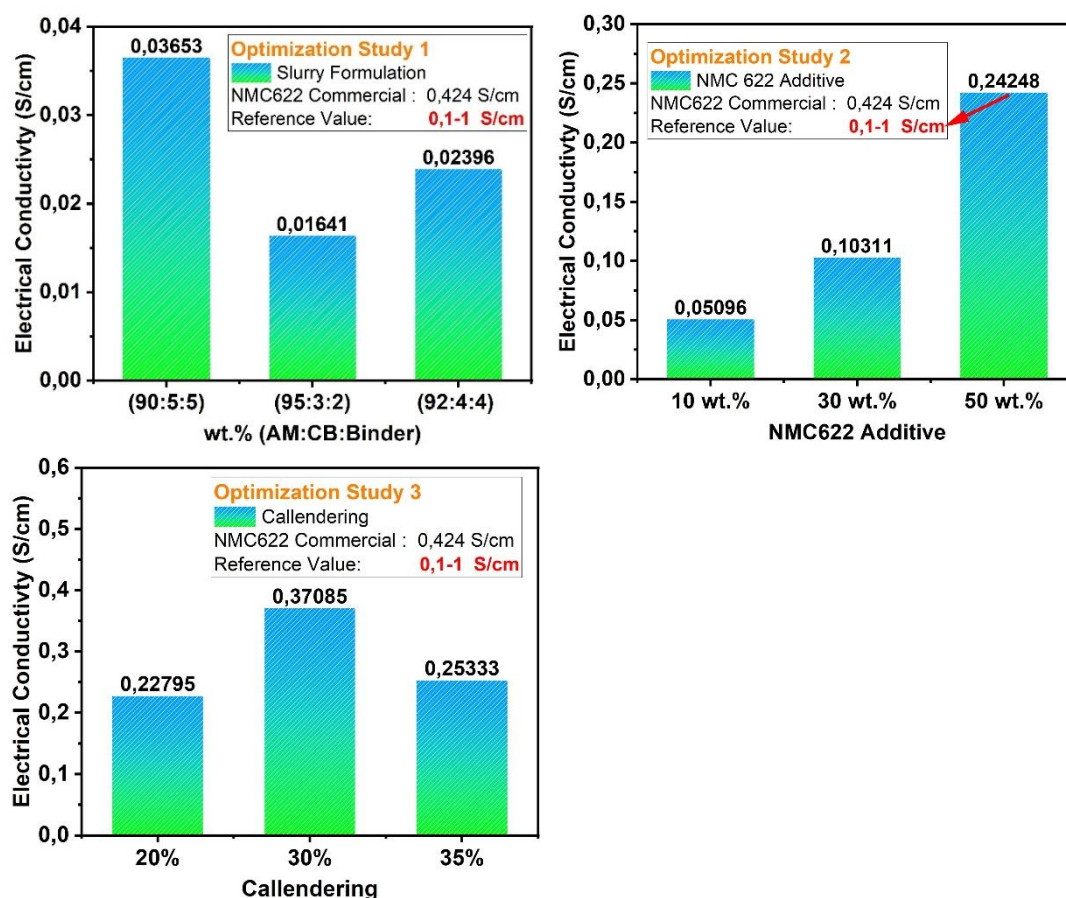


Figure 3.3 Electrical conductivity improvements a. After Slurry optimizations b. After NMC622 Blending c. Calendering.

observed with increasing commercial NMC622 content. The blending ratio was increased until the conductivity approached the literature/industry target window of 0.1–1 S/cm; in this regard, a 50 wt.% addition of commercial NMC622 enabled a conductivity of 0.242 S/cm (Figure 3.3.). In the next step, the electronic conductivity properties of the electrode were further optimized through calendering.

As a first optimization strategy, the electrode slurry composition was modified, and the FRNC-41 active material was blended with 50 wt.% commercial NMC622 (AM:CB:Binder = 92:4:4). The electrochemical performance of the optimized electrode (FRNC-41 v2) was evaluated at C/10 in half-cell configuration (Figure 3.4.).

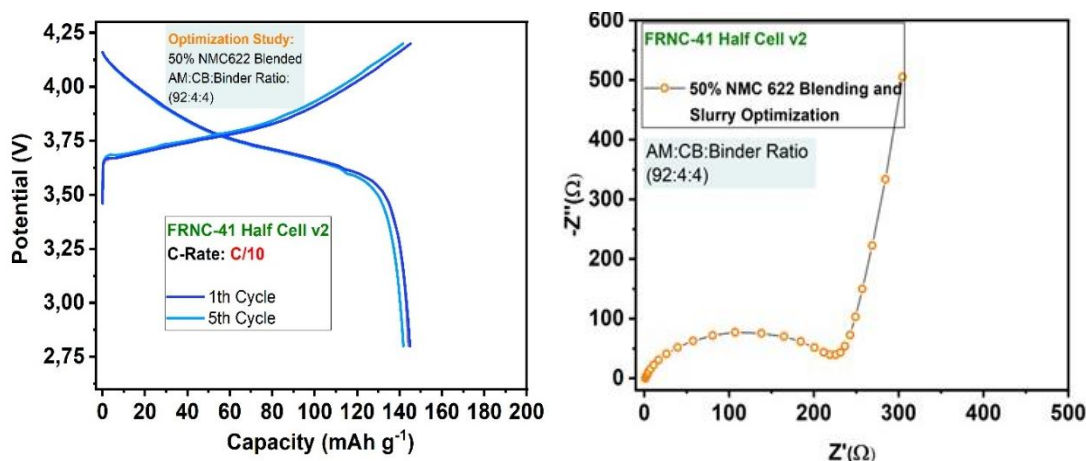


Figure 3.4 Half-cell electrochemical performances after slurry optimization and 50 wt.% NMC622 blending optimizations.

Following the optimization strategy, the charge-transfer resistance (R_{ct}) decreased significantly to approximately 220 Ω . In parallel, galvanostatic charge-discharge testing at C/10 delivered an improved discharge capacity of 143 mAh g⁻¹. Moreover, a substantially more stable and less polarized charge-discharge profile was obtained, indicating enhanced electronic transport and improved electrode integrity.

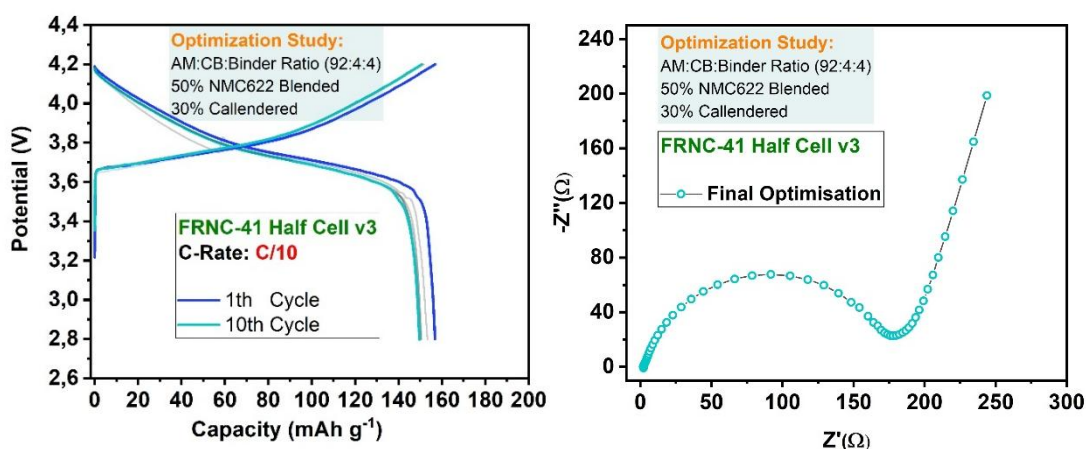


Figure 3.5 Final optimization electrochemical results before pouch cell production.

In the final optimization stage, the combined effects of slurry formulation adjustment, 50 wt.% commercial NMC622 blending (AM:CB:Binder = 92:4:4), and calendering parameter optimization were evaluated (FRNC-41 v3). The Nyquist plot demonstrates a further reduction in charge-transfer resistance compared to the previous optimization step, indicating improved interparticle contact and enhanced electronic percolation. The overall impedance response became significantly more compact, reflecting improved electrode kinetics (Figure 3.5.).

Consistent with the EIS results, galvanostatic charge-discharge measurements at C/10 exhibited improved capacity retention and a markedly more stable

voltage profile with reduced polarization. The optimized electrode delivered approximately 150 mAh g⁻¹ and showed substantially improved electrochemical stability compared to the as-received material (Figure 3.5.). The half-cell optimization phase provided a reliable electrochemical baseline, reducing scale-up risks and enabling a controlled transition to pouch cell production.

Table 3.4 Final Electrode specifications for the half-cell performances and pouch cells.

Parameter	Value	Notes
Electrode diameter	16 mm	Disc type
Electrode area (A)	2.01 cm ²	Calculated as πr^2
Active material mass	29 mg	= 0.029 g
Electrode thickness	90 μ m	= 0.009 cm
Areal loading	14.42 mg cm ⁻²	Based on active material
Active-based volumetric value	1.603 g cm ⁻³	(loading / thickness)
Areal Capacity (mAh/cm2)	2.163 mAh cm ⁻²	= Design Capacity 150 mAh g ⁻¹

In conclusion, the coin cell optimization studies for the FRNC-41 electrode were successfully completed, demonstrating substantial improvements in both electrical and electrochemical performance. Through slurry formulation optimization, conductive additive blending, and calendaring processes, significant enhancements in electrical conductivity, charge transfer resistance (R_{ct}), and overall electrochemical stability were achieved.

The optimized electrode exhibited a considerable increase in specific capacity from 139 to 155 mAh g⁻¹, while the charge transfer resistance was significantly reduced from approximately 800 Ω to 180 Ω . These improvements indicate enhanced electronic conductivity, improved ion transport kinetics, and more stable electrode/electrolyte interfaces within the optimized electrode structure.

Based on the promising electrochemical performance obtained from the coin cell studies, the optimized FRNC-41 electrode formulation was selected for pouch cell fabrication and scale-up investigations. The results demonstrate that the recycled NMC622-based electrode possesses sufficient electrochemical stability and performance potential for practical lithium-ion pouch cell applications.

4. Scale-Up Studies with The Pouch Cell Level

Coin cell optimization studies performed for the FRNC sample resulted in the determination of the optimum electrode formulation and processing parameters for pouch cell scale-up production. Based on the electrochemical performance obtained from coin cell testing, the selected electrode parameters were established as follows: AM:CB:Binder ratio of 92:4:4 (wt.%), a commercial NMC622 blending ratio of 50 wt.%, and a calendaring ratio of 30%. Following the optimization stage, large-scale electrode fabrication for pouch cell production was initiated using the optimized formulation and processing conditions.

Electrode slurries were prepared using a 500 mL vacuum mixer can under controlled vacuum conditions to obtain homogeneous slurries. Each batch enabled the processing of approximately 300 g of active material, providing sufficient capacity for laboratory-scale electrode production and pouch cell fabrication studies (Figure 4.1.). The vacuum-assisted mixing process played a critical role in minimizing trapped air bubbles within the slurry, which is particularly important for achieving uniform electrode coatings and preventing structural defects during printing and drying stages. In addition, vacuum mixing improved the dispersion of conductive additives and binder components throughout the electrode formulation, resulting in enhanced slurry homogeneity and more stable rheological behavior. The improved dispersion quality and controlled viscosity contributed to better coating consistency. Furthermore, homogeneous slurry preparation supported the formation of uniform conductive pathways within the electrodes, which is essential for obtaining reliable electrochemical performance in lithium-ion pouch cell applications.



Figure 4.1 High shear, planetary vacuum mixing process.

The prepared electrode slurry was casted onto the current collector foil using a doctor blade-based tape casting process (Figure 4.2.). During the casting stage, a casting speed of 10 mm s⁻¹ was applied to ensure controlled and uniform slurry deposition across the foil surface. The doctor blade gap was adjusted depending on the targeted film thickness and desired electrode loading. Following the casting process, the electrodes were dried at 100 °C to remove residual solvent from the slurry and promote the formation of a stable electrode structure. The controlled drying process contributed to improved adhesion between the active layer and the current collector while helping to obtain uniform electrode morphology suitable for subsequent pouch cell assembly steps.

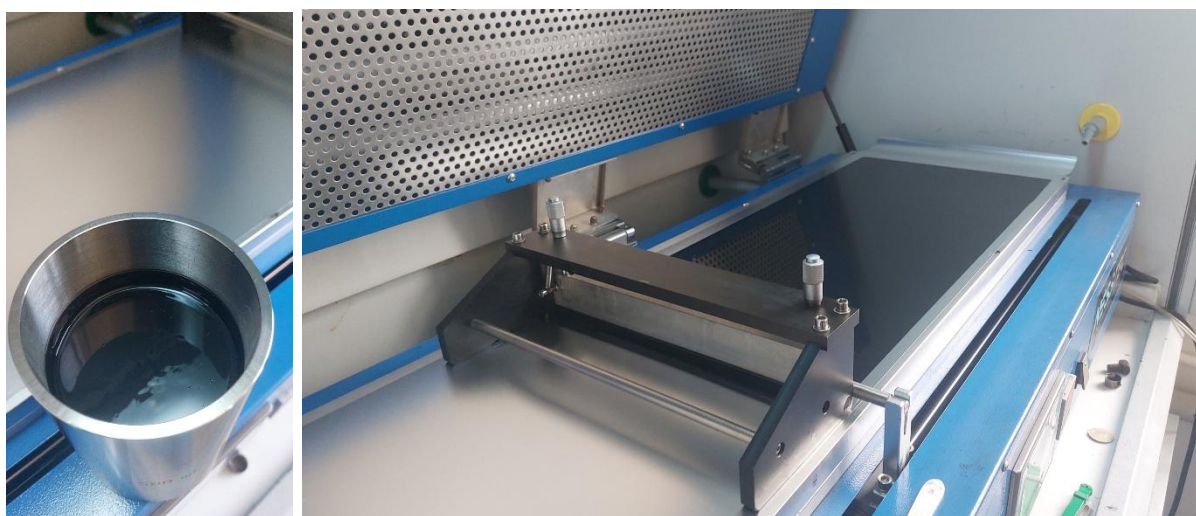


Figure 4.2 Double side tape casting process using a high-precision doctor blade applicator.

After the drying stage, the electrodes were cut into appropriate dimensions for the hot calendaring process (Figure 4.3.). The calendaring treatment was subsequently carried out using a heated calendaring/rolling system. This process was applied to improve electrode densification, enhance particle-to-particle contact within the active layer, and strengthen adhesion between the electrode tape and the current collector foil. In addition, the hot calendaring process contributed to reducing electrode porosity and improving the overall electrical conductivity of the electrode structure. The controlled compression of the electrode layer also helped achieve more uniform thickness distribution and enhanced mechanical stability prior to pouch cell assembly.

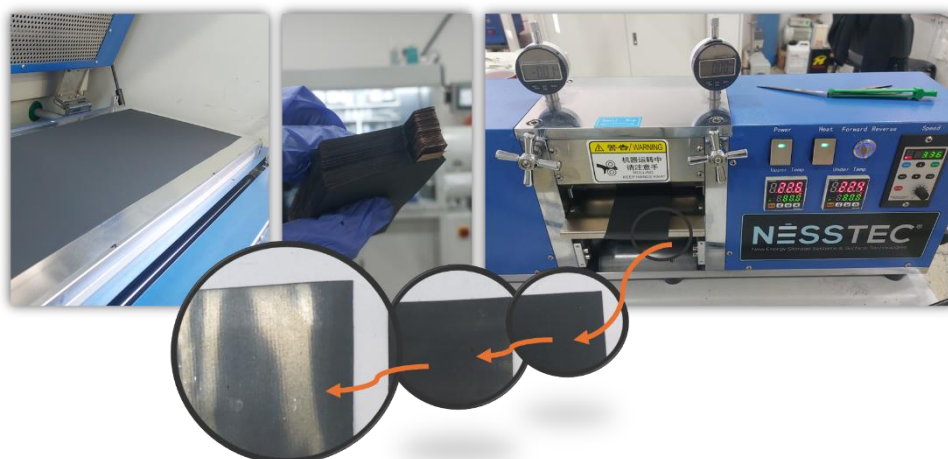


Figure 4.4 From Tape Casting to Hot Calendaring Process.

Following the calendaring process, the electrodes were stamped into the desired dimensions using a die-cutting system according to the targeted pouch cell geometry. Precise electrode cutting is a critical step for achieving consistent stacking alignment and ensuring dimensional compatibility during pouch cell assembly. The laser assisted die-cutter enabled the preparation of uniformly shaped electrodes with controlled dimensions suitable for the selected pouch cell design. In addition, accurate electrode sizing contributed to improved stack stability, optimized active area utilization, and more reliable cell assembly during subsequent manufacturing stages.

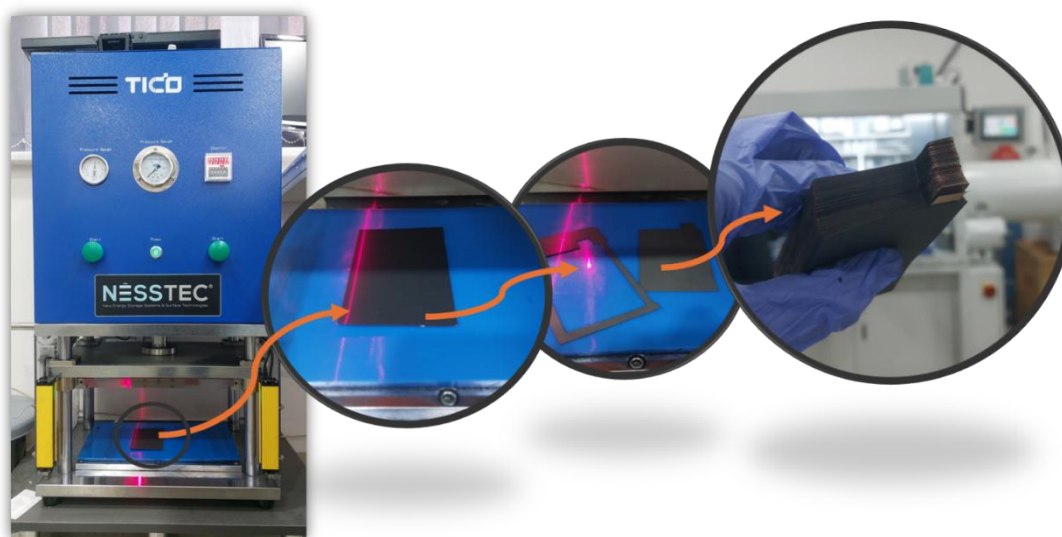


Figure 4.3 Electrode Stamping Process.

Following electrode preparation, the pouch cell architecture was assembled using a semi-automatic stacking machine (Figure 4.5). During this process, cathode and anode electrodes were sequentially stacked together with separator layers in a controlled manner according to the targeted cell configuration and desired stack number. The semi-automatic stacking process

enabled accurate electrode alignment and reproducible layer positioning throughout cell assembly. Proper separator integration between the anode and cathode layers played a critical role in preventing internal short circuits while maintaining ionic transport within the cell structure. In addition, controlled stacking improved the mechanical integrity and uniformity of the electrode assembly, contributing to stable electrochemical performance and reliable pouch cell fabrication. The resulting multilayer electrode architecture was subsequently prepared for electrolyte filling and pouch sealing processes.

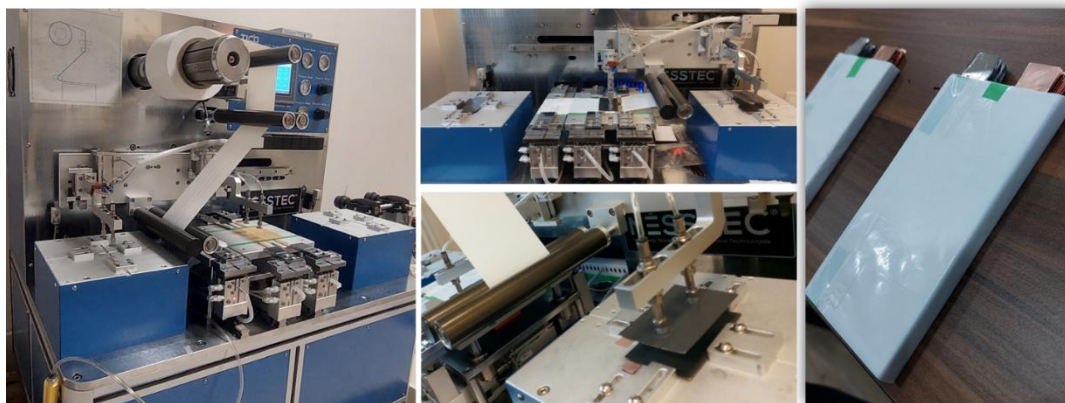


Figure 4.5 Semi-Automatic Cell Stacking Process.

Following the stacking process, the cell assembly was transferred to the Al-laminated film case formation stage (Figure 4.6). In this step, the aluminum laminated pouch film was shaped according to the targeted pouch cell dimensions to create the cell housing structure. The case formation process was carried out using a dedicated forming system to obtain a stable cavity geometry suitable for accommodating the stacked electrode assembly. The formed Al-laminated structure provides both mechanical protection and barrier properties against moisture and oxygen penetration, which are critical for maintaining lithium-ion cell stability and electrochemical performance. In addition, the lightweight and flexible nature of the laminated pouch structure supports compact cell design and improved gravimetric energy density compared to rigid cell housings. After the case formation process, the stacked electrode assembly was carefully placed into the prepared pouch cavity for subsequent electrolyte filling and vacuum sealing stages.



Figure 4.7 Case-formation Process

Following electrolyte filling, the pouch cells were subjected to a vacuum sealing process to minimize the presence of trapped air and moisture within the cell structure. Vacuum-assisted sealing is a critical step in pouch cell fabrication, as residual gases and environmental contaminants can negatively affect electrochemical stability and long-term cell performance (Figure 4.7.).

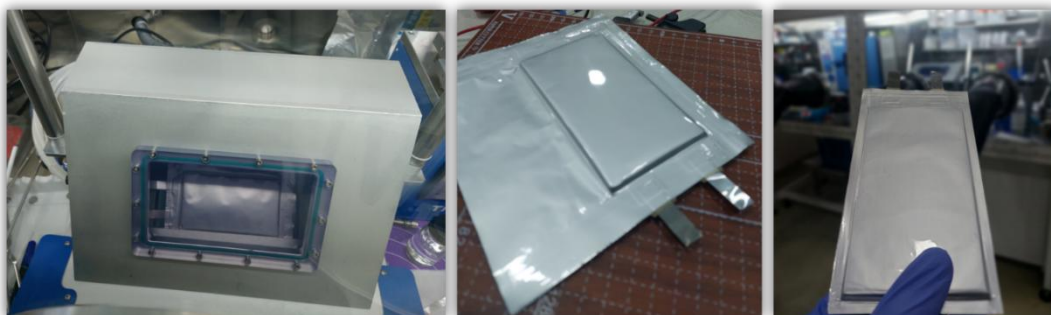


Figure 4.6 Cell Housing, Sealing and Vacuum Sealing Process.

During the initial sealing stage, a gas pocket was intentionally left inside the pouch cell design. This temporary gas chamber allows accommodation of gas generation that may occur during the first charge–discharge cycles, commonly referred to as the formation process. The controlled gas pocket design helps prevent excessive internal pressure buildup during initial electrochemical activation of the cell. After the formation cycles and degassing procedures, the remaining gas pocket can be removed through a final sealing step to obtain the completed pouch cell structure with improved stability and reliability.

Following the sealing stage, the pouch cells underwent formation and aging processes to stabilize the electrochemical structure and activate the electrode interfaces under controlled conditions. The formation cycles were carried out using low current charge–discharge protocols to promote stable solid electrolyte interphase (SEI) formation and improve overall cell stability. After the aging procedure, the electrochemical performance of the cells was further

evaluated through charge–discharge testing at different C-rates (Figure 4.8.). These tests were conducted to verify the maximum achievable performance, rate capability, and cycling stability of the fabricated pouch cells under various operating conditions.



Figure 4.8 Aging, formation and electrochemical tests of produced pouch-cells.

5. Electrochemical Characterization of Produced Pouch Cells

Figure 5.1 presents the galvanostatic charge-discharge profiles of the optimized FRNC-41 pouch cell measured at a C/10 current rate for the 1st and 10th cycles. The pouch cells were fabricated using recycled NMC622 cathode material, and twelve identical pouch cells were produced to evaluate the reproducibility of the cell fabrication process.

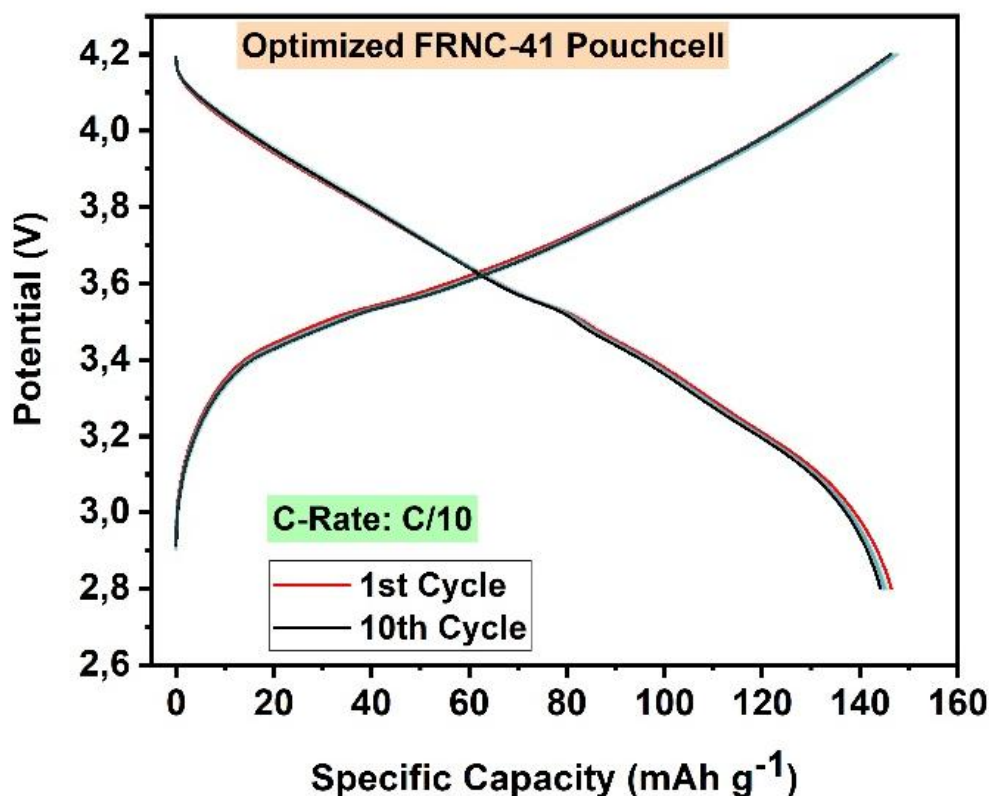


Figure 5.1 Optimized FRNC-41 pouch cell charge-discharge test at C/10.

The average specific discharge capacity obtained from the cells was approximately 150 mAh g^{-1} at C/10, indicating that the recycled cathode material maintained a high reversible lithium storage capability despite the recycling process. The charge discharge curves exhibit the characteristic voltage plateau behavior of layered NMC-based cathodes, centered around 3.6-3.8 V, which is associated with reversible lithium intercalation/deintercalation reactions within the NMC622 structure. The relatively smooth and symmetric profiles between charge and discharge processes suggest stable electrochemical kinetics and good reversibility of the electrode reactions. In addition, the overlap between the 1st and 10th cycle profiles demonstrates that the fabricated pouch cell retained its electrochemical behavior during early

cycling. Only minor polarization differences were observed between the cycles, indicating limited impedance growth and stable electrode/electrolyte interfaces. The absence of significant voltage hysteresis increase further confirms the structural stability of the recycled cathode material and the effectiveness of the pouch cell assembly process. The discharge capacity approaching 155 mAh g^{-1} is considered highly promising for recycled NMC622-based pouch cells and suggests that the recovery and reprocessing steps preserved the electrochemical activity of the cathode material to a large extent. Overall, the results demonstrate that the optimized FRNC-41 pouch cell configuration provides stable cycling performance and competitive energy storage characteristics suitable for further scale-up studies.

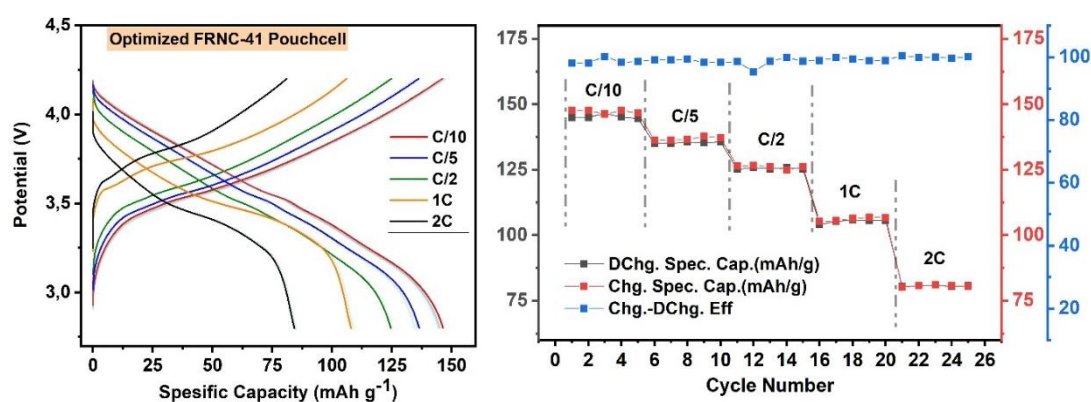


Figure 5.2 Optimized FRNC-41 pouch cell C-Rate performance.

Figure 5.2 presents the rate capability performance of the optimized FRNC-41 pouch cell evaluated at different current rates ranging from C/10 to 2C. The galvanostatic charge-discharge profiles demonstrate the expected influence of increasing current density on the electrochemical behavior of the recycled NMC622-based pouch cell. At lower current rates, particularly at C/10 the pouch cell delivered high specific discharge capacities approaching 150 mAh g^{-1} . The voltage profiles exhibited relatively long and stable discharge plateaus, indicating efficient lithium-ion diffusion and low polarization within the electrode structure. The small voltage hysteresis between charge and discharge curves at low C-rates further confirms favorable electrochemical reversibility and stable electrode kinetics. As the current rate increased from C/2 to 2C, a gradual reduction in specific capacity was observed. This behavior is primarily associated with increased polarization effects and kinetic limitations during rapid lithium insertion/extraction processes. At high discharge rates, especially at 2C, the voltage gap between charge and discharge curves became more pronounced, indicating higher internal resistance and reduced lithium diffusion efficiency under fast cycling conditions. Nevertheless, the pouch cell still maintained a considerable reversible capacity even at elevated current densities, demonstrating the structural integrity and electrochemical robustness of the recycled cathode material. The corresponding rate capability

plot further supports these observations. The average discharge specific capacities were approximately 145 mAh g^{-1} at C/10, 135 mAh g^{-1} at C/5, 125 mAh g^{-1} at C/2, 105 mAh g^{-1} at 1C, and around 80 mAh g^{-1} at 2C. Despite the progressive decrease in capacity with increasing C-rate, the cell exhibited stable cycling behavior within each current regime. In addition, the coulombic efficiency remained close to 100% throughout the rate capability test, indicating highly reversible charge-discharge reactions and minimal parasitic side reactions during cycling. The stable coulombic efficiency also confirms the effectiveness of the electrode formulation, electrolyte compatibility, and pouch cell assembly process. Overall, the rate performance results demonstrate that the optimized FRNC-41 pouch cell based on recycled NMC622 material possesses good electrochemical stability and satisfactory high-rate capability, making it a promising candidate for practical lithium-ion battery applications.

Figure 5.3 illustrates the long-term cycling performance of the optimized FRNC-

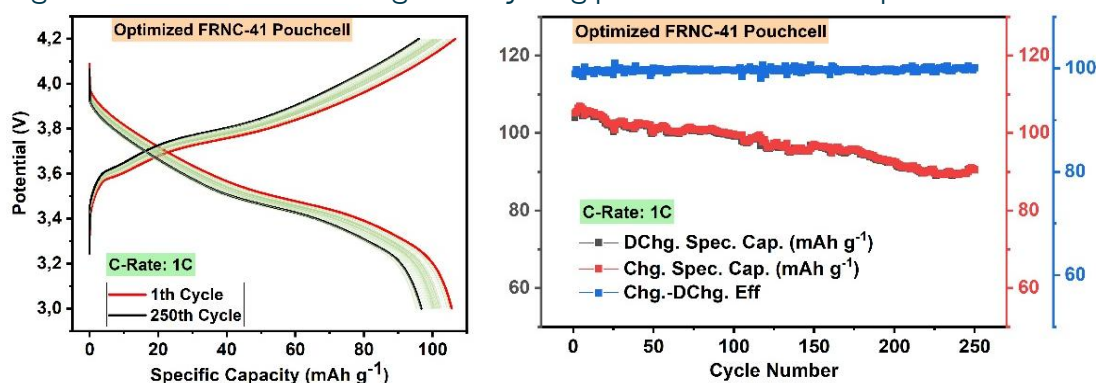


Figure 5.3 Optimized FRNC-41 charge discharge performance at 1C.

41 pouch cell evaluated at a 1C current rate over 250 cycles. The galvanostatic charge–discharge profiles recorded during the 1st and 250th cycles reveal that the pouch cell maintained its characteristic electrochemical behavior even after prolonged cycling.

The initial discharge capacity at 1C was approximately 105 mAh g^{-1} , while the discharge capacity after 250 cycles remained close to 90 mAh g^{-1} . This corresponds to an approximate capacity retention of:

$$\frac{90}{105} \times 100 = 86 \%$$

indicating good long-term cycling stability for the recycled NMC622-based pouch cell.

The voltage profiles of the 1st and 250th cycles showed only moderate changes in polarization during cycling. Although a slight increase in voltage hysteresis was observed after extended cycling, the general shape of the charge-discharge curves remained highly consistent. This behavior suggests that the layered

structure of the recycled NMC622 cathode preserved its electrochemical integrity throughout repeated lithium insertion/extraction processes. The cycling stability graph further demonstrates a gradual and stable capacity decay during prolonged operation at 1C. No sudden capacity drop or severe instability was detected during the 250-cycle test, confirming the mechanical and electrochemical stability of both the electrode formulation and the pouch cell assembly.

In addition, the coulombic efficiency remained close to 99-100% throughout the cycling test, indicating highly reversible electrochemical reactions and minimal parasitic side reactions. The stable coulombic efficiency also confirms the effectiveness of the electrolyte system and the compatibility of the recycled cathode material with the overall cell chemistry.

Overall, the long-cycle performance results demonstrate that the optimized

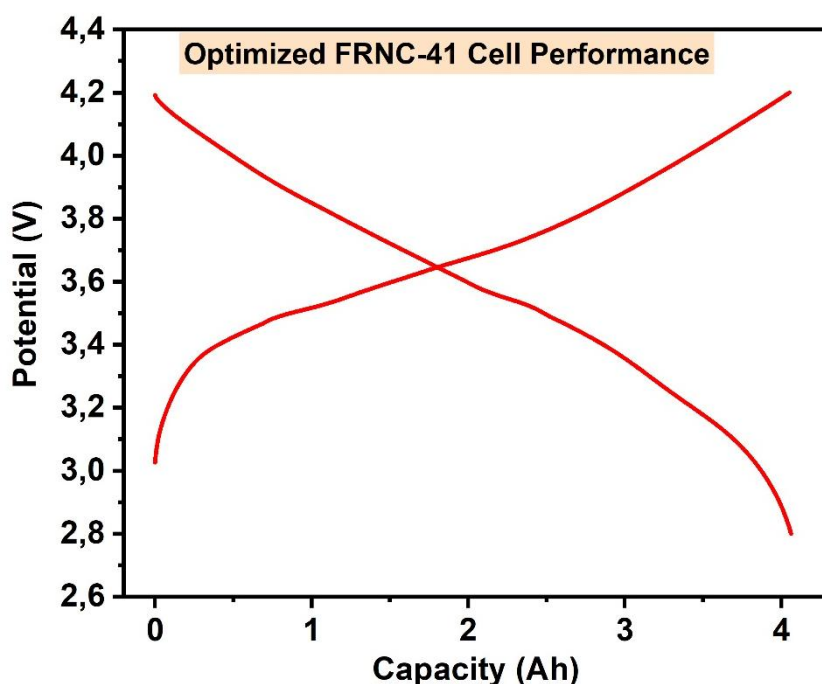


Figure 5.4 Maximum pouch cell capacity obtained with FRNC-41

FRNC-41 pouch cell exhibits stable electrochemical behavior and satisfactory capacity retention under relatively demanding 1C cycling conditions. These findings indicate that recycled NMC622 materials can provide reliable long-term performance in practical pouch cell applications.

According to the electrochemical results;

The specific capacity of the cell is **161 mAh g⁻¹**.

The cell capacity is measured as **4.09 Ah**.

The nominal operating voltage is **3.65 V**,

Therefore, the nominal cell energy calculated as: 4.09 Ah x 3.65 V = **14.9 Wh**

The total pouch cell mass was measured as **69.4 g**.

Finally, the gravimetric energy density of the fabricated pouch cell was calculated according to:

$$E_{grav} = \frac{V_{nom} \times Q_{cell}}{m_{cell}}$$
$$E_{grav} = \frac{14.9 \text{ Wh}}{0.0694 \text{ kg}} \approx 215 \text{ Wh kg}^{-1}$$

Thus, the fabricated pouch cell achieved a gravimetric energy density of: **215 Wh kg⁻¹**.

6. Conclusions

This deliverable demonstrates the successful optimization and pouch-cell integration of recycled NMC622-based cathode material within the FREE4LiB project. The initial evaluation of the FRNC-41 recycled cathode showed that further electrode optimization was required due to limited electronic conductivity and relatively high charge-transfer resistance. Through slurry formulation adjustment, 50 wt.% blending with commercial NMC622, and calendering optimization, the electrochemical and transport properties of the electrode were significantly improved.

The optimized formulation was successfully transferred from coin-cell validation to pouch-cell production. The fabrication workflow included vacuum-assisted slurry mixing, doctor blade casting, drying, hot calendering, electrode stamping, semi-automatic stacking, pouch forming, electrolyte filling, vacuum sealing, formation, and aging. This process enabled the production of reproducible pouch cells and confirmed the feasibility of scaling recycled cathode materials into practical cell formats.

Electrochemical testing showed that the optimized FRNC-41 pouch cells delivered approximately 150 mAh g^{-1} at C/10 and maintained stable performance across different C-rates up to 2C. Long-term cycling at 1C over 250 cycles showed gradual capacity fade without sudden failure, while coulombic efficiency remained close to 99–100%, indicating good reversibility and stable electrode/electrolyte interfaces. The maximum achieved pouch cell capacity was 4.09 Ah, corresponding to 14.9 Wh nominal energy and approximately 215 Wh kg^{-1} gravimetric energy density.

Overall, the results confirm that recycled NMC622-based cathode materials can be effectively reprocessed, optimized, and integrated into pouch-cell architectures. The work provides an important demonstration of the technical feasibility of recycled-material-based lithium-ion cells and supports the wider FREE4LiB objective of closing the material loop in the European battery value chain. Beyond laboratory-scale electrochemical validation, the produced pouch cells were also transferred to project partners for module and battery pack assembly activities. This represents an important step toward demonstrating the applicability of recycled-material-based lithium-ion cells in practical energy storage systems. The integration of the fabricated pouch cells into a demonstrator-level electric bicycle platform further strengthens the technological relevance of the developed recycling and cell manufacturing approach and supports the transition from material recovery to real-life application validation within the FREE4LiB project.