Demonstration of Si-C performance

20x 2Ah Multi-layer Pouch cells

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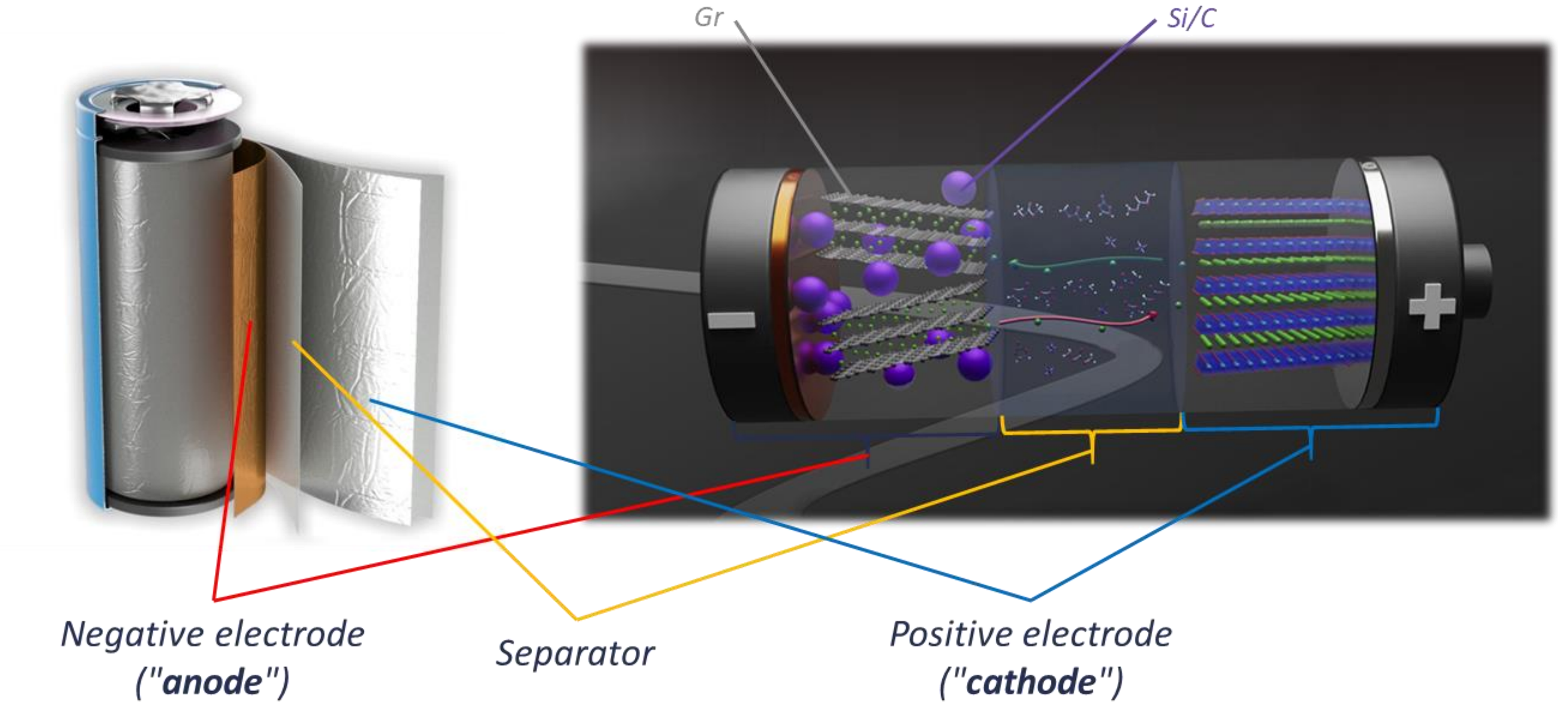
CELL DESIGN

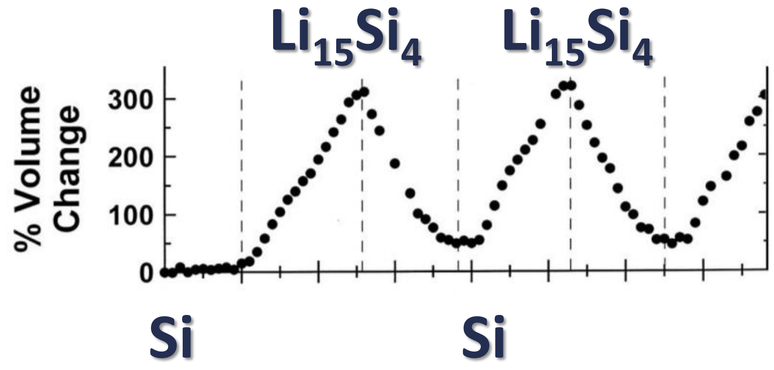


From LiC_6 to $Li_{15}Si_4$
→ +20% in Energy density
(Wh/kg = Ah x V / kg)

3579 mAh/g ~ 2194 Ah/L for **$Li_{15}Si_4$**

VS

372 mAh/g ~ 719 Ah/L for **LiC_6**



Beaulieu *et al.*, *Electrochem. Solid-State Lett.* (2001).

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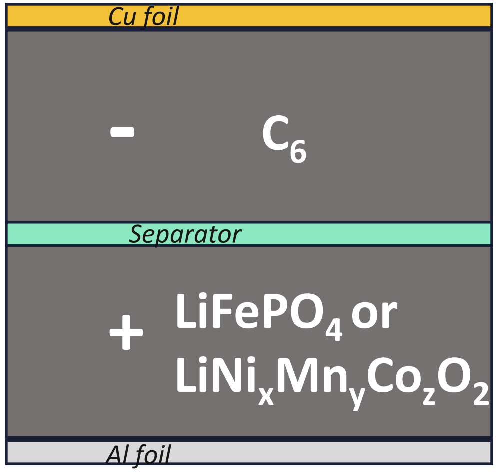
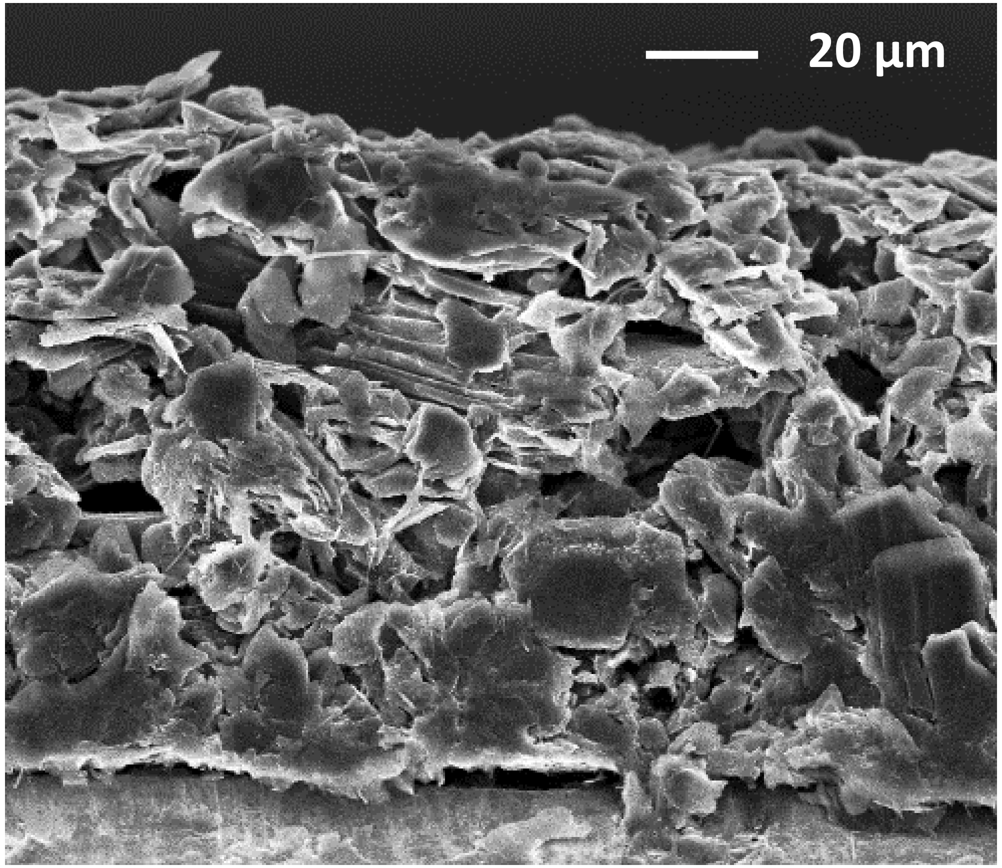


Conventional cell design in commercial Li-ion battery



An electrode has typically:

- 50 – 100 µm active layer thickness + 6-20 µm current collector = 100 – 200 µm in double-side
- Active material (> 95%) + binder (adhesion, flexibility, contribute to deposition process,…) + conductive additive (carbon black or carbon nanotubes)
- < 30% porosity to maximize active material content

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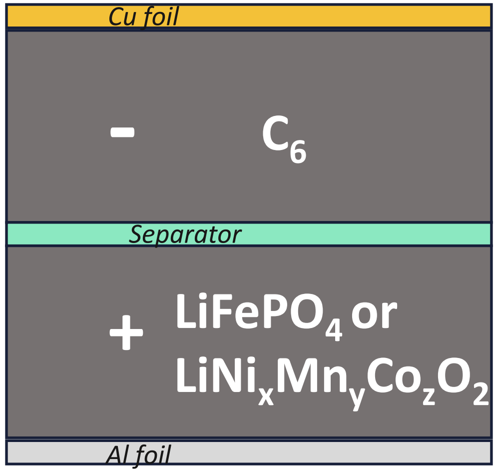

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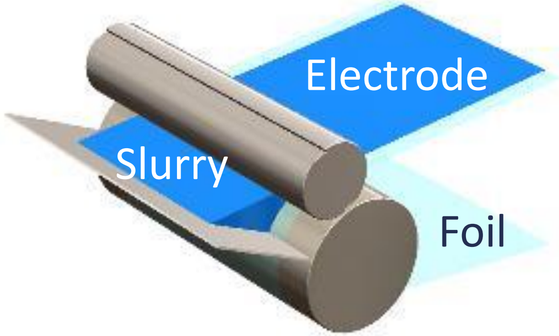

An electrode is typically:

- 50 – 100 µm active layer thickness + 6-20 µm current collector = 100 – 200 µm in double-side
- Active material (> 95%) + binder (adhesion, flexibility, process properties, dispersion…) + conductive additive (carbon black or carbon nanotubes)
- < 30% porosity to maximize active material content
- Obtained from a slurry (solvent (H_2O) + powders dispersed by mixing), deposited by coating process with high precision (< 0.1 mg/cm²) and solvent drying in-line

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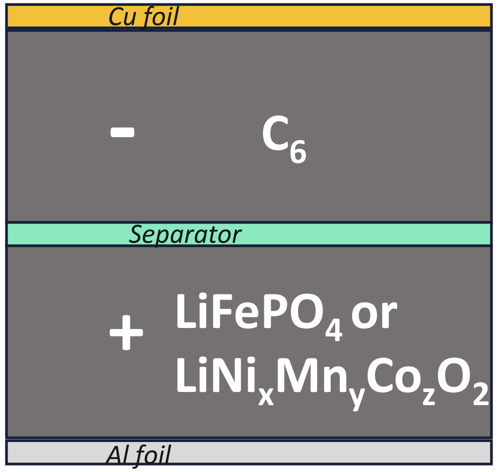

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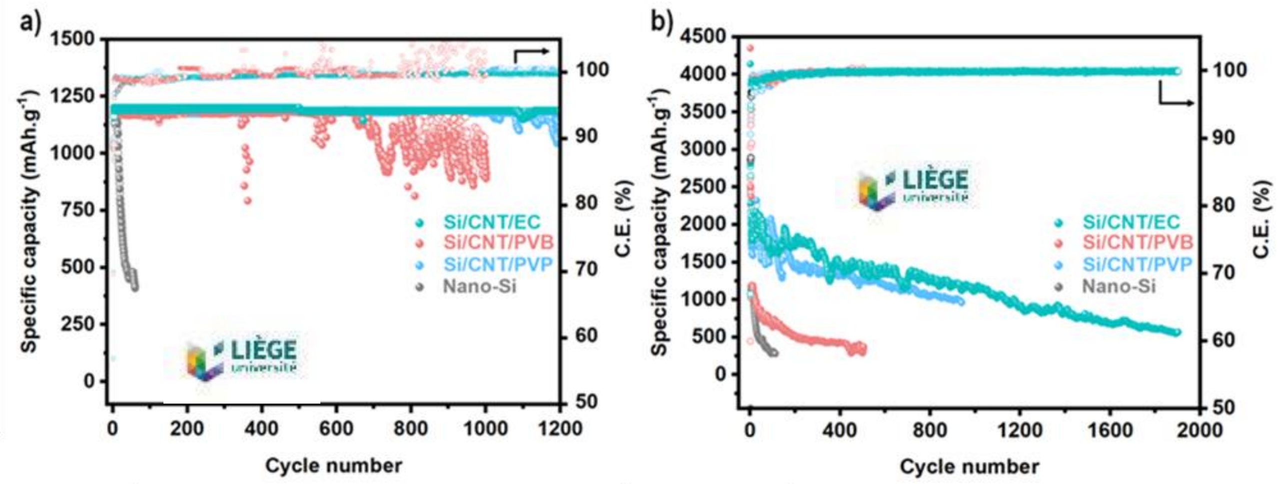
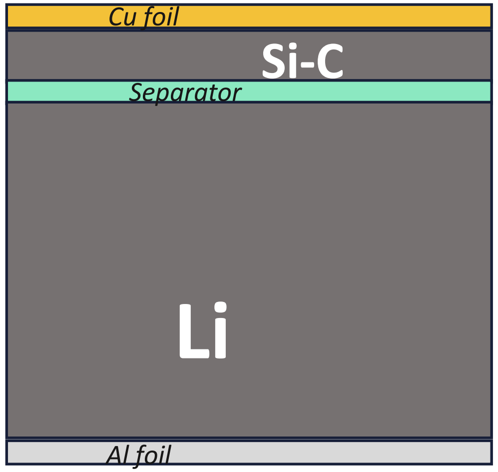


Impressive results of Liege university in capacity retention with > 1000 cycles of stable capacity



Lab cell design

- 0.5 mAh/cm² vs > 3 mAh/cm² in LiB

→ 0.3 mg/cm² of active material vs at least 3 mg/cm² in LiB

- 33% of SiC in the electrode vs > 95% for conventional electrode
- No pressure applied on the electrode vs < 30% porosity

- Infinite Li content vs limited Li content in LiB

→ Challenges for upscaling

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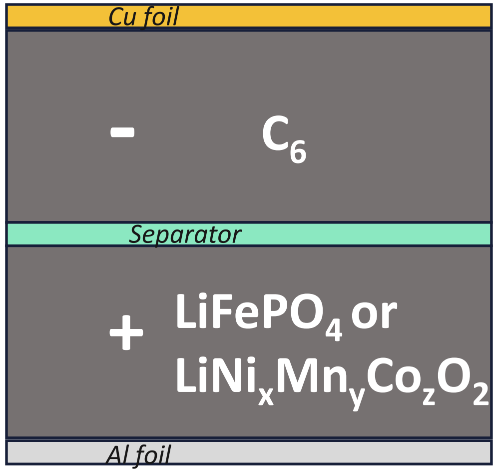
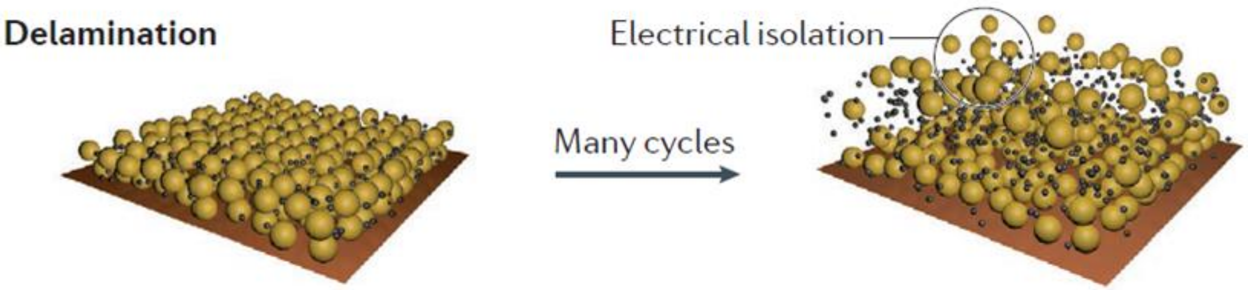
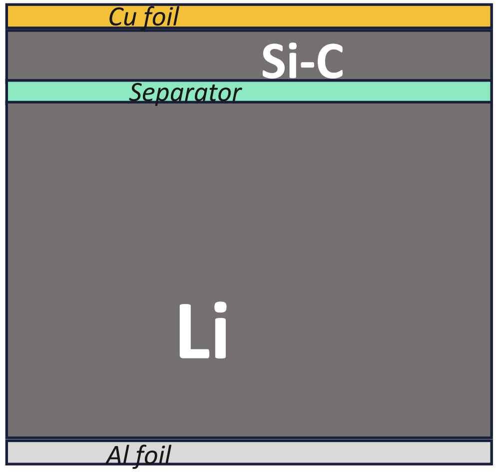


Conventional cell design in commercial Li-ion battery



→ **Si based anodes need also to manage swelling (specific binder / higher porosity)**
Much more challenging when thickness increases



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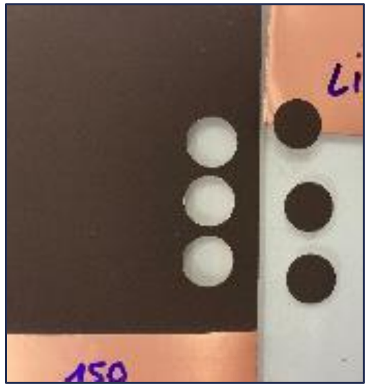
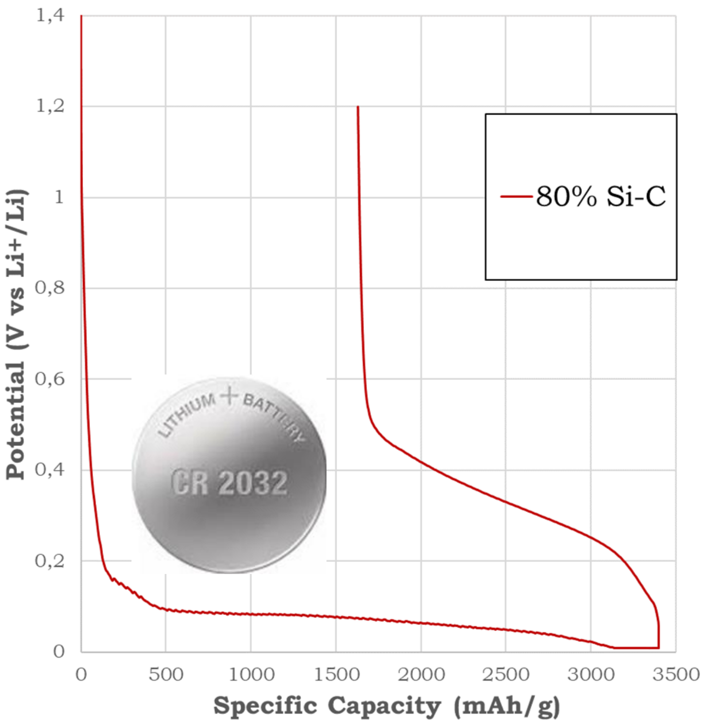
Electrode DESIGN: 1st evaluation

GREEnSiBat–Si (wt. %)	C65 (wt. %)	Na-CMC (wt. %)	Latex (wt. %)	Loading (mg/cm²)	Porosity (%)
33%	33%	33%	0%	0.9	>50%
80%	5%	12%	3%	3.3	35%

Incorporation and dispersion easy to do (D_{100} < 20 µm)



Handle to be assembled in half cell (CR2032)



3400 mAh/g in 1st lithiation but only 1750 in 1st delithiation
→ Initial coulombic efficiency (ICE) of 52.6%

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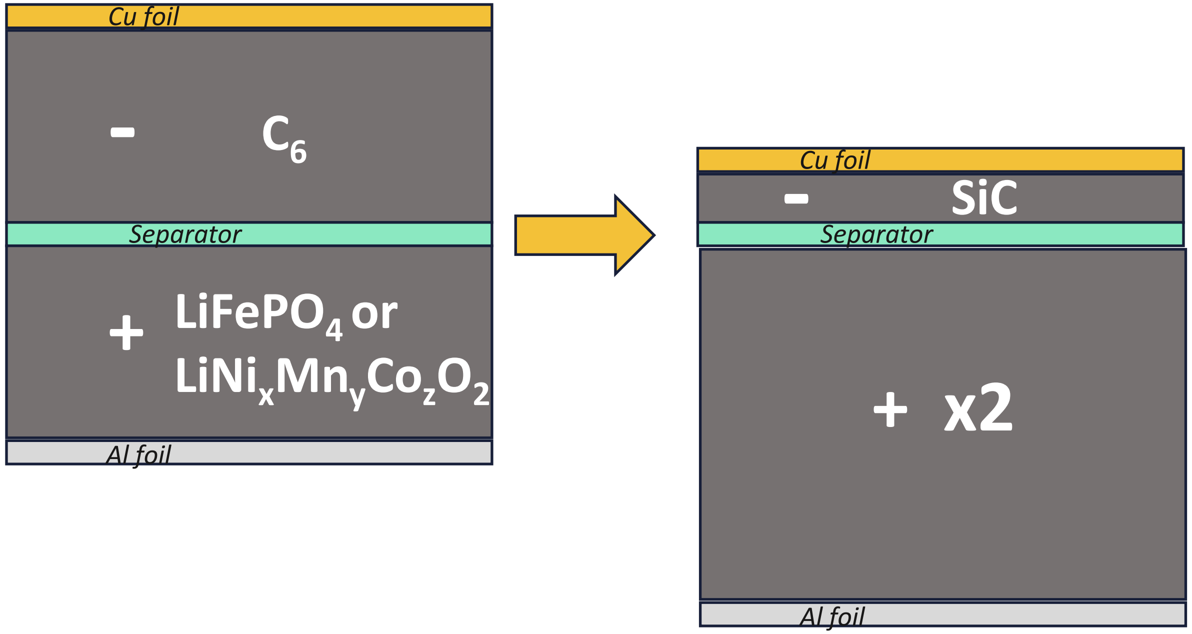
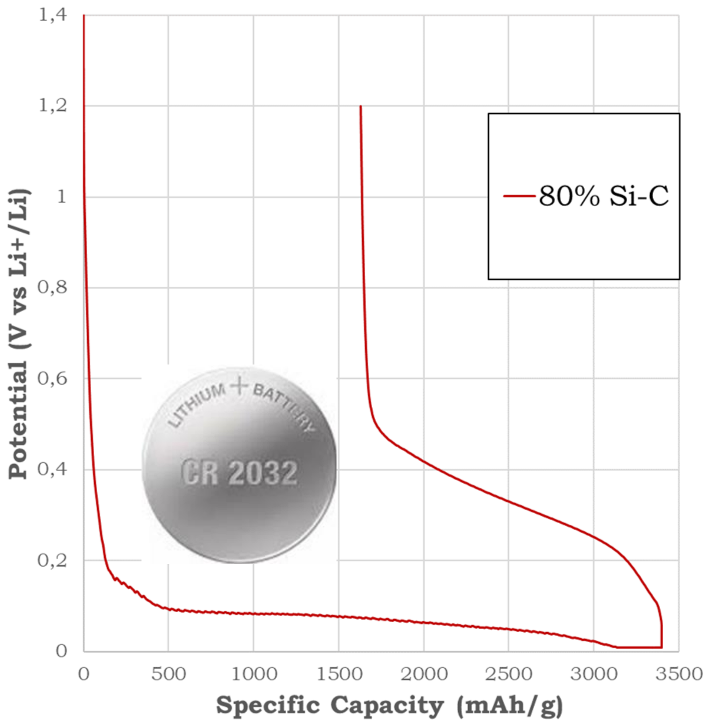


With low ICE, needs to compensate the Li loss by thicker positive electrode and no benefit at the end on the cell energy density



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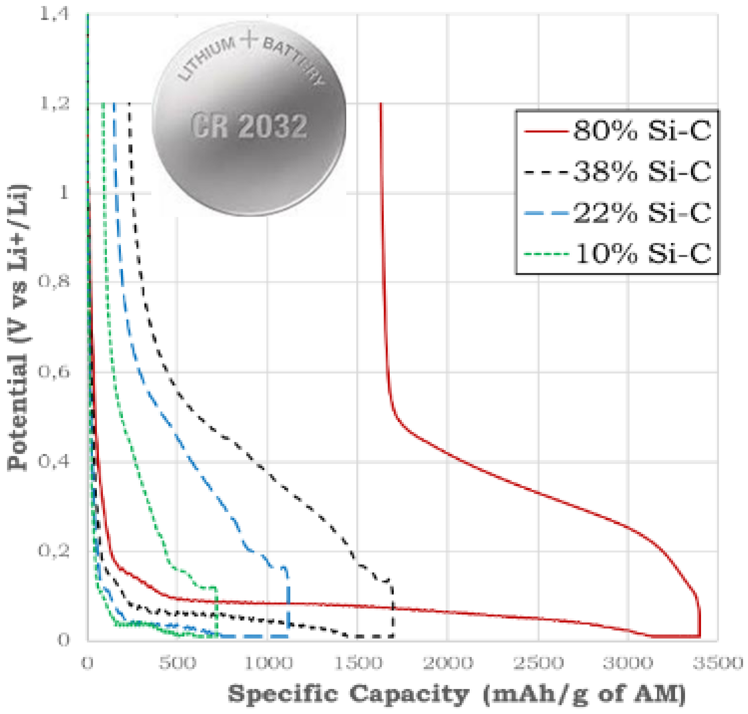
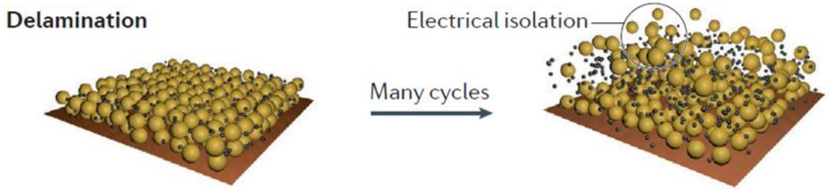
Electrode DESIGN: SiC content investigation

GREEnSiBat–Si (wt. %)	Graphite (wt. %)	C65 (wt. %)	SW-CNT (wt. %)	Na-CMC (wt. %)	Latex (wt. %)	Loading (mg/cm²)	Porosity (%)
33%	0%	33%	0%	33%	0%	0.9	>50%
80%	0%	5%	0%	12%	3%	3.3	35%
38%	50%	2.8%	**0.2%**	5%	4%	3.5	34%
22%	68%	2.8%	**0.2%**	4%	4%	3.3	30%
10%	82%	2.8%	**0.2%**	2.5%	3.5%	5.0	30%



GREEnSiBat–Si (wt. %)	Reversible capacity (mAh/g)	Initial coulombic efficiency (%)
80%	1795	52.7%
38%	1465	86.2%
22%	969	86.8%
10%	632	87.7%

- ICE jumps to suitable value (same order than positive electrode)
- Graphite contribution is visible (2 plateaus below 200 mV)
- Electrical connection in the full electrode thickness should be managed



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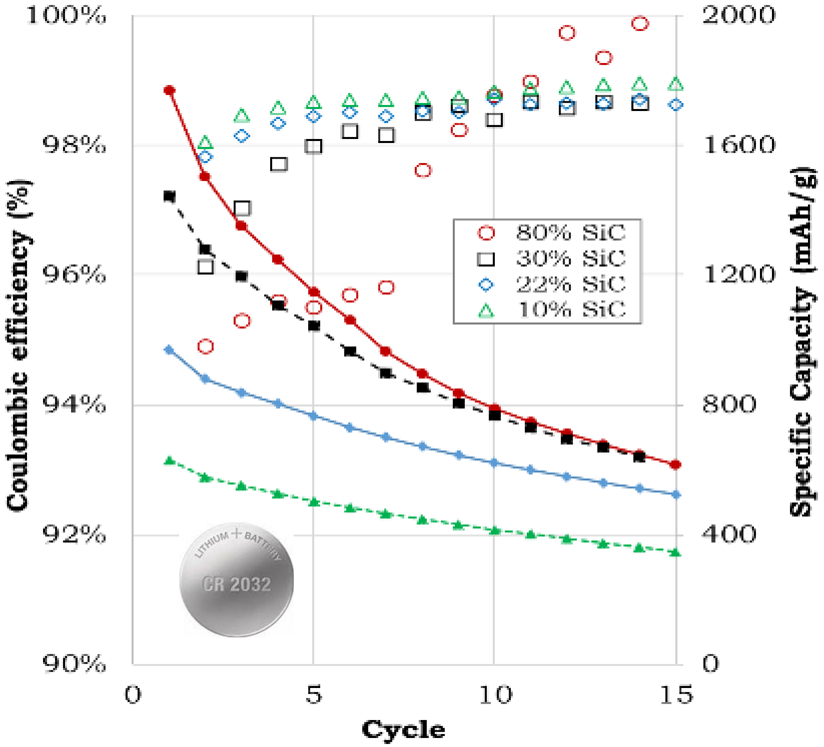
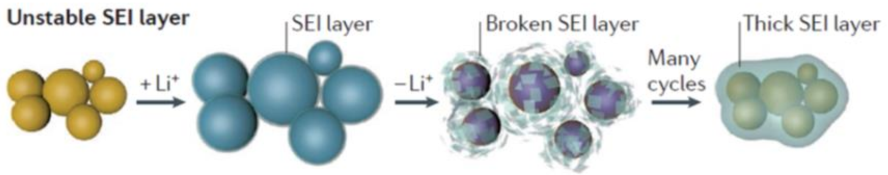


GREEnSiBat–Si (wt. %)	Capacity (mAh/g)	ICE (%)	Mean CE 5th- 15th cycle (%)
80%	1795	52.7%	98.2%
38%	1465	86.2%	98.4%
22%	969	86.8%	98.6%
10%	632	87.7%	98.8%

- Low capacity retention with coulombic efficiency < 99% (to achieve 80% of Q_{ini} after 300 cycles need CE > 99.9%)
- However, no capacity drop – electrode should keep integrity



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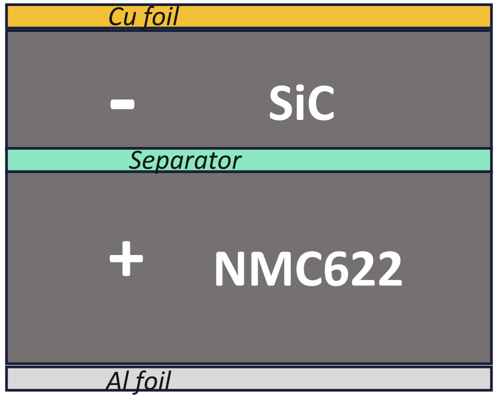
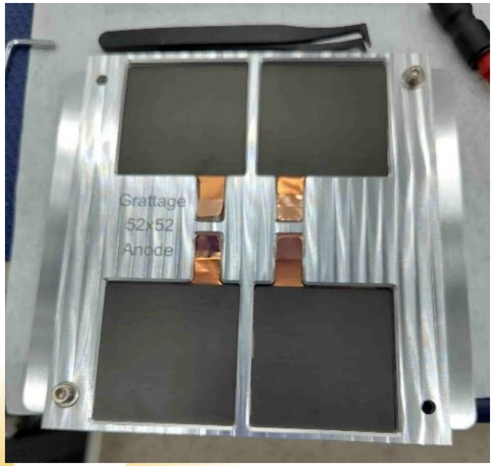
2 Ah Multi-Layer Pouch (MLP) cell assembly

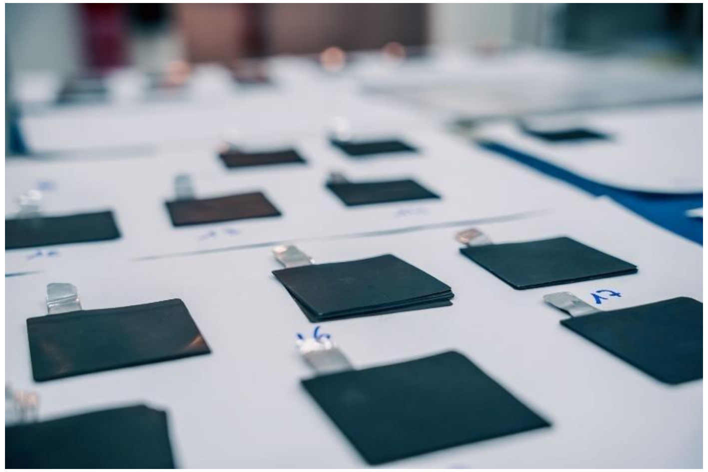


GREEnSiBat–Si (wt. %)	Graphite (wt. %)	C65 (wt. %)	SW-CNT (wt. %)	Na-CMC (wt. %)	Latex (wt. %)	Loading (mg/cm²)	Porosity (%)
10%	82%	2.8%	0.2%	2.5%	3.5%	5.3	40%

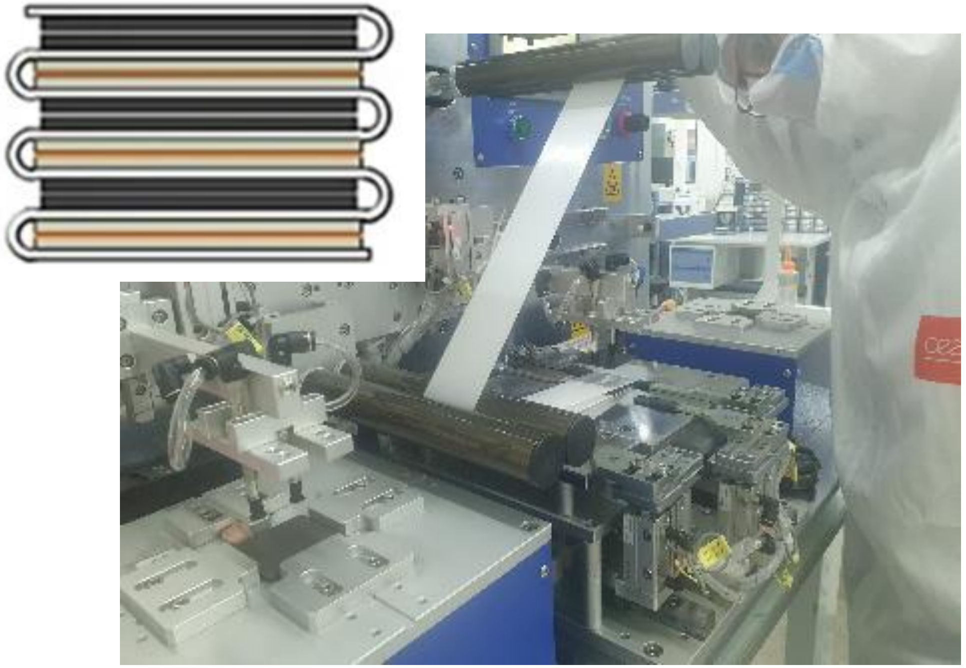
Anode 850 Ah/L vs 570 Ah/L for graphite

NMC622	C65 (wt. %)	PVdF (wt. %)	Loading (mg/cm²)	Porosity (%)
96%	2%	2%	17.0	28%





b) Z-stacking



Anode: 52 x 52 mm² - Cathode: 50 x 50 mm²

Laser cutting of electrodes and scratching on the tab

→ 17 anodes and 16 cathodes per cells = 800 electrodes

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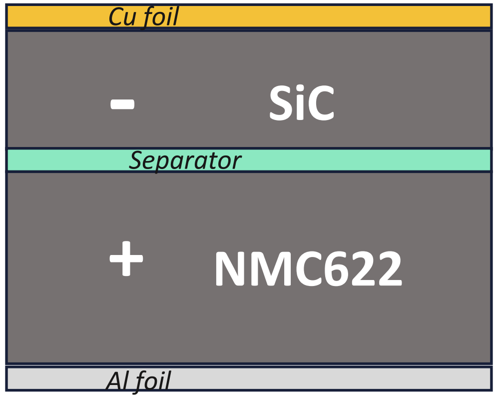
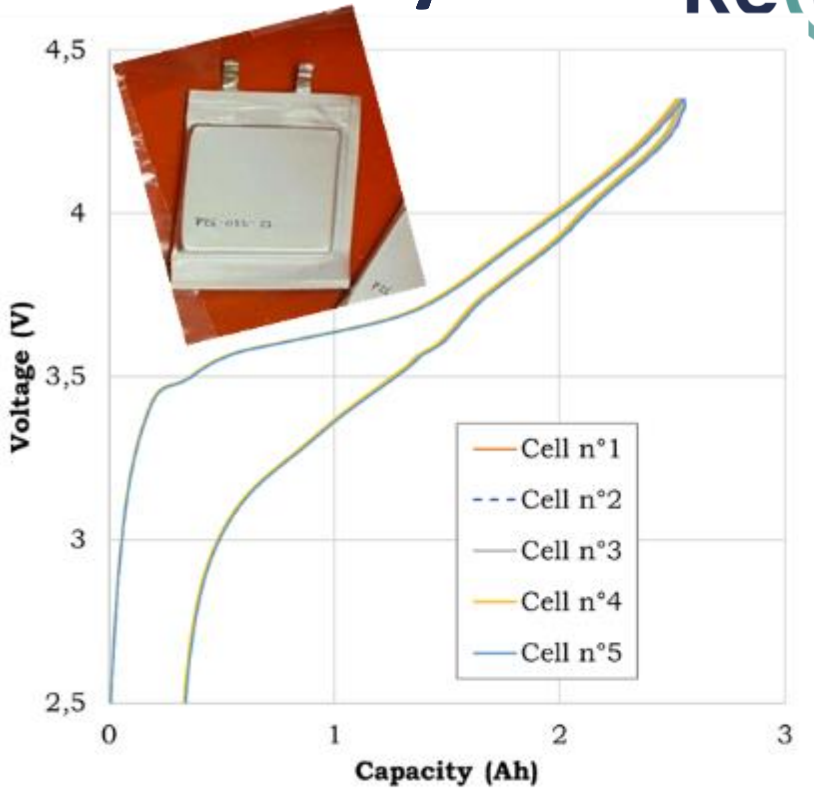


Basic Foil : 20 and 10 µm for Al & Cu
20 µm separator



MLP5050	Thickness before / after formation (mm)	Mass (g)	Capacity (Ah)	Efficiency (%)	Energy density (Wh/kg)	R (mΩ)
22 / 24 cells	4.2 / 4.4	32	2.20 ± 0.02	86.7%	250	36

Cell balancing was 1.10 (that means anode has 10% mean capacity than cathode)
It is for safety reason to not have metal Li (as higher anode surface vs cathode surface)
Efficiency of 87.7% in half-cell → 86.7% due to anode extra surface/loading
→ **250 Wh/kg** (High energy density for 2 Ah cells with basic foils/ separator/ NMC622)

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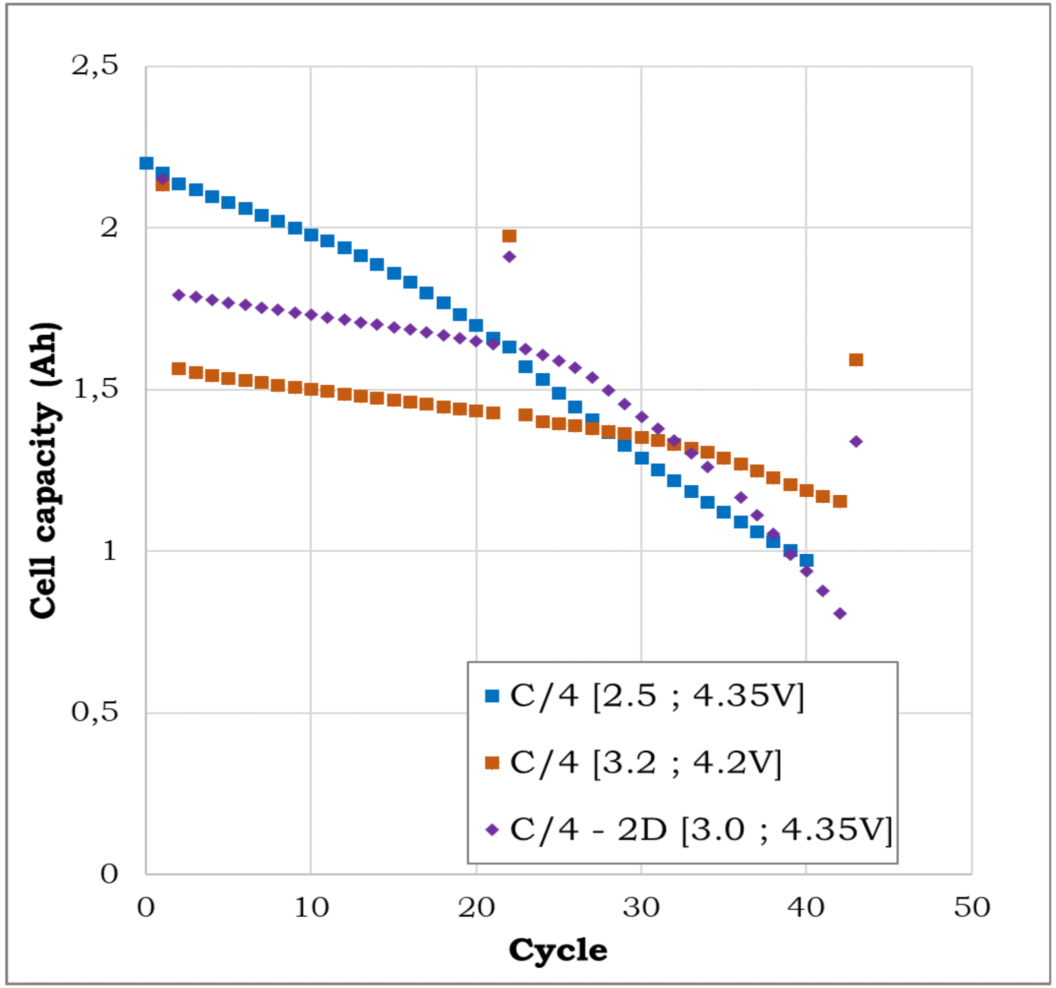
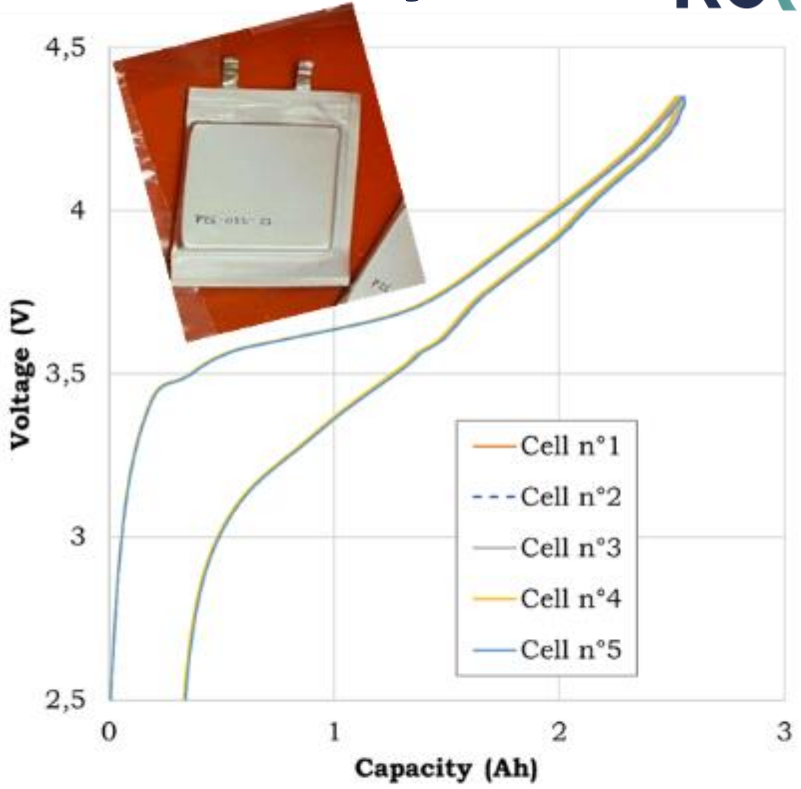
2 Ah Multi-Layer Pouch (MLP) cell assembly





- Capacity loss per cycle of 1% for the 1st cycles when cycling in full window
  → after 15 cycles, fading acceleration > 2% per cycle and < 1Ah after 40 cycles
- Capacity loss of 0.5% when limiting the voltage window
  → Capacity loss mainly due to negative electrode

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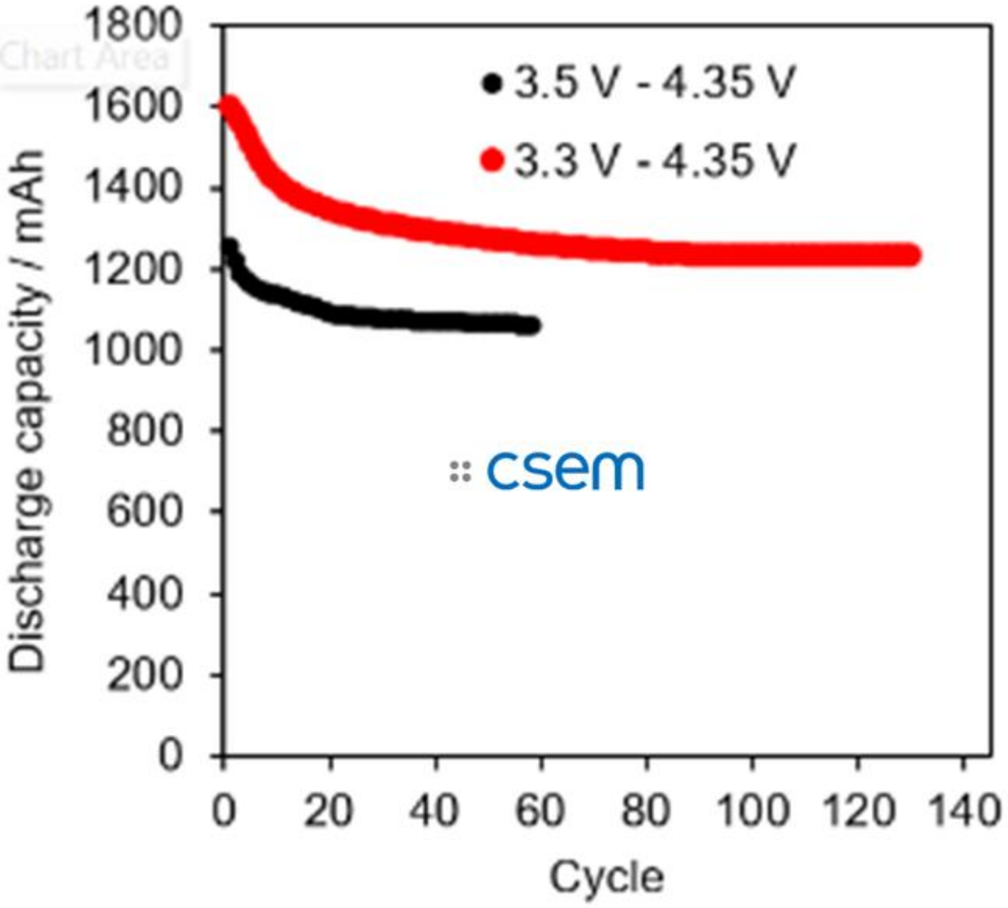
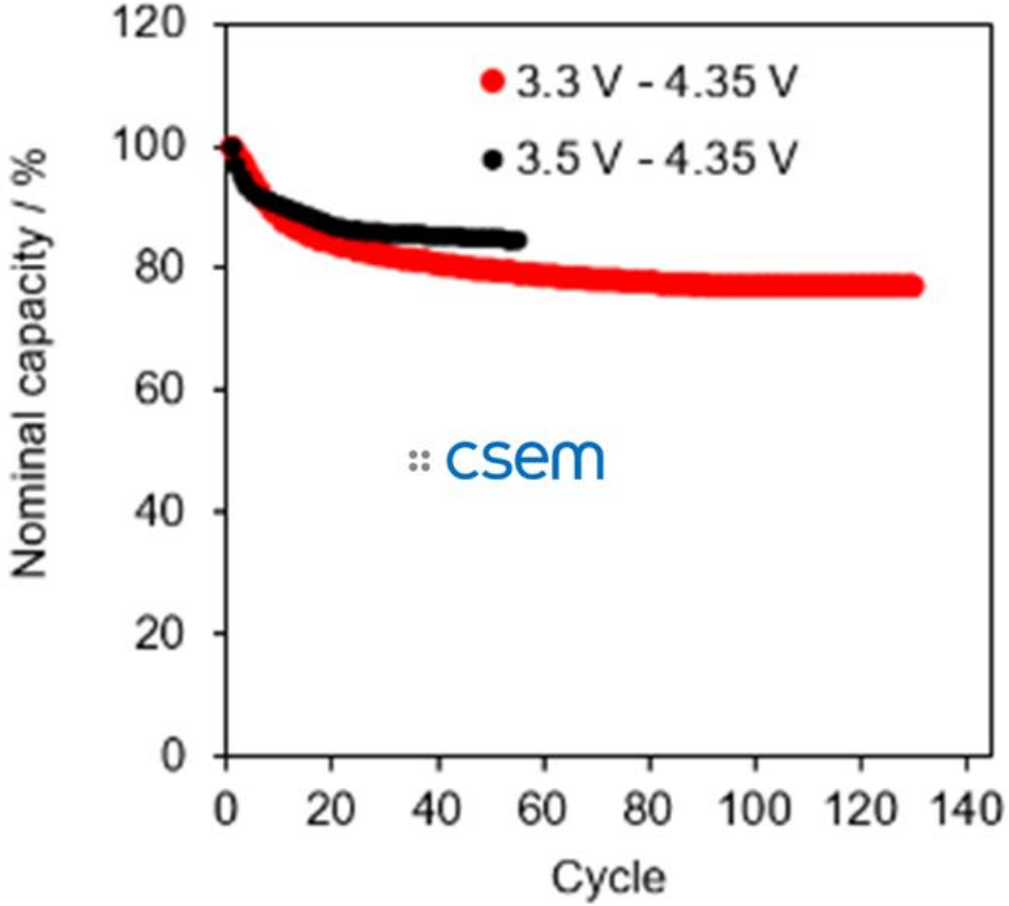
2 Ah Multi-Layer Pouch (MLP) cell assembly





- When limiting the voltage window with remaining capacity to 80 or 60%, the capacity stabilizes (CSEM)

→ Possible to achieve long cycling

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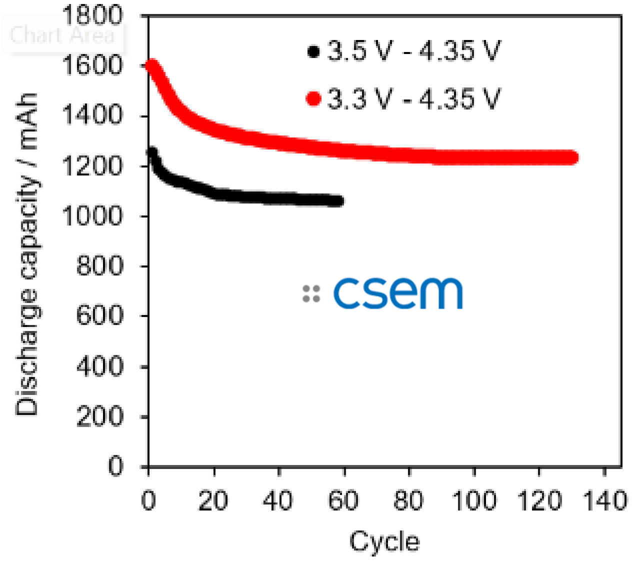

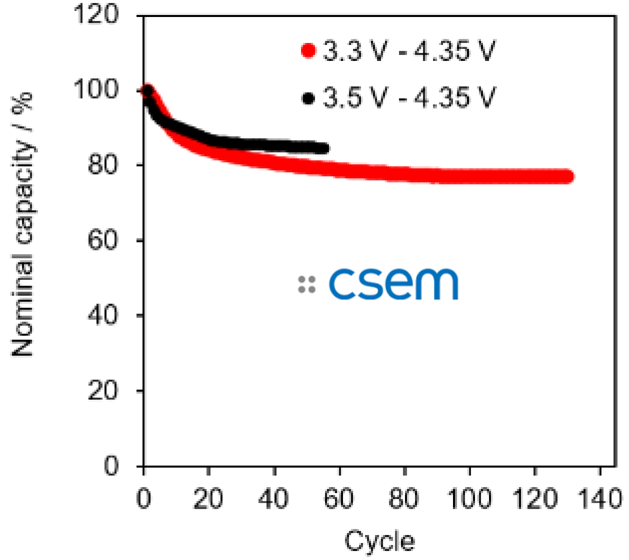

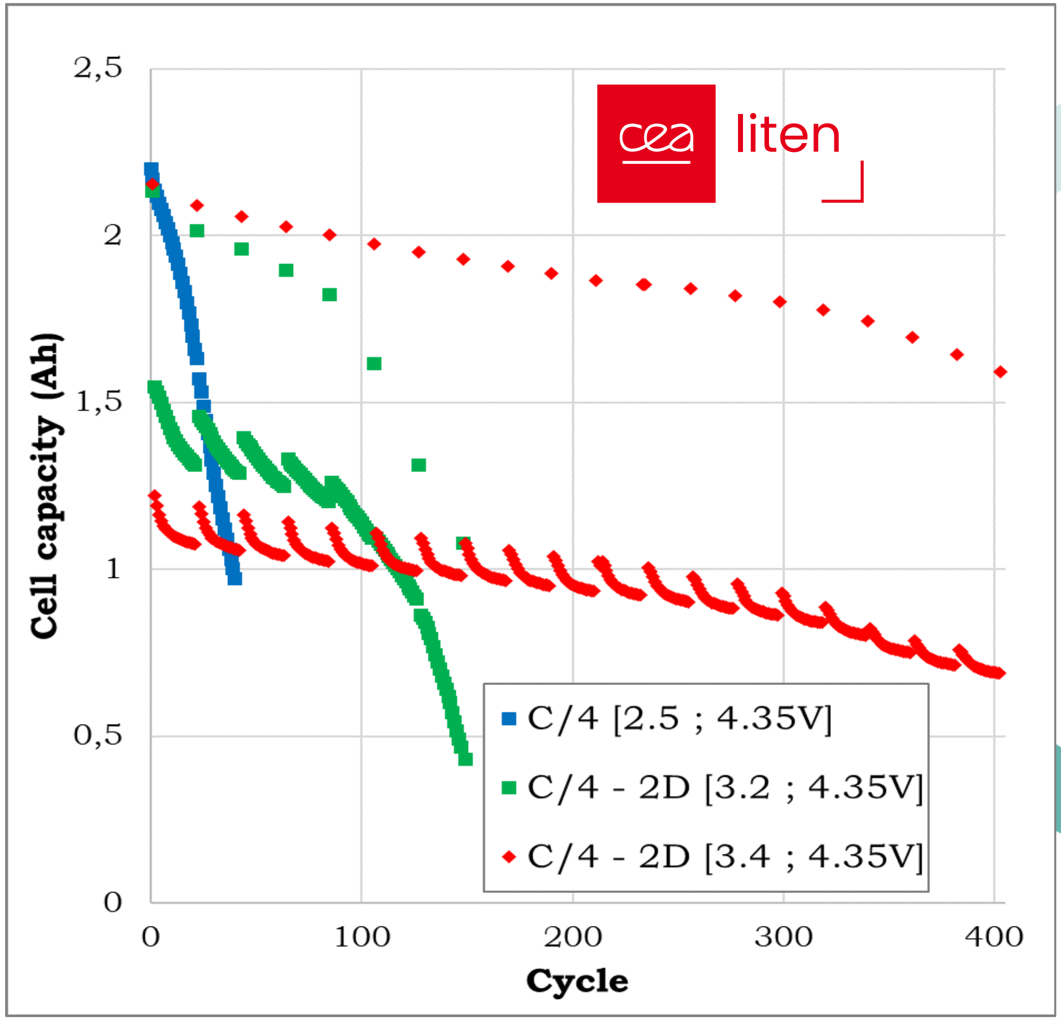

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→ Possible to achieve long cycling

→ Validated at CEA (cycling with regular check-ups (every x20 cycles))

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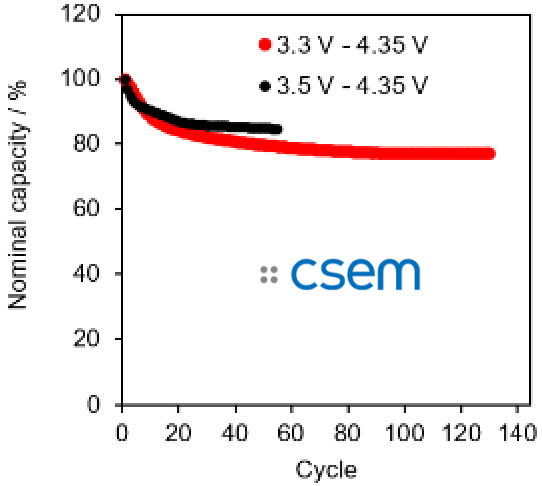

- Electrode formulation with SW-CNT keeps electrical conductivity during electrode breathing
  → initial coulombic efficiency (> 86%) compliant with high energy density cell
- Graphite dilution is a supplemental way to improve the ICE (87.7%)
  → 850 Ah/L at anode level (+50% vs conventional graphite anode)
- 24 MLP cells assembled with 17 anodes and 16 cathodes stacked by Z-folding process:
  - 1% of capacity deviation : 2.20 ± 0.02 Ah
  - Energy density of 250 Wh/kg with basic components
- Long capacity retention demonstrated by CSEM and CEA by limiting the voltage window

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MERCI POUR VOTRE ATTENTION

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