



Nissan Leaf

Electronic pathway length manipulation and impact on conductivity networks in lithium-ion battery electrodes

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 THE FARADAY
INSTITUTION
ELECTRODE MANUFACTURING

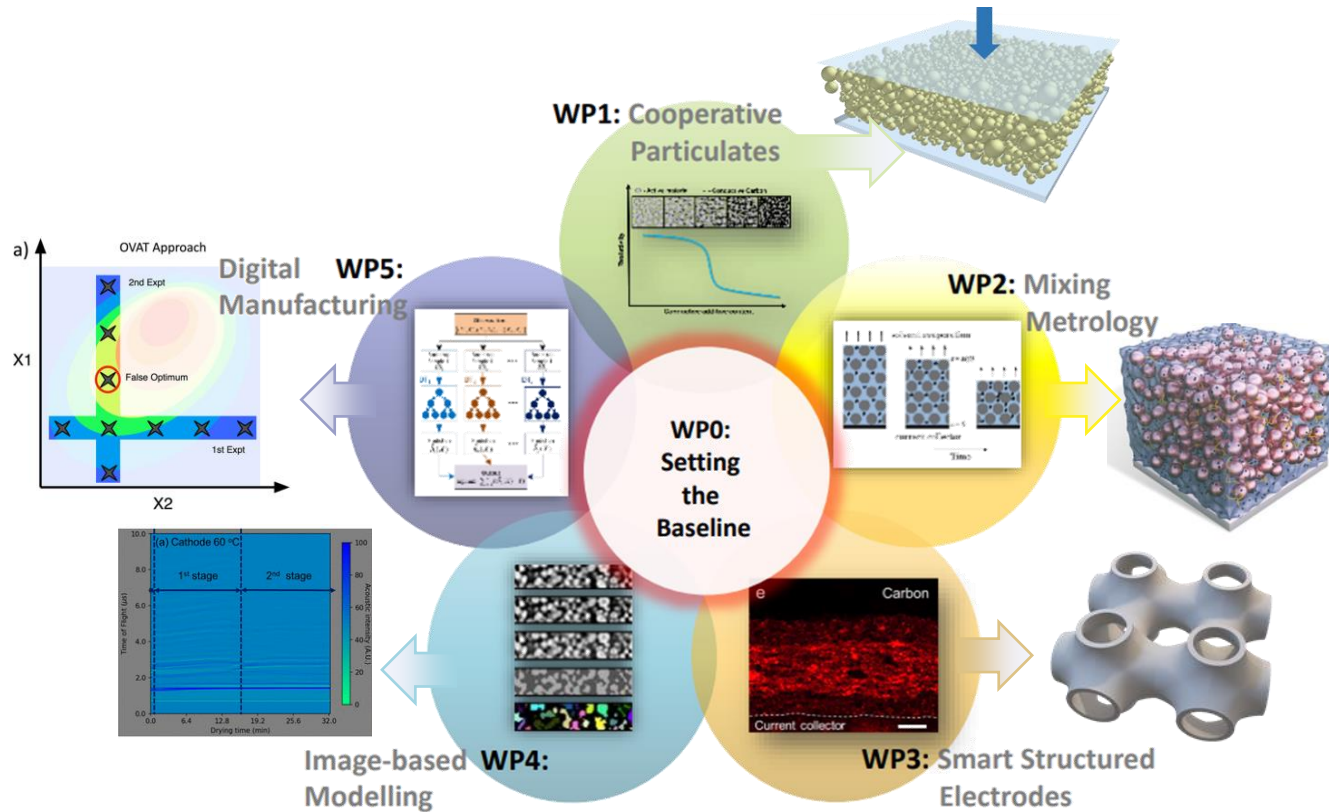
NEXTRODE



Next Generation Electrodes (NEXTRODE)



➤ The Faraday Institution funded project- NEXTRODE



- **WP1: Denis Cumming**
Ruihuan Ge (2nd Nov. 12:30)
- **WP2 and WP4: Yeshui Zhang (poster)**
ACS Appl. Mater. Interfaces 2021, 13, 30, 36605–36620
- **WP3: Yige Sun (1st Nov. 14:50)**
- **WP5: Geanina Apachitei (2nd Nov. 15:35)**

ELECTRODE ELECTRONIC CONDUCTIVITY



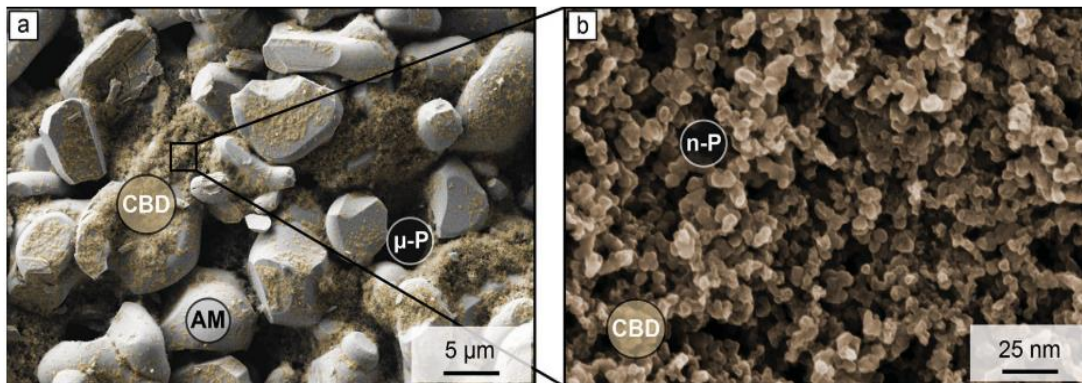
Particle design

Active material
(AM) particle
engineering

- Particle surface coating
- Core-shell structured particles
- Hybrid composite particles (coating, doping)

Carbon binder
domain (CBD)
engineering

- Carbon coating on AM particles
- Graphite/carbon nanotubes
- Intensive dry mixing for homogenous carbon coverage



Electrode design

- High power and energy density requires thicker electrodes
- Efficient electron transport networks with decreasing proportions of conductive additives (CA)
- Incorporation of CA has secondary implications
 - Electrode porosity and tortuosity
 - Compressibility and mechanical properties
 - AM-binder interactions
 - Electrolyte reservoirs and wettability
 - SEI formation
 - Electrode thermal conductivity

LONG- AND SHORT-RANGE ELECTRONIC CONTACTS

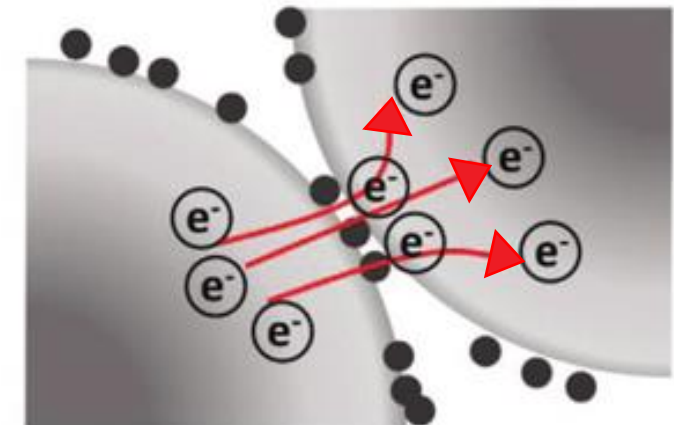
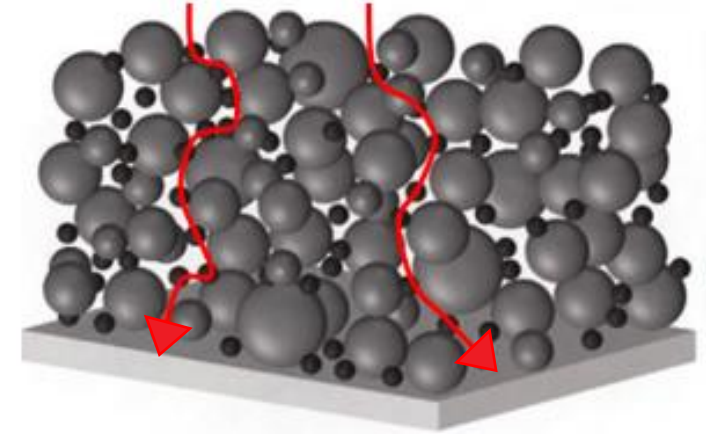


What we know

- Importance of long- and short-range electrical networks
- Destructive and constructive impacts of heterogeneity impacts rate capability^[1]
- Processing techniques affects CBD formation and electronic and ionic networks^{[2][3]}
- Targeted distribution of CA with binder in clusters localized next to contact areas with other AM particles^[4]

No fundamental understanding

- Optimal quantity and location/ distribution of CA in an electrode
- Optimal combination of long- and short-range contacts
- Advance characterization of contact structure
- Simulation of traditional composite or structured electrodes



● Active particle • Carbon black
● Coated active particle

[1] Yari et al., *ACS Appl. Energy Mater.* **2020** 3, 11820-11829

[2] Saraka et al., *ACS Appl. Energy Mater.* **2020** 3, 11681-11689

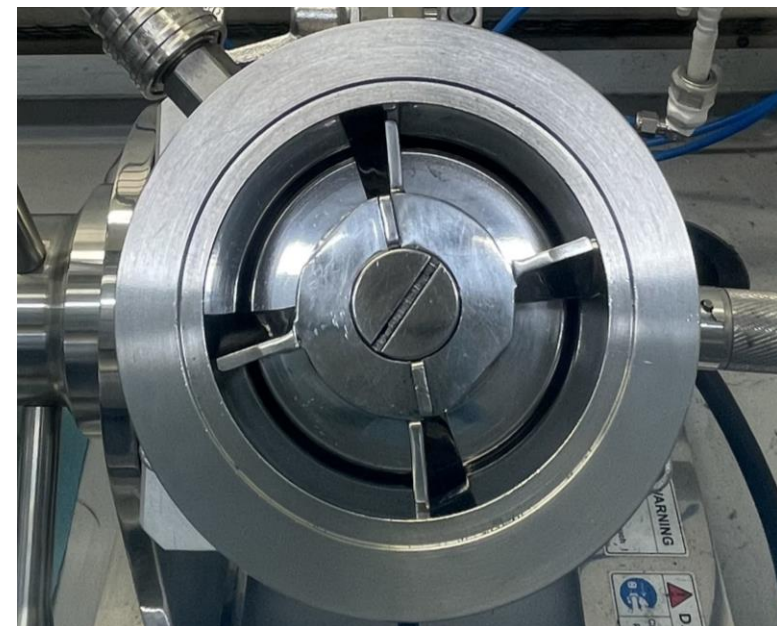
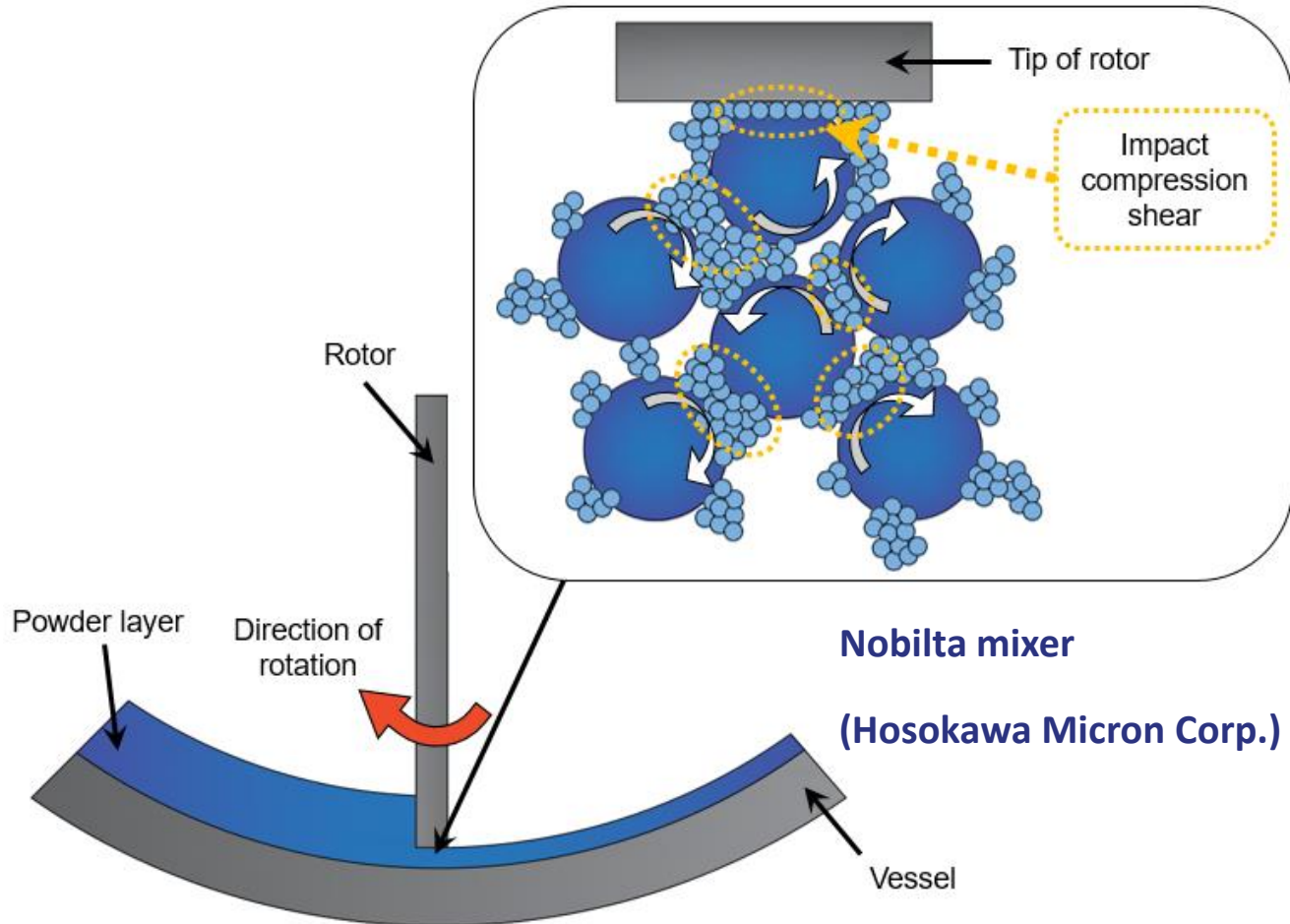
[3] Dreger et al., *Journal of Elec Mater.* **2015** 44, 4434-4443

[4] Bauer et al., *Journal of Power Sources.* **2015** 288, 359-367

Park et al., *Adv. Energy Mater.* **2020** 10

MECHANOFUSION PROCESS

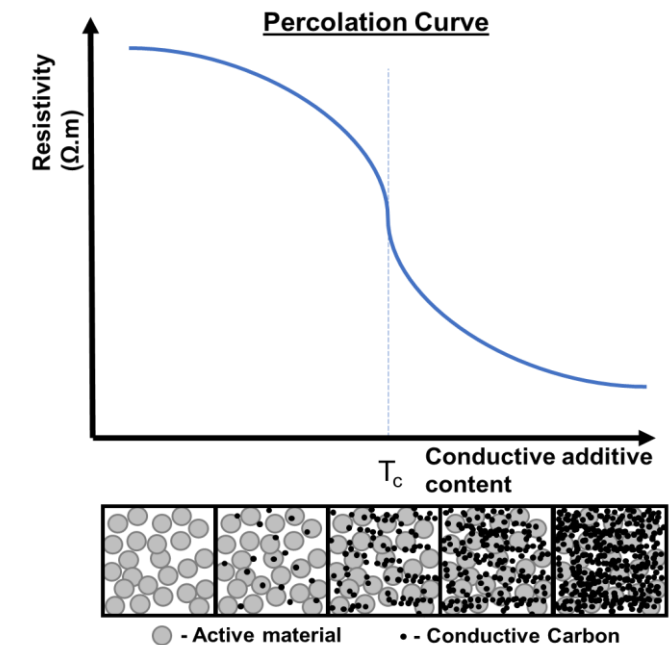
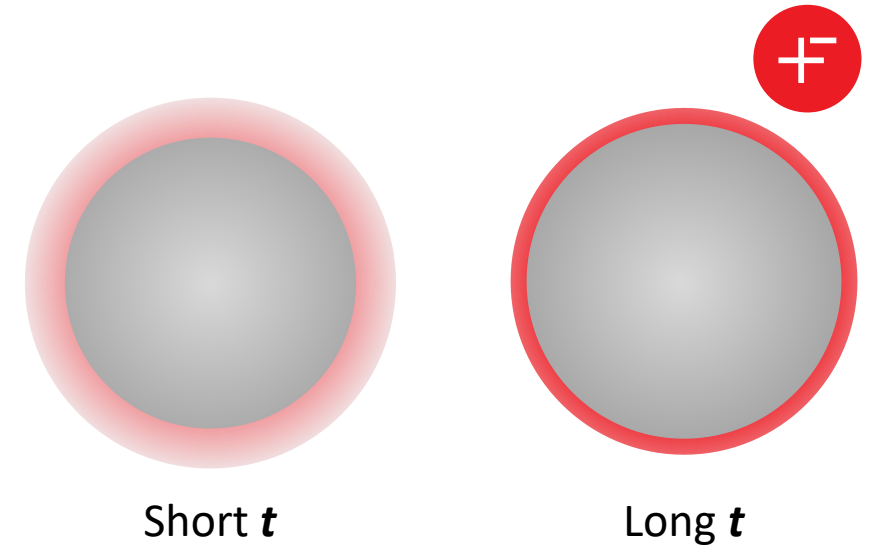
Use of mechanofusion to control degree of carbon black de-agglomeration as a measure of short-range contacts



MECHANOFUSION CARBON COATING

- Constant solids to AM (NMC622 BASF) : binder (PVDF 5130 Solvay)
- Conductive additives – Imerys C65 and KS6 graphite
- AM:B ratio at 98:2wt%
- NMC coated over a range of wt% CB to develop variation in local percolation curve
- Percolation threshold – critical conductive additive concentration

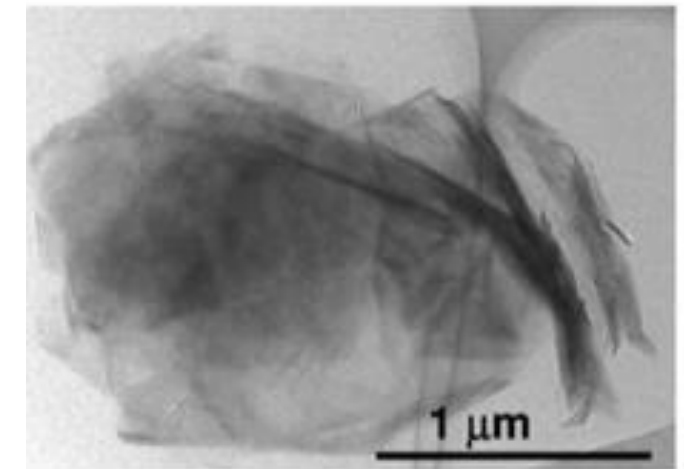
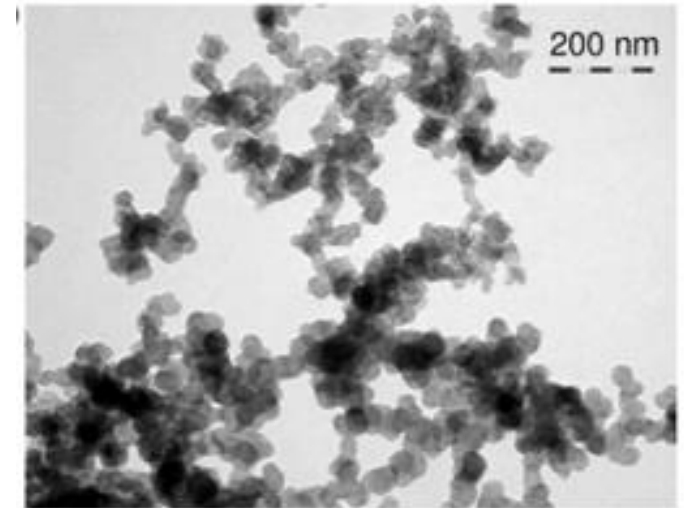
- Short to moderate mixing times
 - CB agglomerates broken down and NMC progressively coated
 - Improvements in electronic conduction
- Long mixing times
 - Compaction resulting in a thin and dense coating
 - Dense layer may impede ionic conduction



LONG- AND SHORT-RANGE CONTACTS



Formulation		Nobilta Mixed (AM:CA wt %)	Slurry Mixed
96:2:2 NMC:C65:PVDF (wt%)			
Baseline (control)	Unknown	-	96:2:2wt% (all slurry mixed)
2% C65 Nobilta	100% short-range	96:2	2wt% – Binder
1% C65 Nobilta +1% C65 in slurry	50% long-range 50% short-range	96:1	1wt% – C65 2wt% – Binder
1% C65 Nobilta +1% Graphite in slurry		96:1	1wt% – Graphite (KS6) 2wt% – Binder



- Studying C65 and graphite and their ability to provide long range contacts
- Graphite used as an alternative LRC
- Comparing this to other formulations with lower carbon amounts (1wt%)
- Potential additional conductive additives e.g. carbon fibers, nanotubes have not been investigated

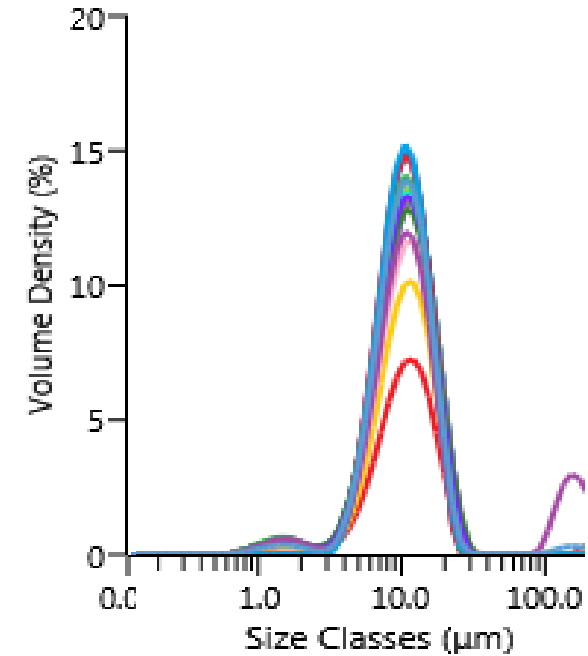
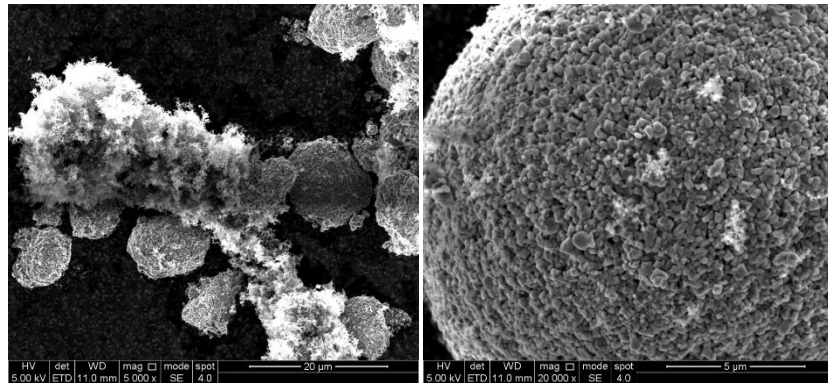
MECHANOFUSION EXPERIMENTS



Investigated 1 -10 wt% coating of C65 on NMC

- Inspected with SEM at mixing times from 0 to 60 min
- Powder conductivity measured at time intervals

t=0 min



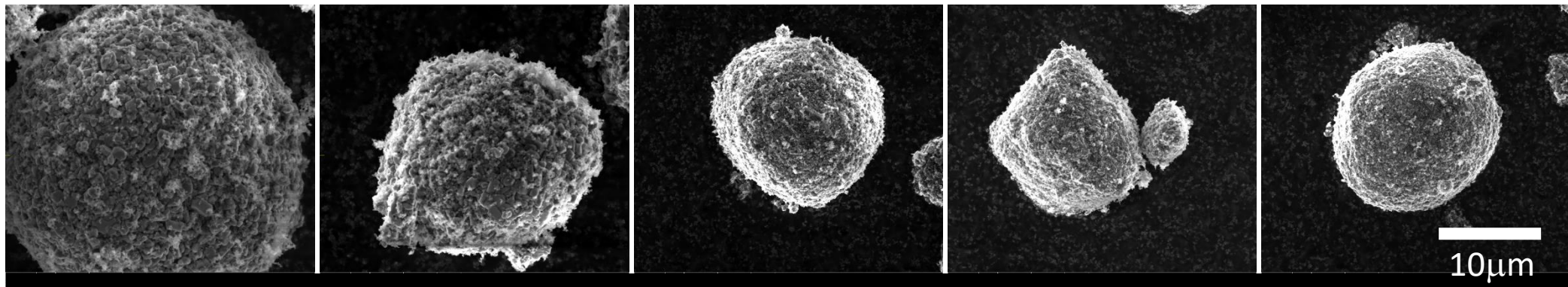
t=2 min

t=5 min

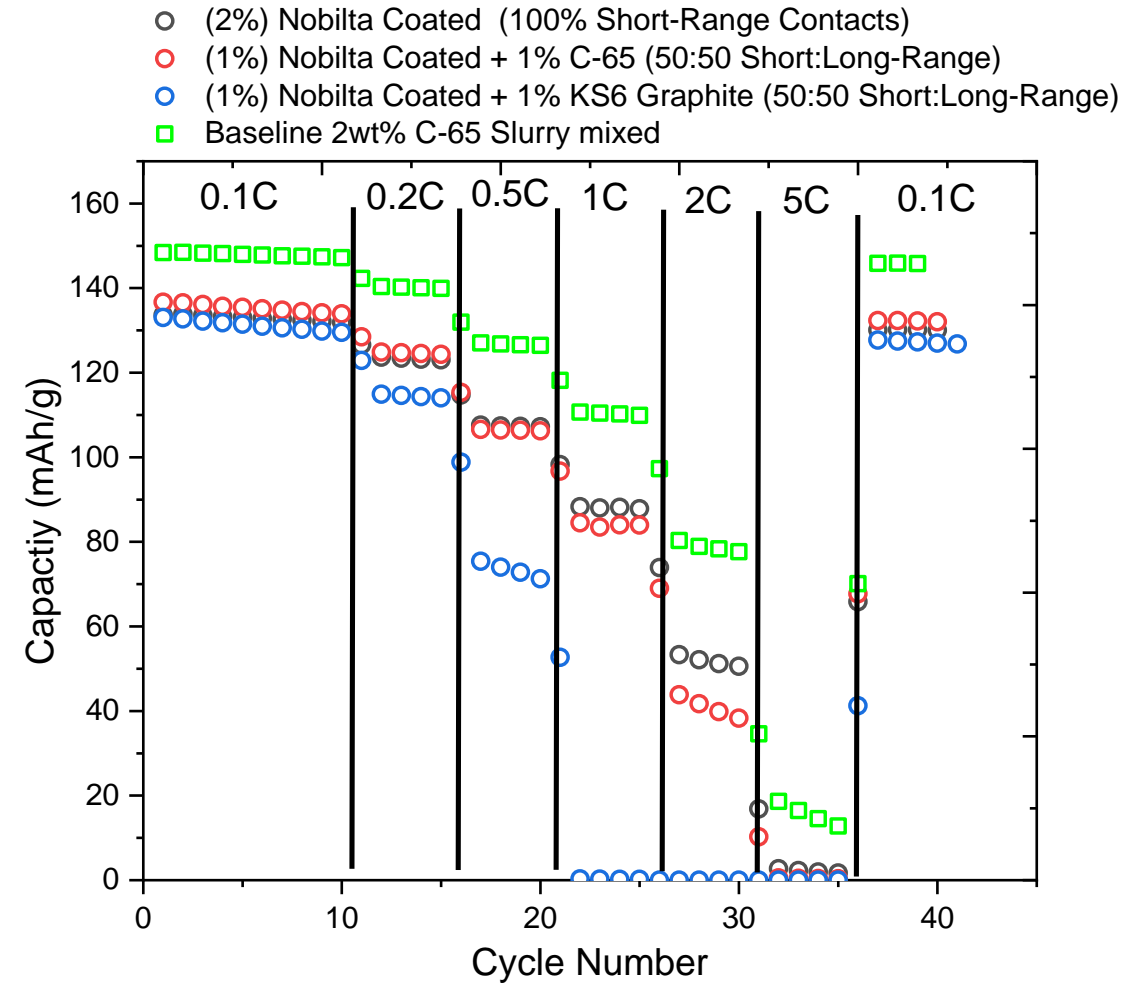
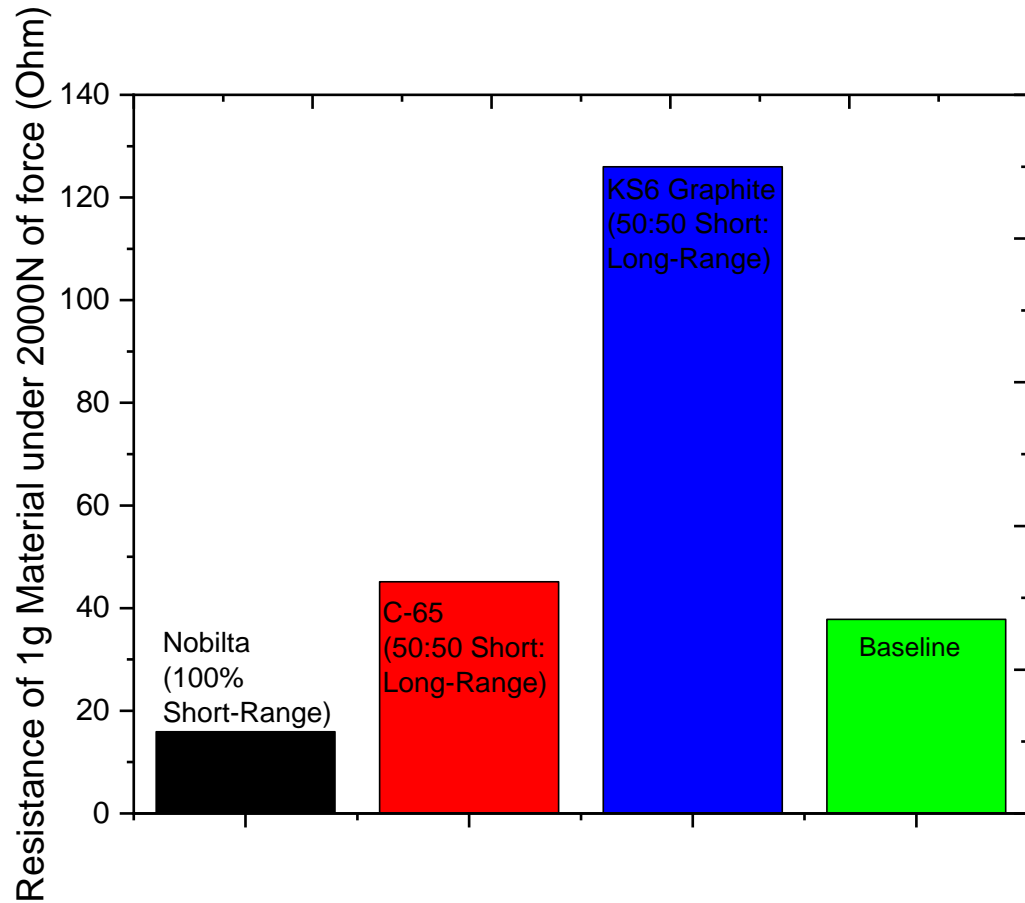
t=10 min

t=30 min

t=60 min



CONDUCTIVITY AND ELECTROCHEMISTRY



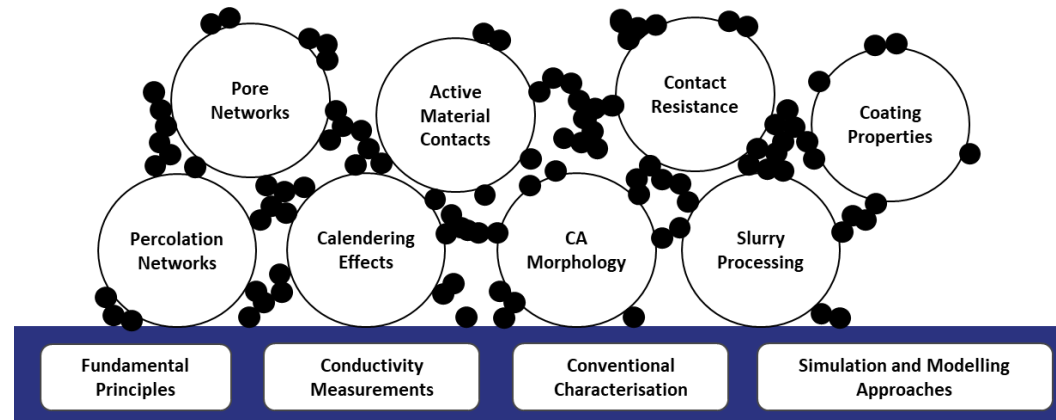
OUTLOOK

Conclusions

- Demonstrated mechanofusion as rapid and effective deagglomeration technique
- Manipulation of carbon black controls electronic conductivity
- Complex relationship between CBD processing and morphology on cell performance
- Binder location may differ in SR and mixed range structures
- Unknown impact of coating on calendering mechanics

Future Work

- Investigate mechanofusion mechanisms
- Further determination of control in long- and short-range electrical contacts
- Why does slurry casting perform so well?
- Best conductive additive for long-range contacts
- Better characterisation techniques to quantify pathways
- **Use our understanding and methods to produce designed conductive pathways and 3D architectures.**



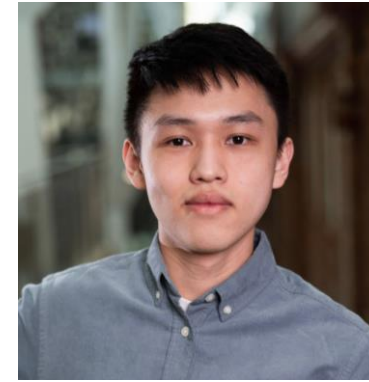
ACKNOWLEDGEMENTS & PARTNER UNIVERSITIES



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Work Package Lead



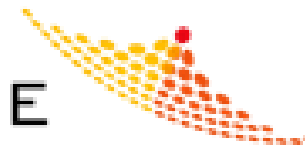
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NEXTRODE



The
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Of
Sheffield.



UNIVERSITY OF
Southampton



UNIVERSITY OF
BIRMINGHAM



WARWICK

Thank you



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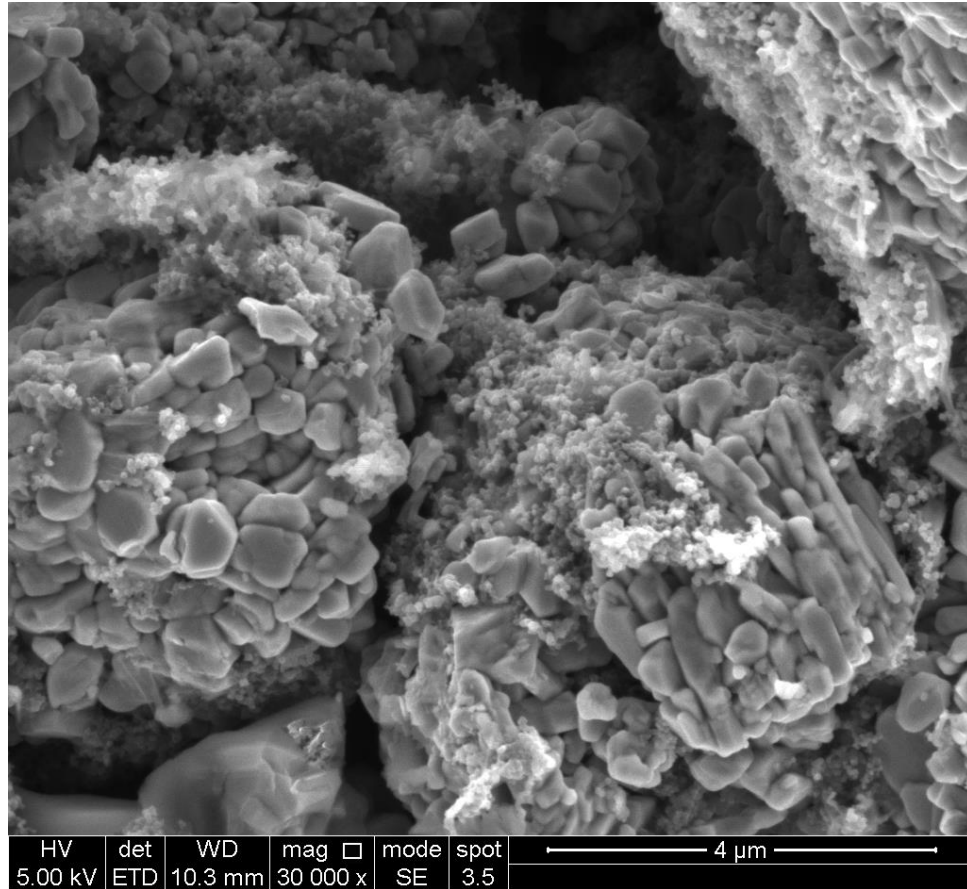
- www.nextrode.web.ox.ac.uk



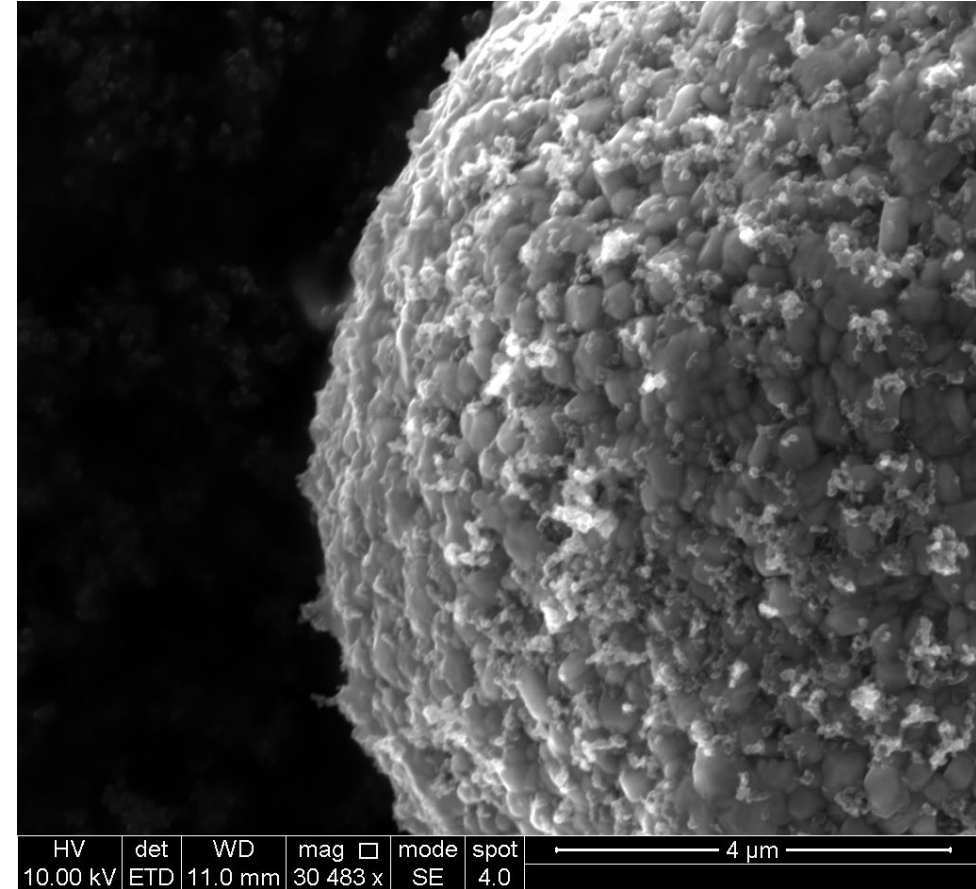
SHORT RANGE CONTACTS



Conventional slurry electrode NMC particle



Mechanofusion coated NMC particle



ELECTROCHEMICAL RESULTS



- Complex relationship between cell performance and CBD morphology
- Further work to investigate electrode pore properties, electrolyte reservoirs, EIS response etc.
- How does coating for short-range contacts change calendaring mechanics

