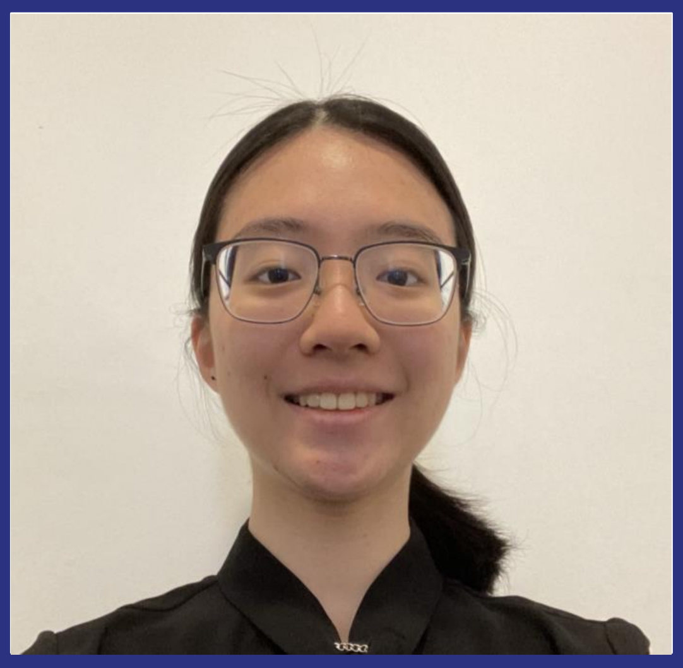
Towards Solvent Free Battery Manufacturing

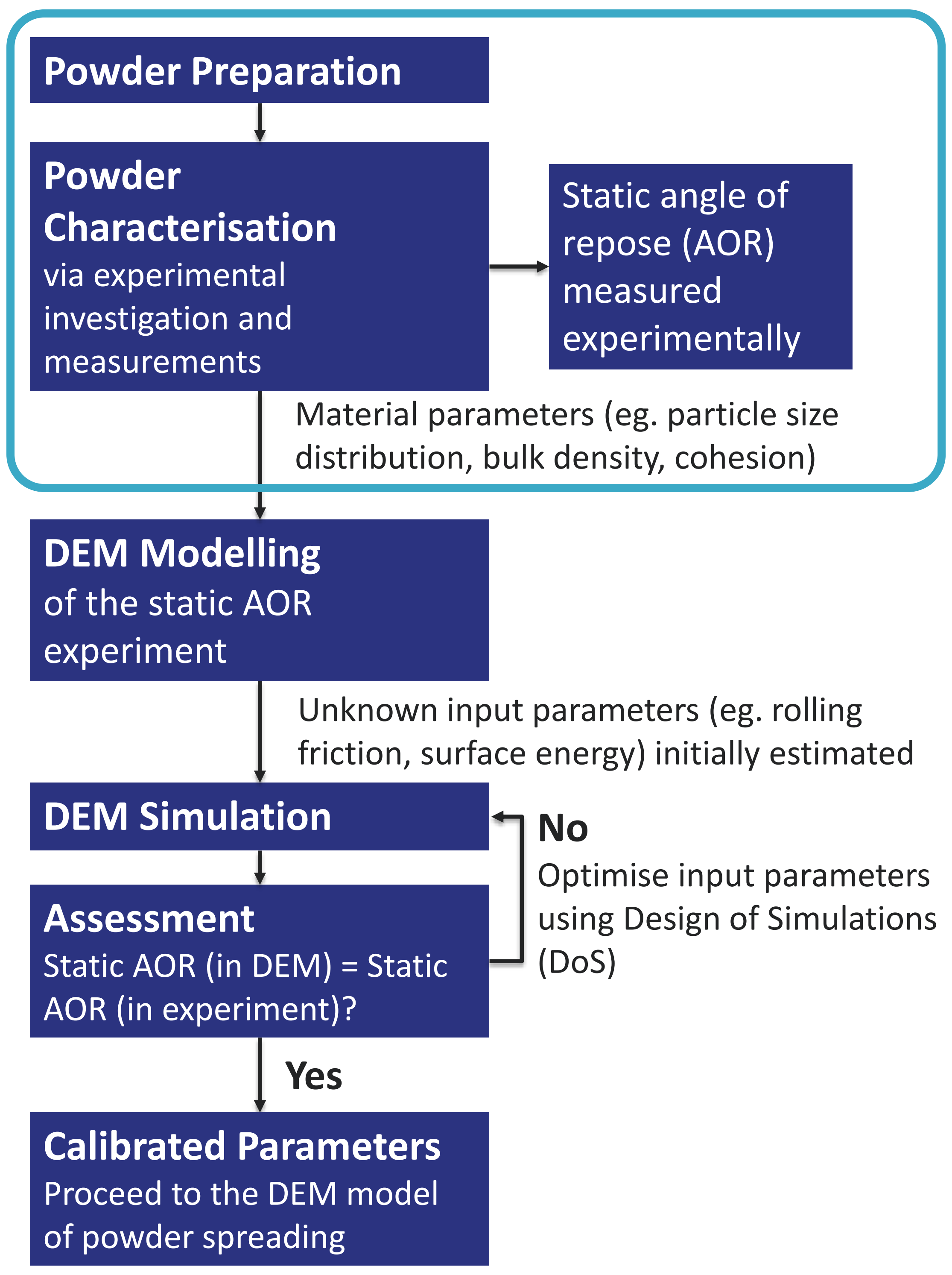
Investigating the characteristics of Lithium-ion cathode active materials for the calibration of Discrete Element Method simulations



Eunice Tan Hamza Mohamed, Dr Ruihuan Ge, Dr Rachel Smith

ABSTRACT

- The interest in additive manufacturing is growing in many applications, including battery production.[1]
- The use of modelling techniques such as DEM helps us predict the behaviour of powder during spreading. [2],[3]
- The calibration will help us understand the powder spreading mechanism to optimise our processes further.



Powder Preparation

Powder Characterisation via experimental investigation and measurements

Static angle of repose (AOR) measured experimentally

Material parameters (eg. particle size distribution, bulk density, cohesion)

DEM Modelling of the static AOR experiment

Unknown input parameters (eg. rolling friction, surface energy) initially estimated

DEM Simulation

No

Optimise input parameters using Design of Simulations (DoS)

Assessment Static AOR (in DEM) = Static AOR (in experiment)?

Yes

Calibrated Parameters Proceed to the DEM model of powder spreading

MOTIVATION

- Dry battery manufacturing methods such as powder spreading is an excellent alternative to conventional wet slurry casting → eliminates the use of toxic solvents and reduces costs
- Experimental investigations are crucial to characterise and calibrate the relevant input parameters for the DEM model of powder spreading.

Project aims to:

1. provide an extensive characterisation on NMC622 and the effects of mechanofusion and carbon additives,
2. calibrate the parameters required for the DEM simulation of NMC622.

METHODS

Powder Preparation

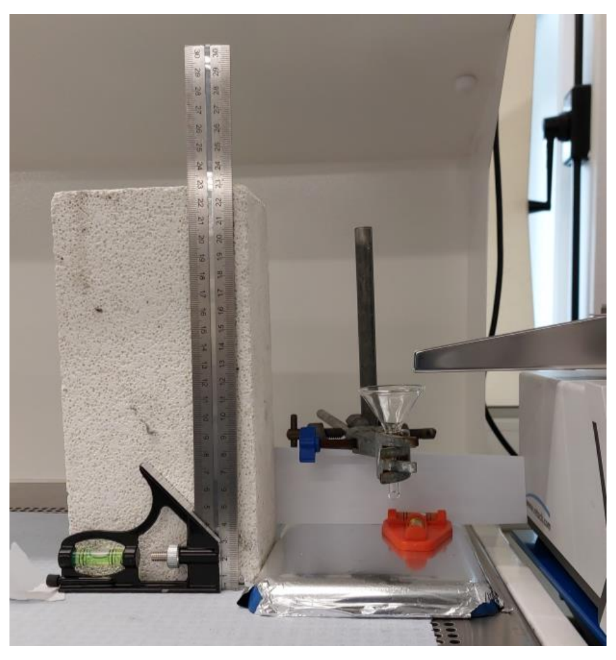
1. NMC622
2. NMC622 MF
3. NMC622:C65 98:2 wt% *(aged)* MF
4. NMC622:C65 98:2 wt% MF

*MF – Mechanofused for 1 hour at 2300 rpm

Powder Characterisation

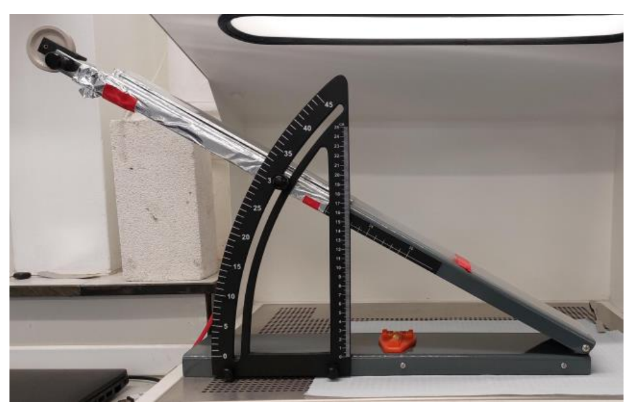
Static angle of repose

Measure angle with cone height diameter method



Inclined plane test

Pelleted tablet tilted on surface till point of sliding

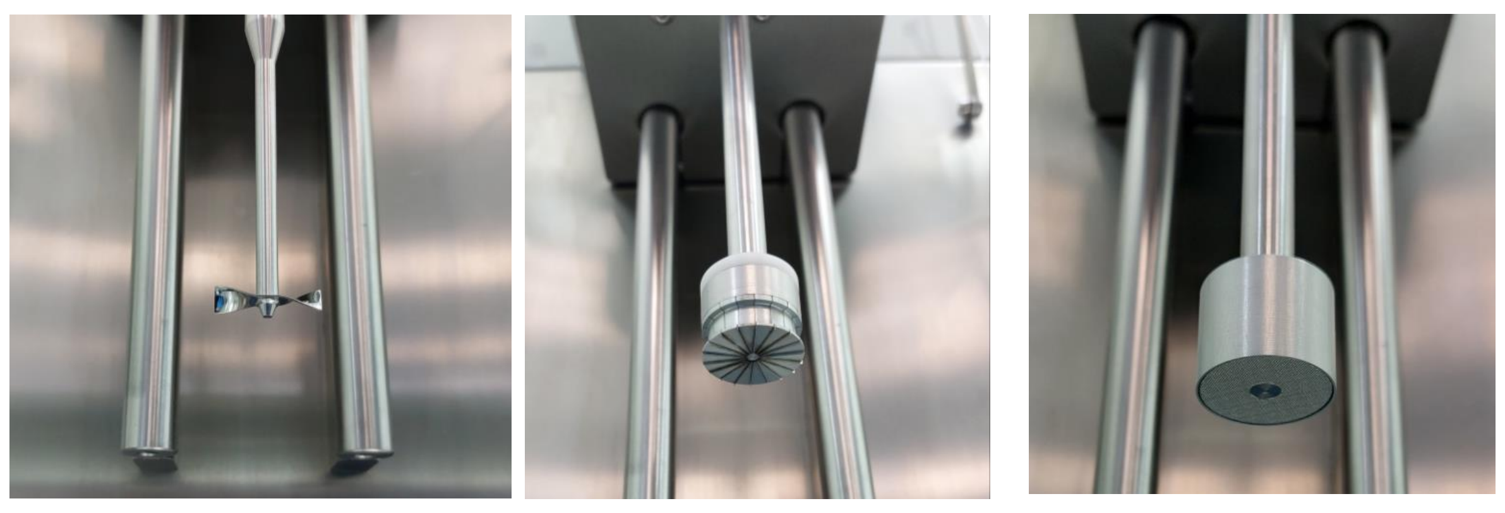


Pre-programmed by FT4 Rheometer

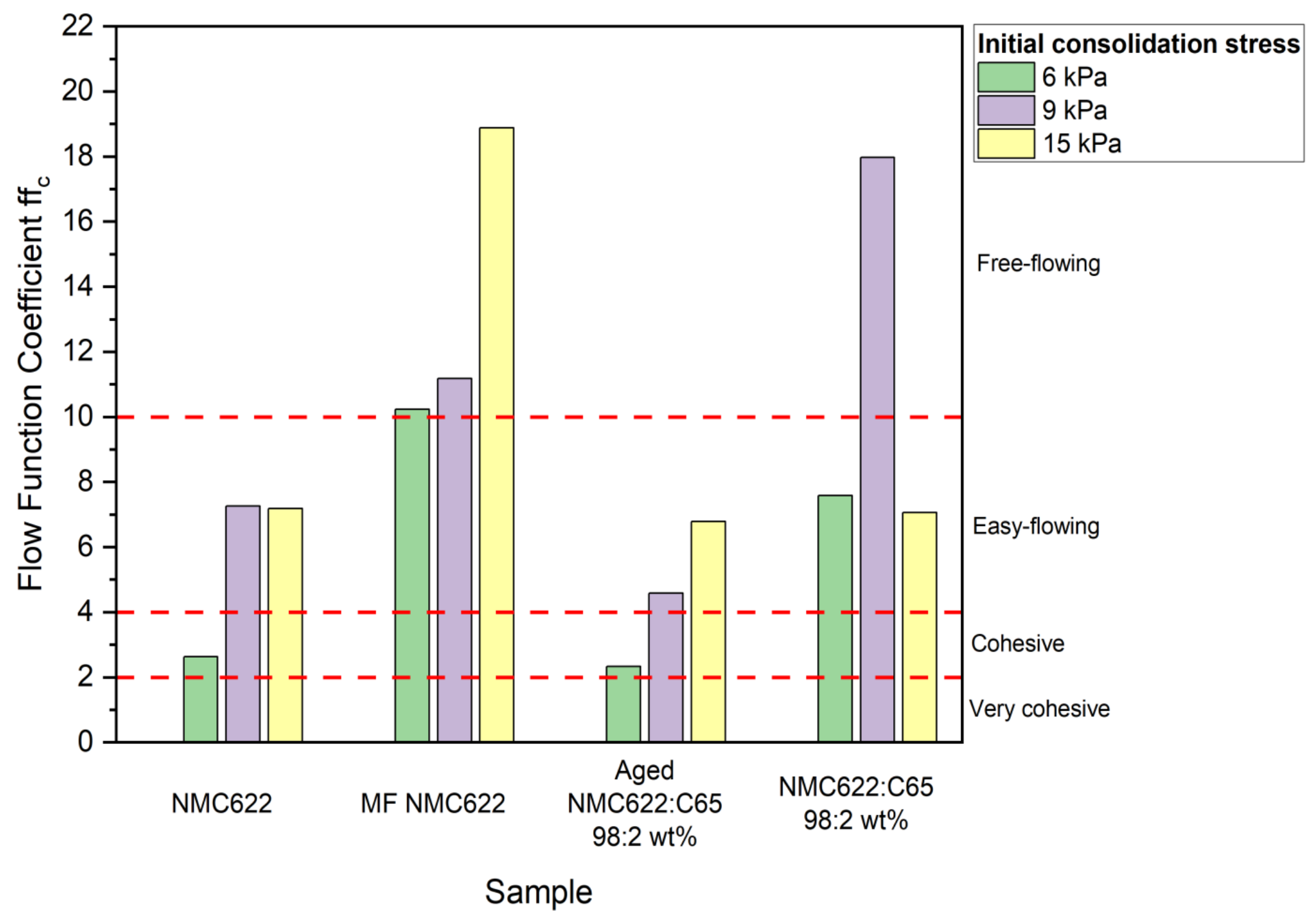
Stability & variable flow rate

Shear cell

Wall friction



FLOWABILITY: ff_c, SPECIFIC ENERGY & BFE



- Low cohesion leads to better unconfined flow and confined flow
  - Low specific BFE = Good confined flow
  - Low specific energy = Good unconfined flow
- The flowability of the powder samples is highly dependent on the consolidation stress applied.
  - Generally observing improved flow at higher consolidation stresses for all samples.

What improves the flowability of powder?

- ✓ Pure mechanofusion
- ✓ Low moisture content (no ageing)
- ✗ Mechanofused addition of C65

**BFE – Basic Flowability Energy*

ff_c – Flow Function Coefficient

COEFFICIENT OF STATIC FRICTION (PARTICLE – SURFACE)

Inclined plane test

Pressed Tablet

Approx. 3.5 g under 3 tonnes

- ✓ Quick and easy measurement
- ✗ Pressed tablet is extremely fragile and prone to breakage

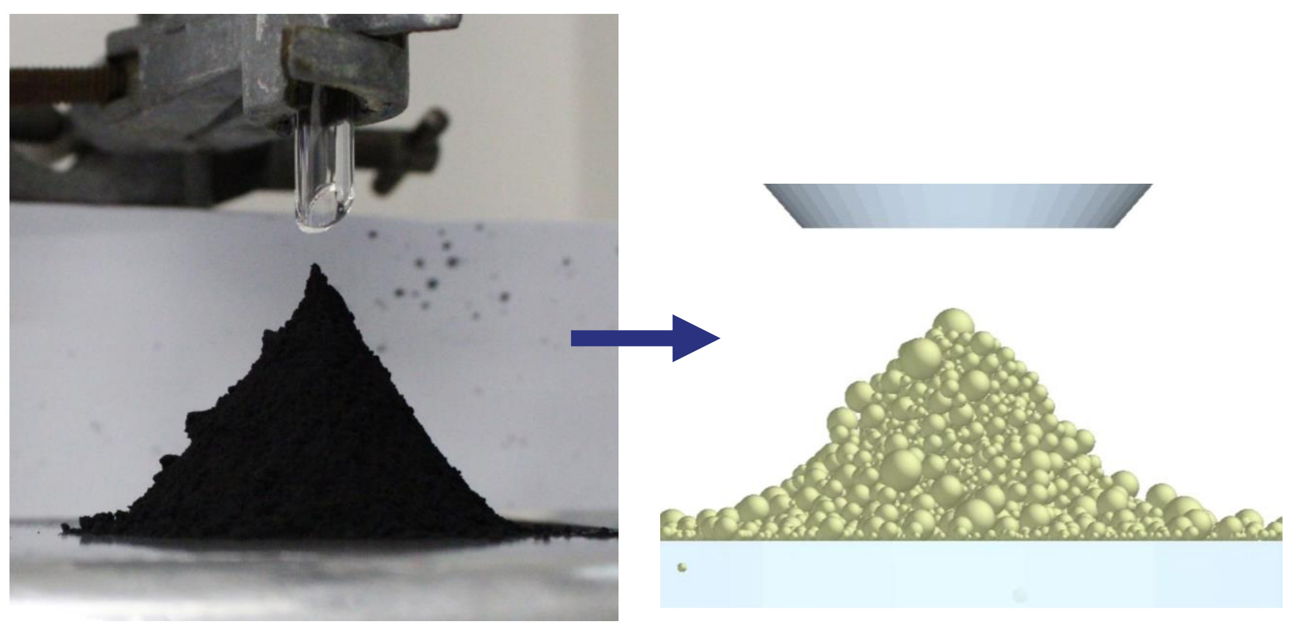
Wall friction test

- Current collector foil: Ra 0.85 µm
- ✓ Provides the option to measure the desired surface roughness
- ✗ Neglects the effect of surface energy

Sample 1: NMC622

Test method	Static friction coefficient
Inclined plane	0.45
Wall friction	0.445 – 0.497

STATIC ANGLE OF REPOSE (AOR) CALIBRATION



Experimental Results

- Sample 3 has a much higher AOR than the rest
  → aged sample has **higher moisture content, which increases cohesivity**
- AOR has low sensitivity towards the effects of mechanofusion and C65 additives despite observing obvious changes in flowability.

Sample	Static AOR (°)
1	43.6 ± 1.8
2	44.3 ± 2.1
3	47.3 ± 2.0
4	43.1 ± 0.7

CONCLUSION

- Based on the characterisation, a DEM model was successfully calibrated for Sample 1, with the following finalised input parameters:
  Sliding friction (particle – surface): 0.3; surface energy (between particles): 1.5 mJ/m^2; surface energy (on current collector): 1 mJ/m^2
- The effects of C65 addition and ageing produces a change in flowability based on the FT4 measurements, but subtle changes in the static AOR experiment.

FUTURE PLANS

1. Calibrating **all four samples** on DEM will require the assessment of more than one bulk property.
   - Construct a DEM model of the wall friction and uniaxial compression tests → compare with the existing experimental data obtained
2. Investigate the effects of increasing the percentage of C65 on the flowability of NMC622.
   - Identify the optimum amount of C65 to achieve desired flow
3. Experimentally obtain the input parameters for DEM to reduce the DoS matrix.

REFERENCES

[1] Ghadiri, M 2018, 'Jamming during particle spreading in additive manufacturing'. *Powder Technology,* Vol. 338, https://www.sciencedirect.com/science/article/pii/S0032591018305278.

[2] Lupo, M, 2021, 'Calibration of DEM for Cohesive Particles in the SLS Powder Spreading Process'. *Processes*, Vol 9, https://doi.org/10.3390/pr9101715.

[3] Lischka, C. Nirschl, H. 2023, 'Calibration of Li-Ion Cathode Materials for Discrete Element Method Simulations'. *Energy Technol.*, Vol 11, https://doi.org/10.1002/ente.202200849.

BIOGRAPHY

Eunice is currently studying Mechanical Engineering at Imperial College London. She is interested in exploring the ever-growing battery industry. In the future, she aspires to work in impactful research that would lead towards the sustainable future of batteries.

THE FARADAY INSTITUTION
ELECTRODE MANUFACTURING
NEXTRODE
University of Sheffield
UK Research and Innovation