Supporting Information for

Simulation of Bi-layer Cathode Materials with Experimentally Validated Parameters to Improve Ion Diffusion and Discharge Capacity

Authors:

Ridwanur Chowdhury[a] , Aayan Banerjee[a,b] ,Yan Zhao[c], Xinhua Liu[d] , Nigel Brandon[a]

[a]Department of Earth Science and Engineering, Imperial College London, UK
[b]Catalytic Processes and Materials, Faculty of Science and Technology, University of Twente, Netherlands
[c]Department of Mechanical Engineering, Imperial College London, UK
[d]School of Transportation Science and Engineering, Beihang University, China

SEM and PSA data for the as-received secondary (big) NMC811 particles

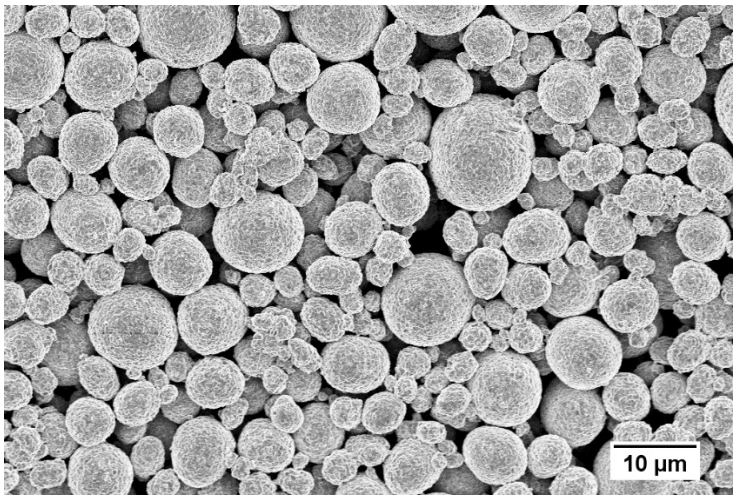

Figure S1. SEM image of as-received secondary NMC particles (powder sample)

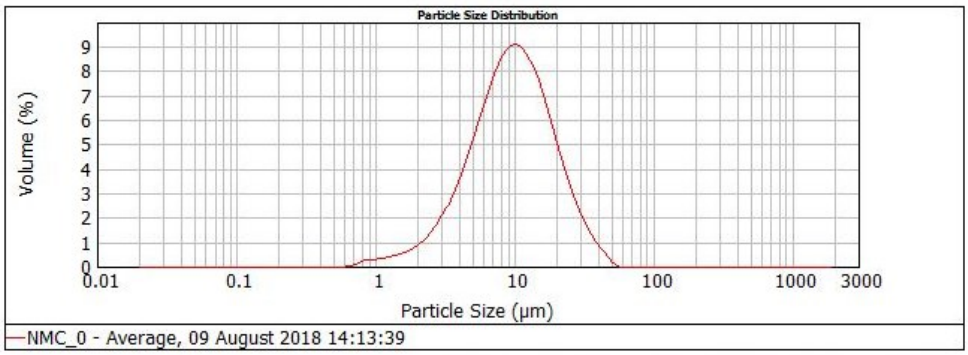

Figure S2. Size distribution of secondary (big) particles using PSA

XRD patterns comparison for as-received (0 hr) and 10 hours (10 hr) ball-milling

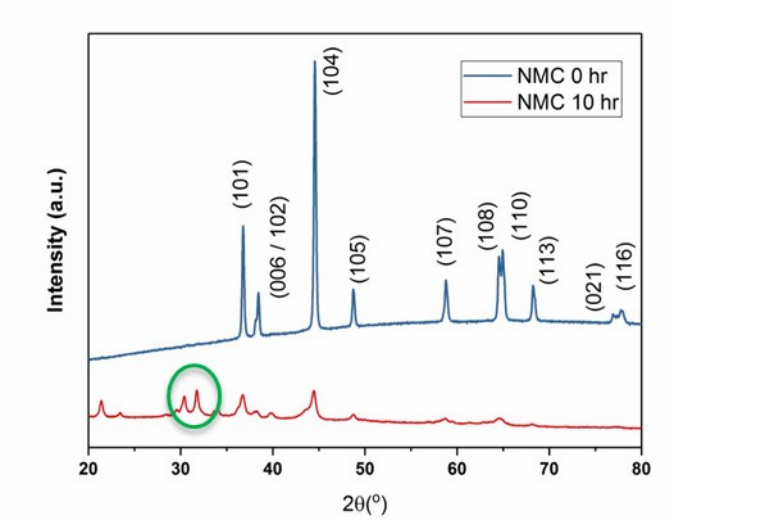

Figure S3. XRD patterns for NMC811 powder samples

The green circle in Fig. S3 shows possible formation of Li_2CO_3 due to longer interaction with deionized water during increased ball milling time (10 hours).

Mass loading data for both small and big particles

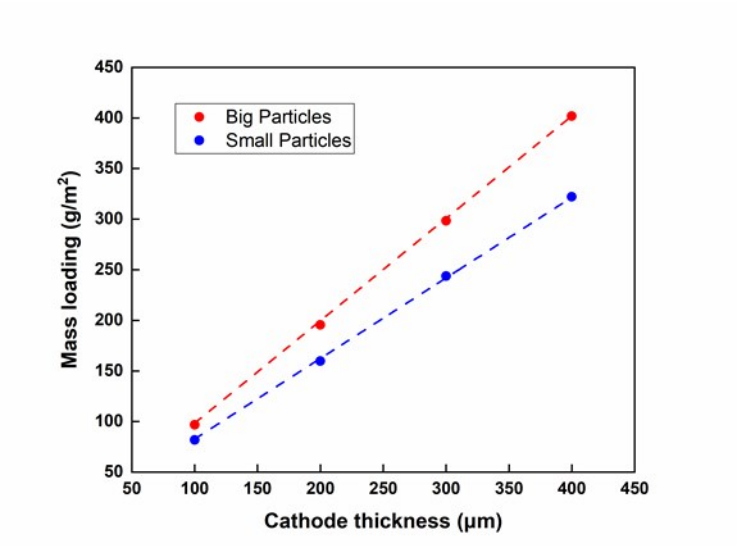

Figure S4. The mass loading as a function of electrode thickness

Impact of drying temperature on microstructure

The density for NMC811, PVDF and CB are 4.87, 1.78 and 1.95 g cm^{-3}. This makes the active material the heaviest material. A preferential binder deposition on the electrode surface was observed when the drying temperature was reduced to 60^0C. A slow drying process may provide the heavier NMC

particles enough time to migrate closer to the current collector due to gravity. On the other hand, an increase in drying temperature (80^0C) results in crack formation due to surface tension related to faster solvent evaporation. An initial drying temperature of 70^0C is found to be optimal based on SEM analysis as presented in Fig. 4.

EIS data for small and big particles

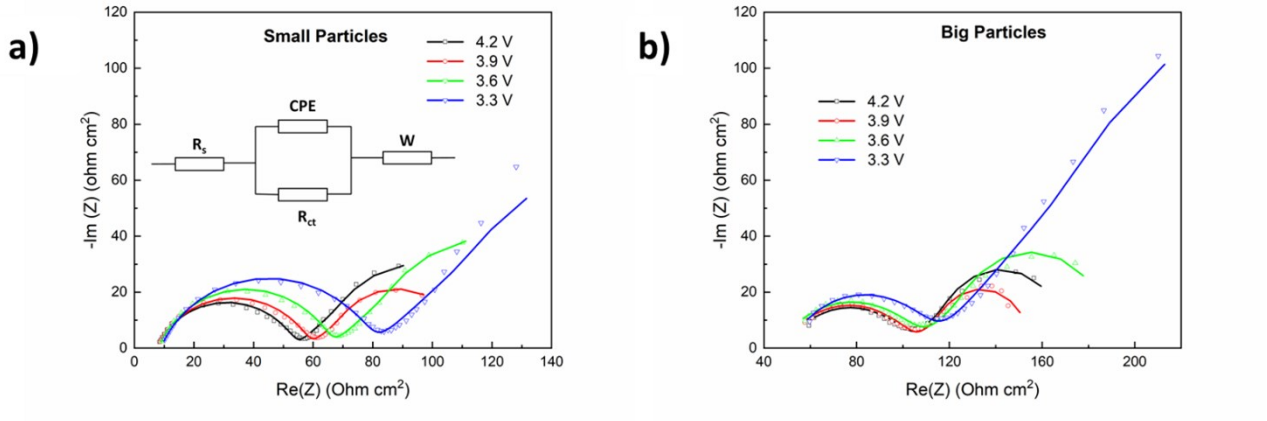

Figure S5. Nyquist plots at different cell voltages for (a) small and (b) big particles

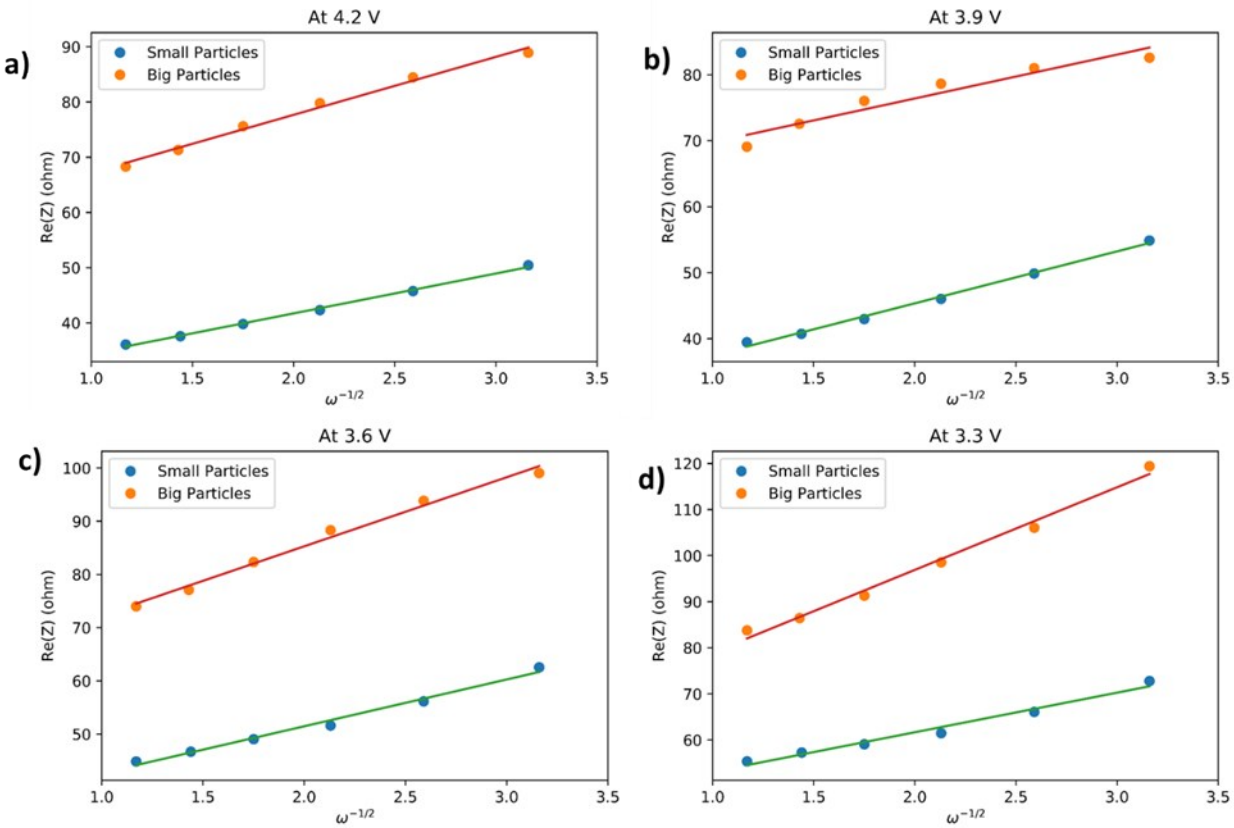

Figure S6. Linear fitting between Re(Z) and $\omega^{-1/2}$ for small and big particles at (a) 4.2V, (b) 3.9V, (c) 3.6V and (d) 3.3V