

Supplementary Data

Table S1. Characteristic particle diameter d_p of graphite material.

| | d_{10} [μm] | d_{50} [μm] | d_{90} [μm] | $(d_{90} - d_{10})/d_{50}$ [-] |
|------------|-------------------------------|-------------------------------|-------------------------------|-----------------------------------|
| Gr_flake | 10.25 | 19.55 | 36.74 | 1.36 |
| Gr15 | 11.90 | 14.85 | 18.03 | 0.41 |
| Gr20 | 11.48 | 14.45 | 17.66 | 0.43 |
| Gr25 | 11.20 | 14.34 | 17.64 | 0.45 |
| Gr30 | 10.79 | 13.75 | 16.82 | 0.44 |
| Gr_pancake | 10.73 | 15.44 | 21.42 | 0.69 |
| Gr15cc | 12.39 | 17.06 | 22.55 | 0.60 |

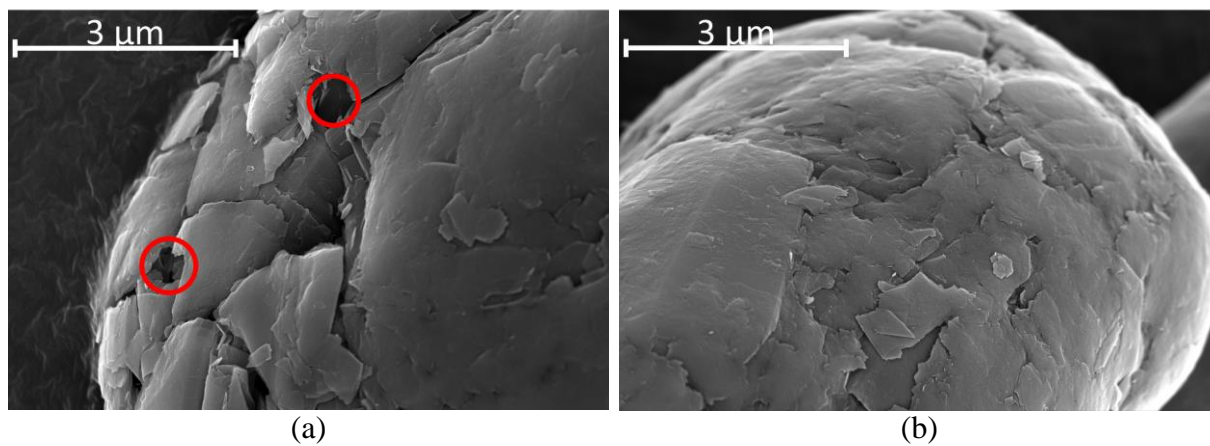


Figure S1. SEM image of surface of (a) Gr15 and (b) Gr30.

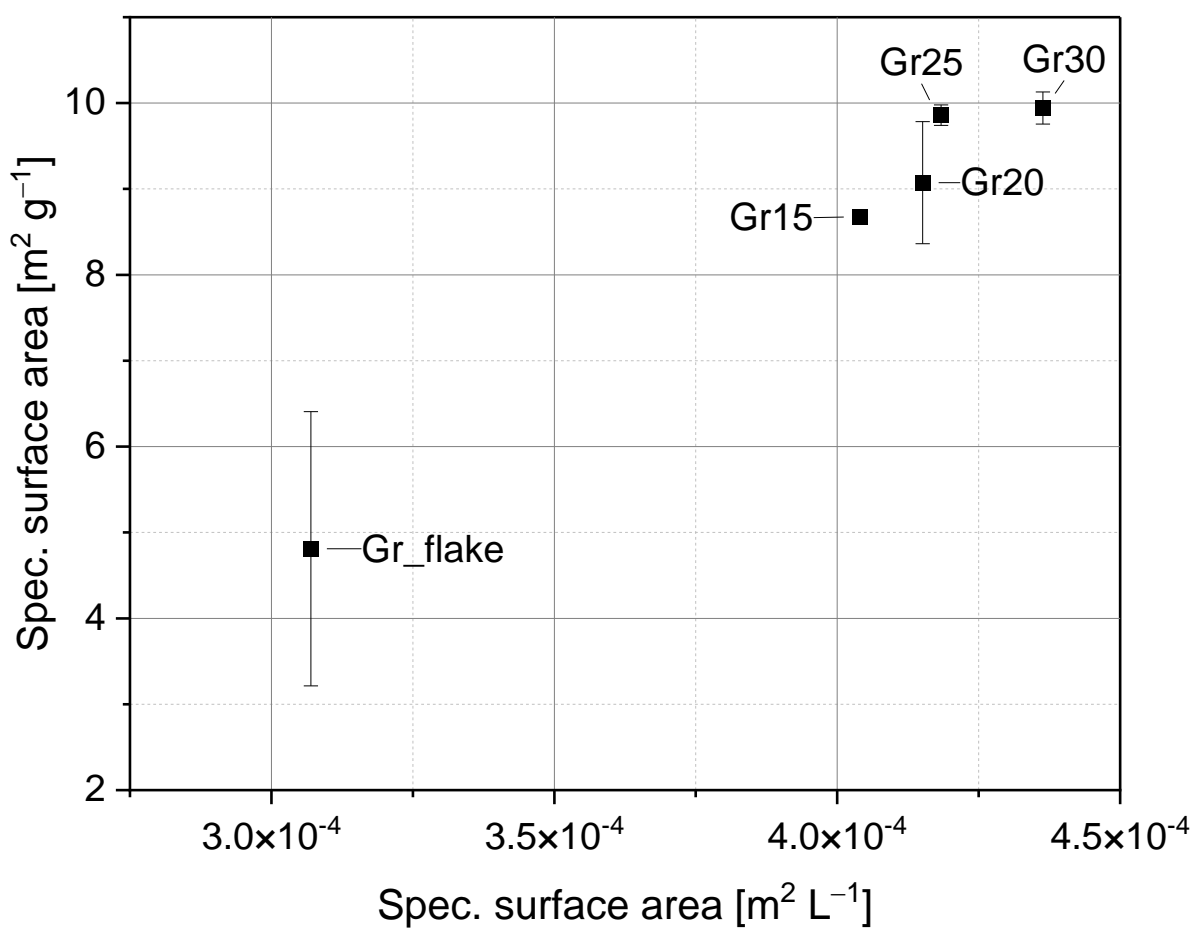


Figure S2. Link between volumetric specific surface area and mass specific surface area.

Table S1. Calendering of anodes with minimal roller gap (25 μm).

| | Initial coating density [g cm ⁻³] | Line load [N mm ⁻¹] | Coating density [g cm ⁻³] |
|------------|--|------------------------------------|--|
| Gr_flake | 0.89 | 338.0 | 2.02 |
| Gr15 | 1.19 | 323.9 | 1.92 |
| Gr20 | 1.19 | 341.3 | 1.94 |
| Gr25 | 1.16 | 352.0 | 1.86 |
| Gr30 | 1.16 | 343.3 | 1.93 |
| Gr_pancake | 1.10 | 293.2 | 1.90 |
| Gr15cc | 1.11 | 308.3 | 1.67 |

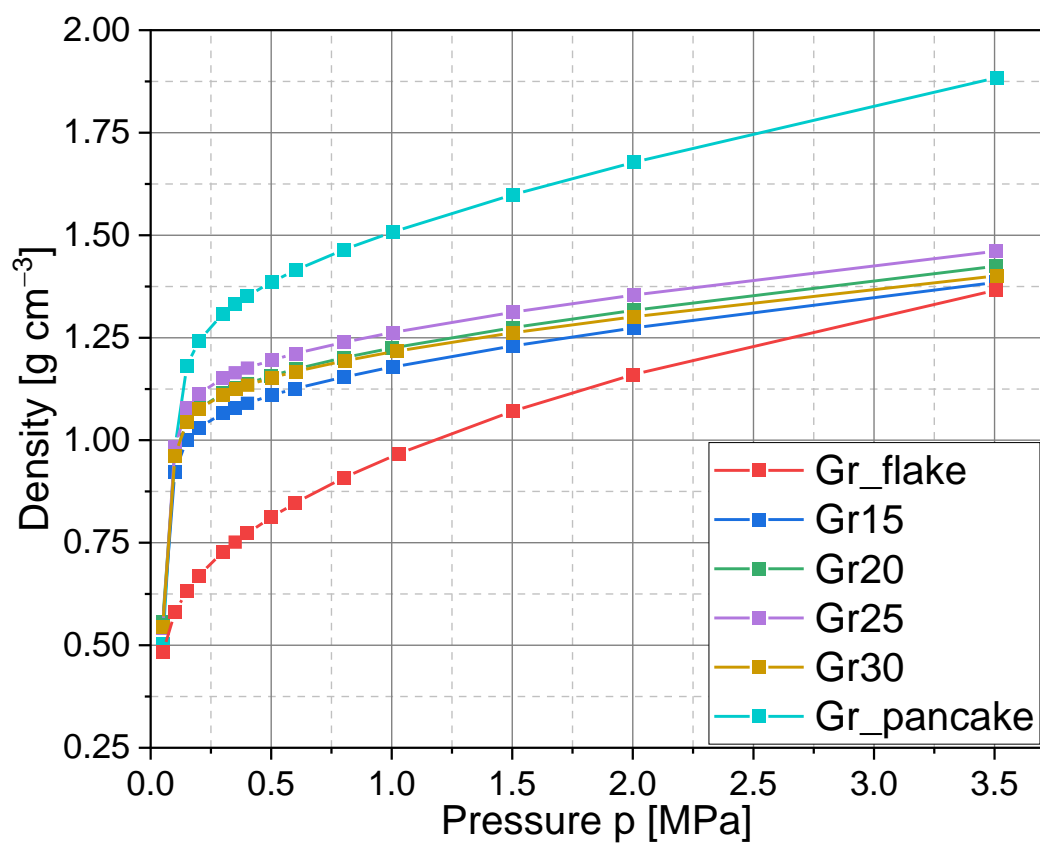


Figure S3. Density of graphite powder vs. pressure during compression.

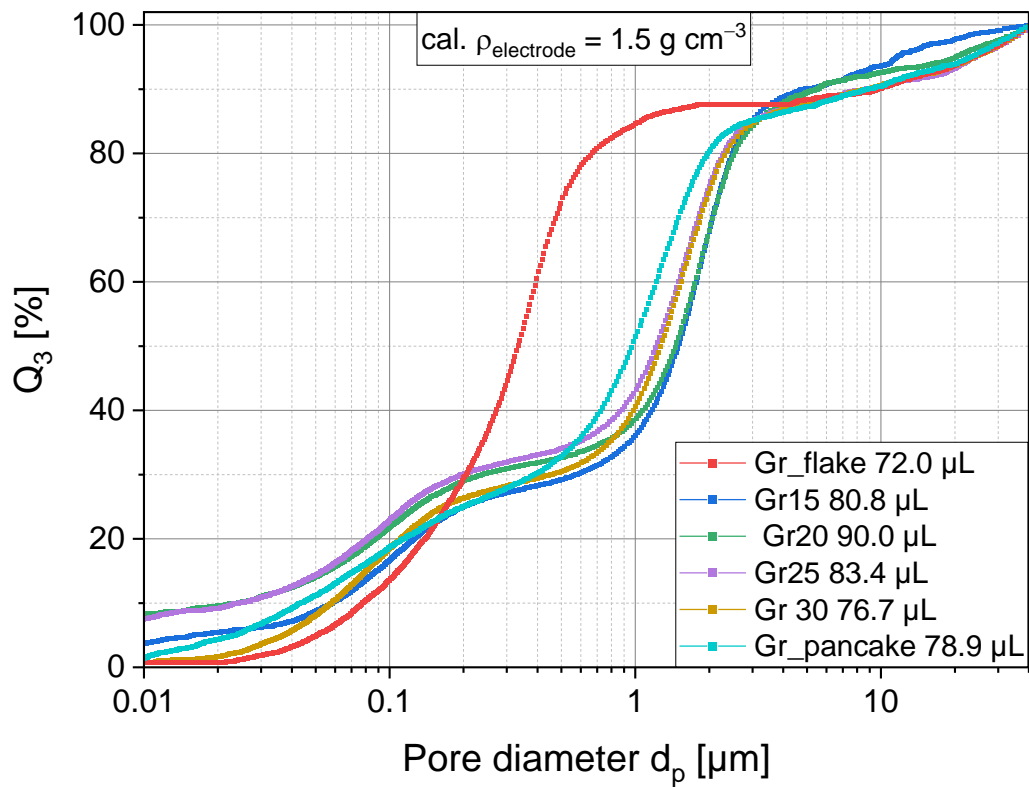


Figure S4. Total distribution of electrode porosity of different particle morphologies.

Table S2. Calendering of anodes to a density of 1.5 g cm⁻³.

| | Initial coating density [g cm ⁻³] | Line load [N mm ⁻¹] | Coating density [g cm ⁻³] |
|------------|--|------------------------------------|--|
| Gr_flake | 0.89 | 63.1 | 1.51 |
| Gr15 | 1.19 | 62.0 | 1.52 |
| Gr20 | 1.19 | 61.8 | 1.51 |
| Gr25 | 1.16 | 66.6 | 1.50 |
| Gr30 | 1.16 | 79.1 | 1.52 |
| Gr_pancake | 1.10 | 84.2 | 1.52 |
| Gr15cc | 1.11 | 160.0 | 1.48 |

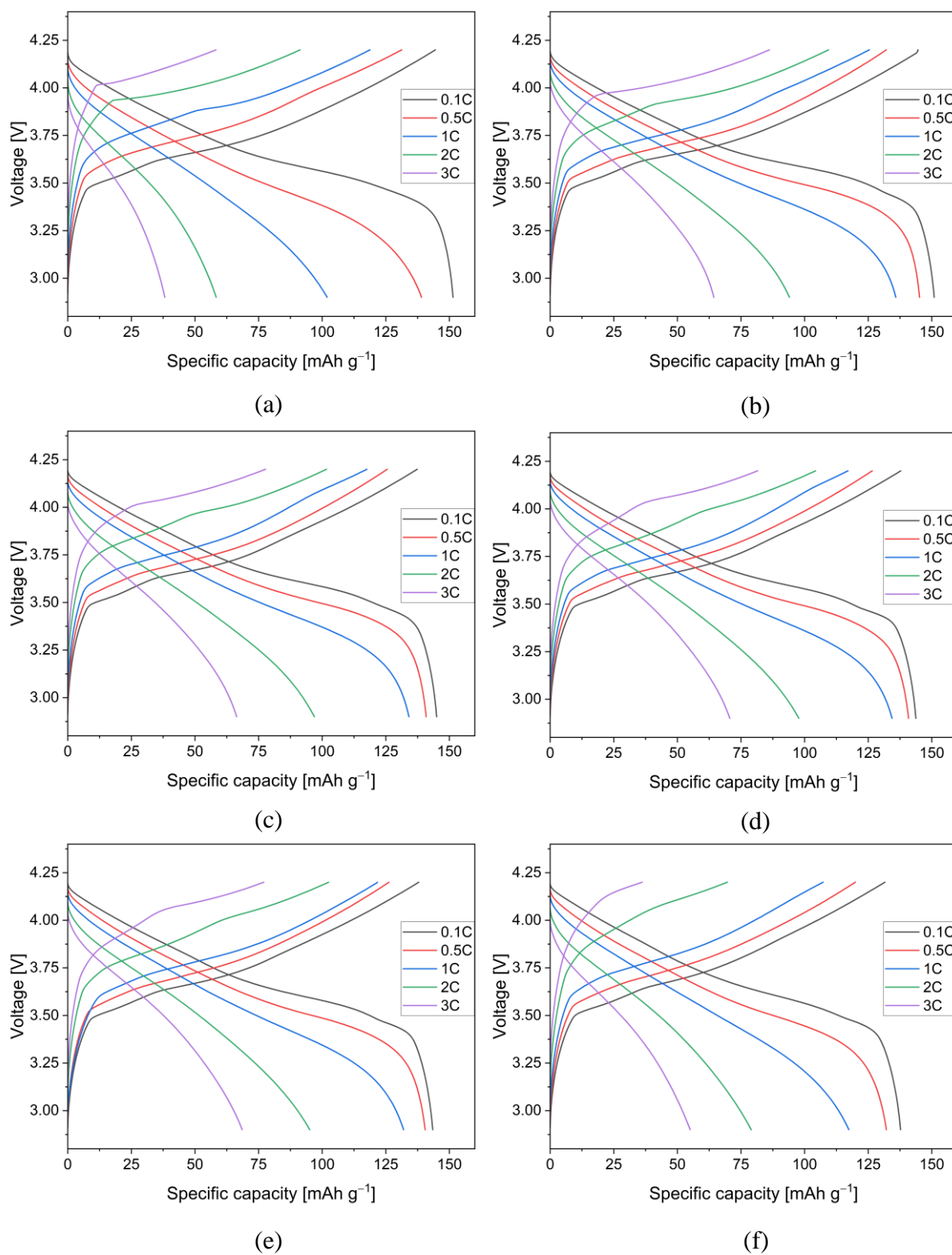


Figure S5 Galvanostatic charge/discharge voltage profiles of (a) Gr_flake; (b) Gr15; (c) Gr20; (d) Gr25; (e) Gr30; and (f) Gr_pancake composite electrodes.

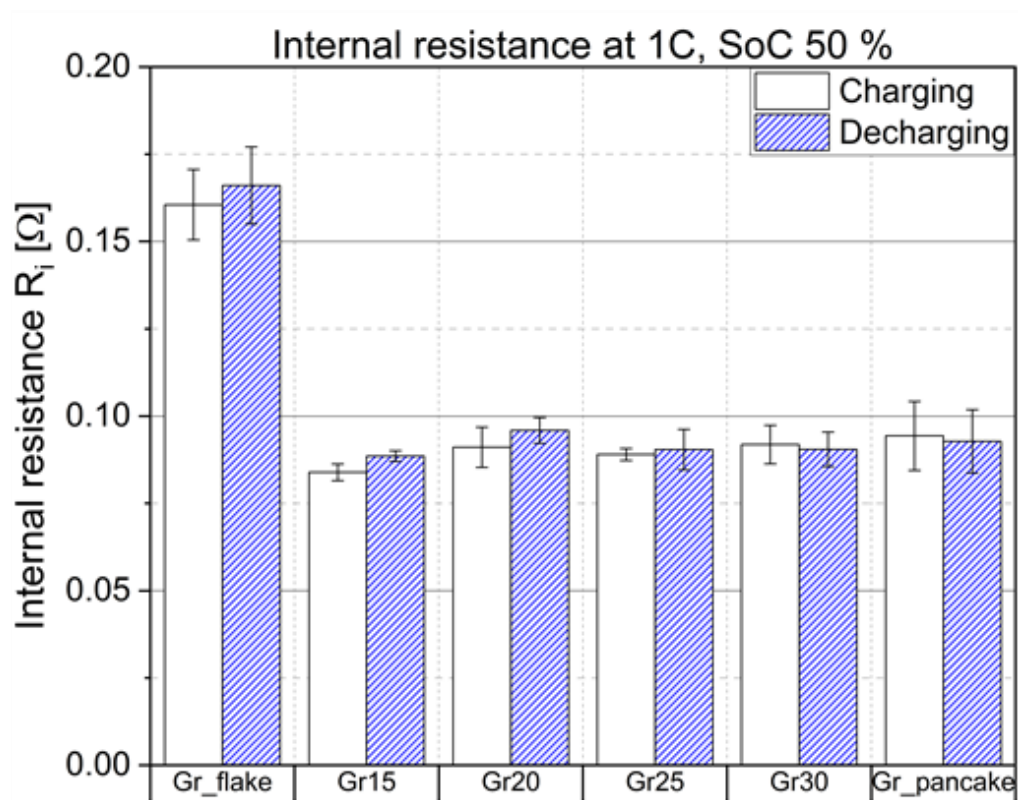


Figure S6 Internal electrical resistance during a cycle at 1C.

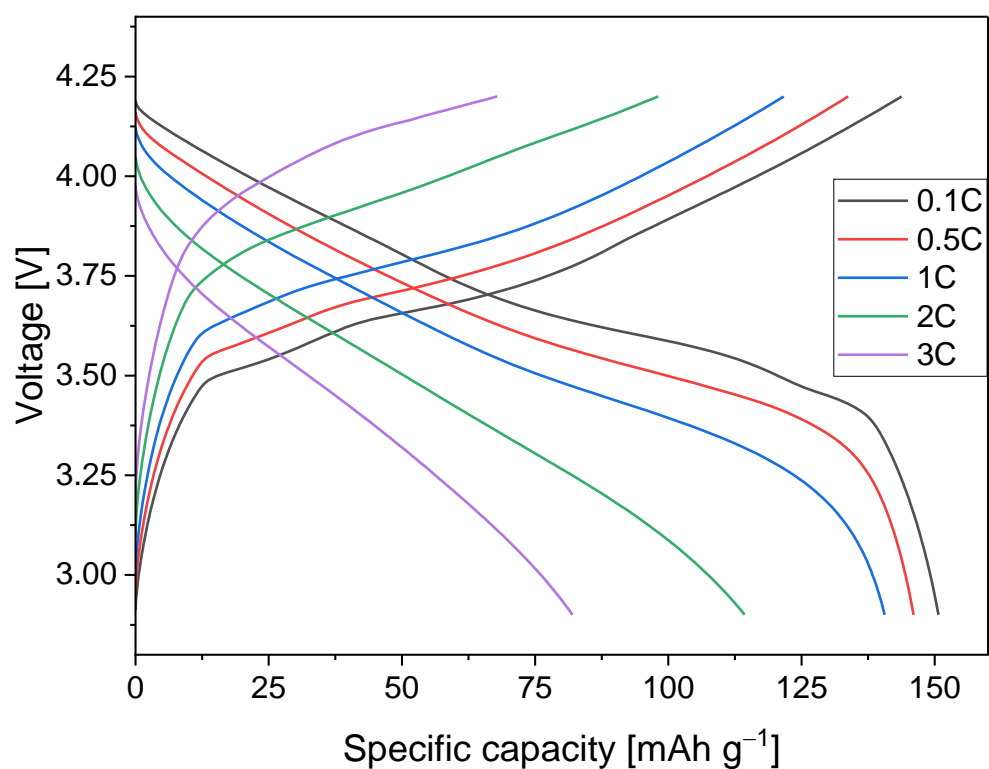


Figure S7 Galvanostatic charge/discharge voltage profiles pitch-coated spherical graphite (Gr15cc) composite electrodes.

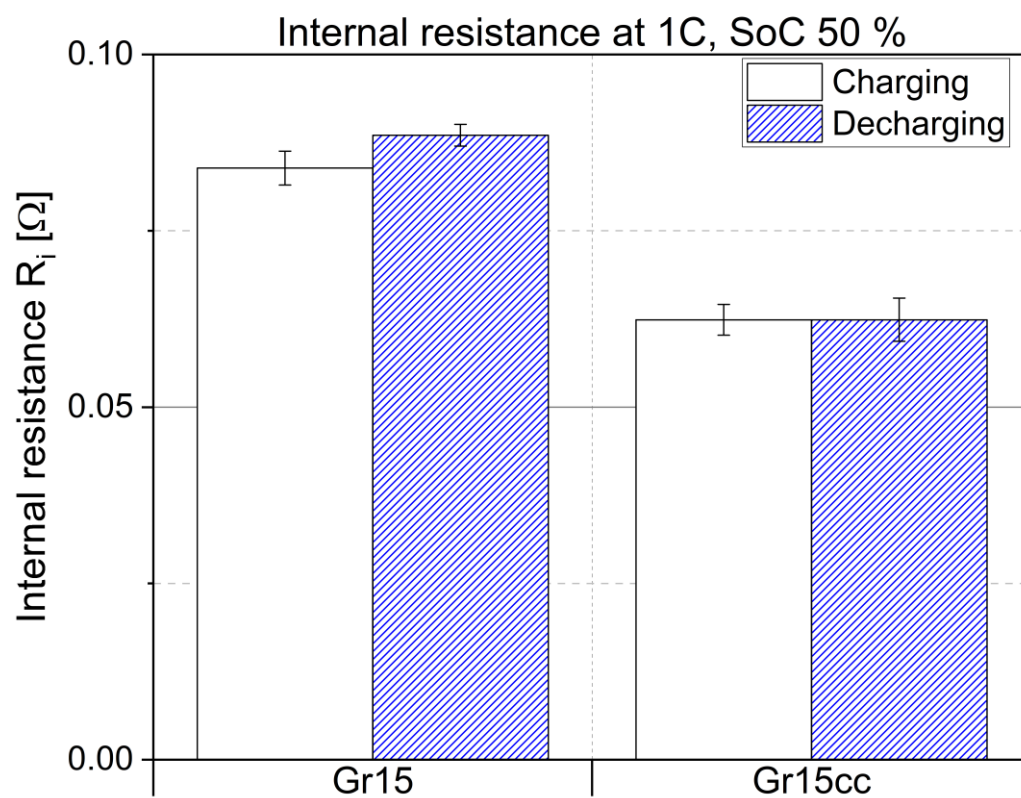


Figure S8 Internal electrical resistance during a cycle of Gr15 and Gr15cc at 1C.