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Supporting Information for

Quantifying the effect of electronic conductivity on the rate-performance of nanocomposite battery electrodes

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Electrode density

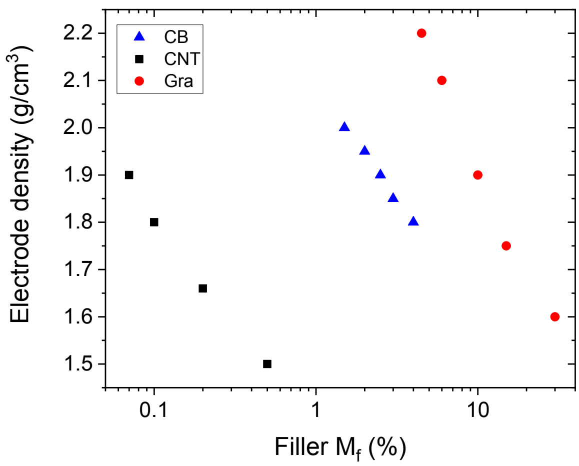


Figure S1: Electrode density versus filler content.

Galvanostatic charge discharge curves

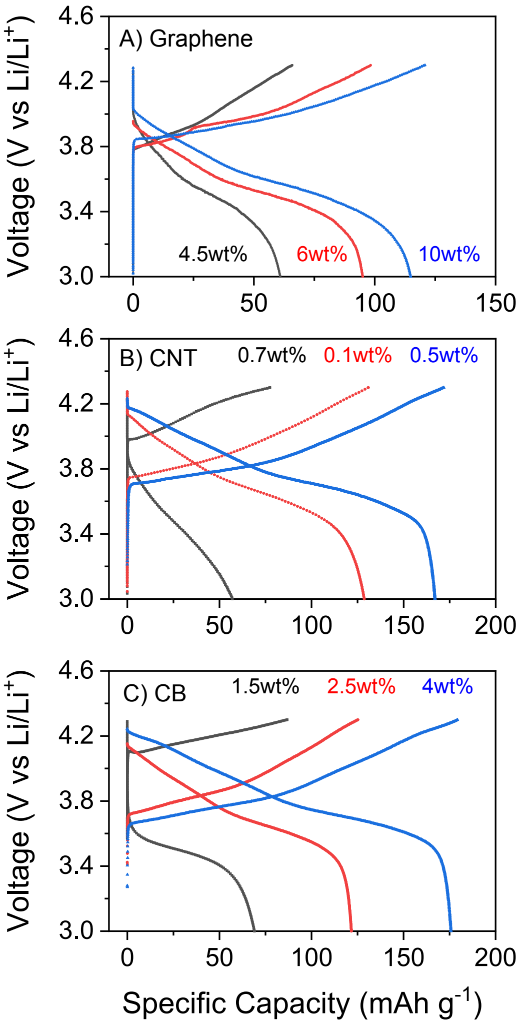


Figure S2: Second cycle charge-discharge curves for electrodes of NMC811 filled with different loadings of A) graphene, B) carbon nanotubes and C) carbon black. All measurements were performed at I/A=17 mA/g.

Calculating the curve in figure 5B

We can plot equation 6a on figure 5B as follows.

Equation 6a can be written as:

$$\frac{\tau}{L_E^2} = \frac{14Q_V}{\sigma_{OOP}} + \left[\frac{28Q_V}{2\sigma_{BL} P_E^{3/2}} + \frac{1}{D_{BL} P_E^{3/2}} + \frac{L_S}{L_E} \frac{28Q_V}{\sigma_{BL} P_S^{3/2}} + \frac{1}{L_E^2} \left(\frac{L_S^2}{D_{BL} P_S^{3/2}} + \frac{L_{AM}^2}{D_{AM}} + t_c \right) \right]$$

When plotted as τ / L_E^2 vs. Q_V / σ_{OOP} the expected slope is 14 F/mAh while the intercept is the set of terms in the square brackets. The intercept can be found when $1/\sigma_{OOP}=0$ or when $\sigma_{OOP}\rightarrow\infty$.

For illustrative purposes, we plot τ / L_E^2 vs. σ_{OOP} below. The intercept in figure 5B is the constant value of τ / L_E^2 when σ_{OOP} becomes large. To plot this, we need to estimate the relevant parameters:

$\langle Q_V \rangle=2.1\times10^8$ mAh/m^3	Found using $Q_V = \rho_E Q_M$ and averaging over all samples.
$\sigma_{BL}=0.5$ S/m	Typical for LIB electrolytes[1]
$D_{BL}=3\times10^{-10}$ m^2/s	Middle of the range for common battery electrolytes[2-3]
$P_E=0.6$	Estimated from mean electrode density
$P_S=0.4$	Typical for commercial separators[4]
$L_S=16$ µm	Celgard 2032 separator
$\langle L_E \rangle=97$ µm	Measured mean thickness
$L_{AM}=r/3\sim2$µm	Proposed relationship between L_{AM} and particle radius[5]
$D_{AM}=5\times10^{-14}$ m^2/s	Diffusivity of Li ions in NMC111[6]
$t_c=25$s	Roughly middle of the range reported by[5]

Using these parameters yields the following graph which clearly shows the limiting value to be 3.5×10^{10} s/m^2.

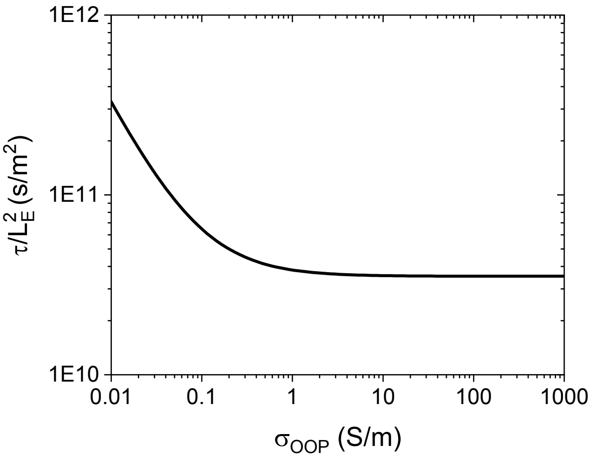
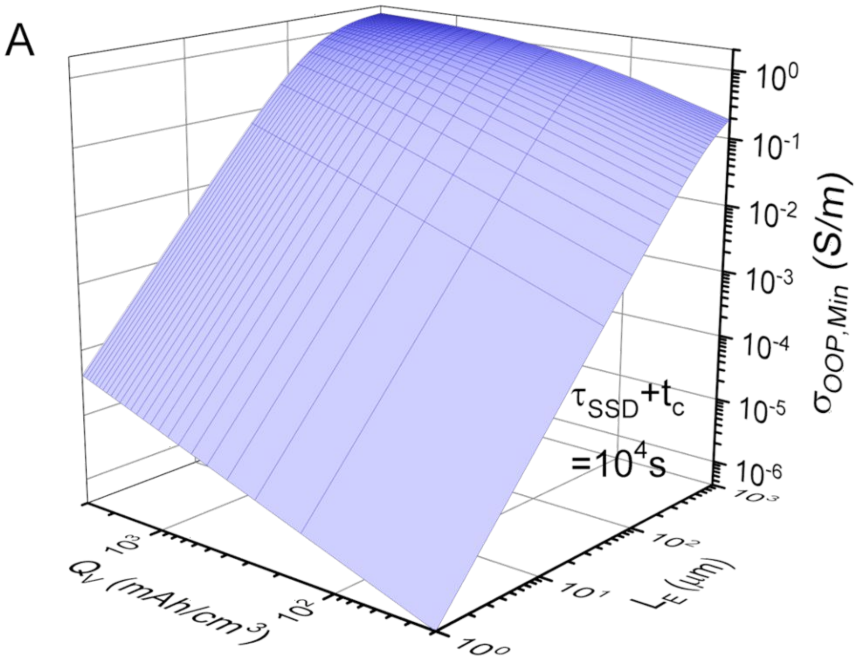


Figure S3: Calculating τ / L_E^2 versus OOP conductivity.



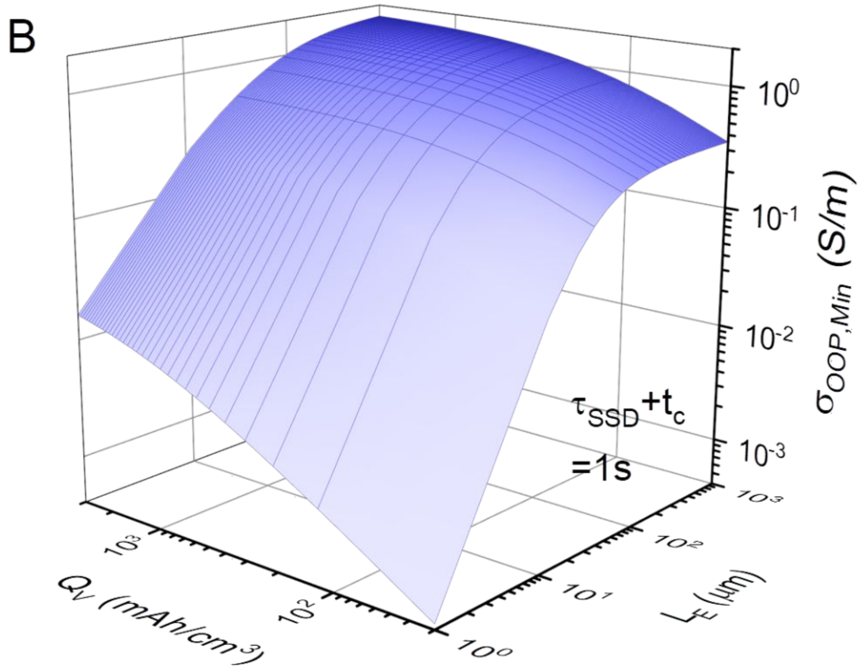


Figure S4: Critical (out-of-plane) electrode conductivity, $\sigma_{OOP,min}$, plotted as a function of electrode thickness (L_E) and low rate volumetric capacity (Q_V). The critical conductivity is that required to reduce the contribution to τ associated with the electrode resistance (first term in equation (6a)) below 10% of the sum of the other contributions to τ (i.e. the other six terms in equation (6a)). Here we calculate $\sigma_{OOP,min}$ using the following parameters: $\langle Q_V \rangle$=2.1×10^8 mAh/m^3, σ_{BL}=0.5 S/m, D_{BL}=3×10^{-10} m^2/s, P_E=0.6, P_S=0.4, L_S=16 μm, $\langle L_E \rangle$=97 μm. In A and

B, this calculation is performed for electrode materials with long (A, $\tau_{SSD}+t_c=10^4$s) and short (B, $\tau_{SSD}+t_c=1$s) combinations of solid-state diffusion and reaction times.

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