Supplementary Information

Carbon-Binder Migration: A Three-Dimensional Evaporation Model for Lithium Ion Batteries

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Figure S1. Comparison of experimental (full line) and simulated (dots) shear-viscosity curve for a slurry containing 94 wt.% of NMC, 3 wt.% of C65 and 3wt.% of PVdF. The simulation was performed by using 94 wt.% of AM and 6 wt.% of CBD. The experimental slurries were prepared and analyzed as previously reported in the experimental section of Reference S1.
**Table S2.** Force fields parameters’ value for the slurry (red) and dried electrode (blue and green) simulations. The blue values correspond to the ones reached when the AM backbone is frozen (Figure 1 in the main text), while the green ones are the values reached at the end of the simulation. The FFs parameters’ value at the end of the drying (green) were kept constant for the calendaring simulation. $d_{AM,\text{exp}}$ and $d_{CBD}$ stand for the AM and CBD diameters, respectively. During the simulation, the AM particles diameter were slightly expanded (14% in the slurry simulation and 2% in the drying one) to account for the particles overlap. For more information on this point, please refer to Reference S1. The diameter and density of the CBD particles when the AM backbone is frozen (blue values) are in between the one of the slurry and the one of the completely dried electrode (green values), and they depend on the CBD type considered (CBD1, CBD2 or CBD3) and to the RSFs applied, which is the reason because they are not explicitly reported in the Table below.

|               | AM                              | CBD               |
|---------------|---------------------------------|-------------------|
| $\epsilon$ [pg $\mu m^2 \mu s^{-2}$] | $[0.01, 100, 500]^* d_{AM}$ | $[1, 150, 650]^* d_{CBD}$ |
| $\sigma$ [$\mu m$]                   | $[0.95, 0.93, 0.93]^* d_{AM}$ | 1.1, 1.1, 1.1 |
| $r_c$ [$\mu m$]                      | $[1.14, 2, 2]^* d_{AM}$       | 2.2, 2.2, 2.2   |
| $d$ [$\mu m$]                        | $[1.14, 1.044, 1.02]^* d_{AM}$ | 5.7, 1.3        |
| $\rho$ [g $cm^{-3}$]                 | 4.65, 4.65, 4.65             | 0.008, 0.95     |
| $k_n$ [pg $\mu m^{-1} \mu s^{-2}$] | 7, 400, 950                  |                   |
| $\gamma_n$ [$\mu m^{-1} \mu s^{-1}$] | 45, 16, 10                   |                   |
| $\nu$                                    | 0.15, 0.3, 0.3               |                   |
| $X_u$                                    | 0.015, 12, 15                |                   |

**S2. Approaches to implement the selected relative shrinking factor**

During the model development, it was observed that applying the relative shrinking factors (RSFs) selected from the beginning of the simulation did not bring to the observed experimental trend, i.e. additives migration occurring in a specific time range and not all along the drying step. Therefore,
instead of applying the selected RSFs from the beginning of the drying, all the CBD particles (the ones in the top region, the ones in the middle region and the ones in the bottom – Figure 2 in the main text) start from a RSF of 1. However, the RSF of the bottom region stick to 1 all along the drying, while the RSF of the bottom and top regions increase from 1 to the selected RSF (as, for instance, 1.2 / 1.8 or 1.3 / 2.2, to name two cases discussed in the main text). The rationality behind this is the fact that the slurry starts the drying from a rather homogeneous condition (at the entrance of the oven the slurry is still at RT), while heterogeneities (for instance in terms of temperature) are developed along the slurry film during the drying step.

The utilization of the procedure discussed above adds an extra degree of freedom to the simulation, i.e. which function to use to go from 1 (beginning of the simulation) to the selected RSFs (at the end of the simulation) for RSF2 (middle region) and RSF3 (top region). Different functions were tested and some of them are reported in Figure S2. On the one side, applying a constant RSFs (pink curve) from the beginning of the drying or a function that lead to a very fast increase of their value (as an exponential function, red curve) lead to additive migration from the beginning of the simulation, which is not what was experimentally observed. On the other side, applying less steep functions (as quadratic, linear, square root, etc.) delay the additives migration (here CBD), as expected from previous experimental findings. All the results showed in the main text were obtained using a linear function (blue curve in Figure S2), which was found to be a good compromise in between all the functions tested.

![Figure S2. Comparison of different functions used to go from RSFs of 1 to the selected values for the case of the drying rate 1.3 / 2.2. Other functions compared to the ones showed here were tested (as quadratic or square cube), showing similar behaviors to the linear/cubic/square root ones. Then, they were not reported for the sake of the graph readability.](image-url)
S3. References

(S1) Lombardo, T.; Hoock, J.; Primo, E.; Ngandjong, C.; Duquesnoy, M.; Franco, A. A. Accelerated Optimization Methods for Force-Field Parametrization in Battery Electrode Manufacturing Modeling. *Batter. Supercaps* **2020**. https://doi.org/10.1002/batt.202000049.