

## Supporting Information

### Evaporation Induced Nanoparticle – Binder Interaction in Electrode Film Formation

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## Relation between Solvent Chemical Potential and Evaporation Kinetics

The chemical potential affects the energy barrier for converting a solvent cell in the liquid phase to the gas phase. The more negative chemical potential indicates a lower energy barrier for the liquid-to-gas phase transition. Therefore, the more negative chemical potential represent a faster evaporation rate at a constant drying temperature. In the pure-liquid system, the critical solvent chemical potential  $\tilde{\mu}$  for liquid/gas coexistence is  $-2$ . When solid-phase components are added into the system, the strong attractive interaction between the nanoparticle and solvent tries to keep the solvent in the liquid phase. In this case, the critical chemical potential for the phase transition can be roughly estimated by the mean-field approach<sup>1</sup>:

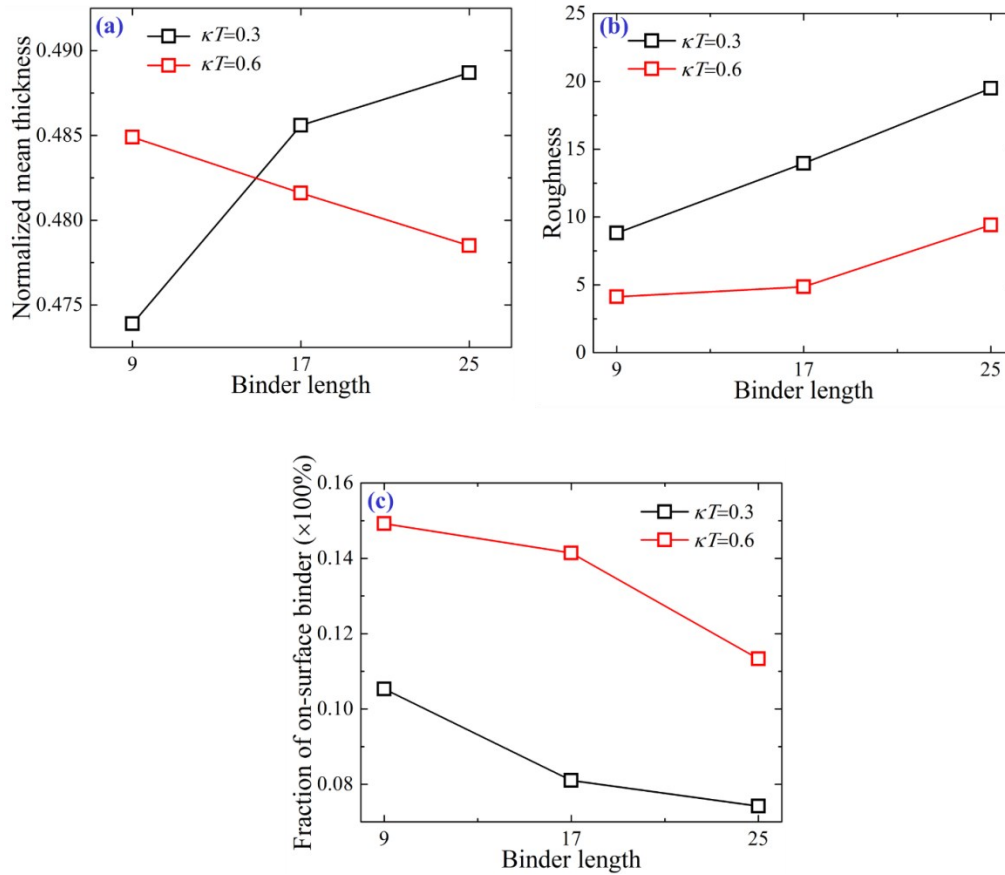
$$\tilde{\mu} = -2 - (\epsilon_{nl} - 1) \times \Phi_{tot}. \quad (\text{S1})$$

In the current study, the solid-solvent interaction  $\epsilon_{nl}$  is predefined to 1.4, and the total volume fraction of solid phases  $\Phi_{tot}$  are 43.6%. The estimated critical chemical potential is  $-2.17$  in this study, which indicates that the evaporation is not energetically and kinetically favored if the solvent chemical potential is higher than  $-2.2$ .

## Reproducibility of KMC Simulations

Three different binder lengths ( $l=9, 17, 25$ ) are chosen to demonstrate the reproducibility of the current CGLG-KMC model. For each case, five different random initial structures are generated for each binder length. All initial structures are dried at both  $\kappa T = 0.3$  and  $\kappa T = 0.6$ . The average thickness, roughness and fraction of on-surface binder for each combination of binder length and drying temperature are calculated and plotted in Figure S1. It can be found that the average thickness increases as the binder length increases at the low drying temperature; oppositely, the average thickness decreases as the binder length increases at the high drying temperature (Figure

S1(a)). The high drying temperature can produce flatter films as shown in Figure S1(b). The binder distribution is the key factor determining the mechanical stability of the electrode film. Figure S1(c) clearly demonstrates that the low drying temperature and long binder are helpful for inhibiting binder migration to the top surface of electrode films. All these results agree well with findings discussed in the main text, which can strongly prove the reproducibility of the current model. In KMC simulations, systems always move to the same final state which has minimum Hamiltonian if interactions energies and temperature are fixed.



**Figure S1.** Reproducibility of CGLG-KMC model. For each case, five equilibrium initial structures are generated and the averaged (a) normalized thickness, (b) roughness and (c) fraction of on-surface binder are calculated based on final structures.

## Reference

1. I. Vancea, U. Thiele, E. Pauliac-Vaujour, A. Stannard, C. Martin, M. Blunt and P. Moriarty, *Physical Review E*, 2008, 78, 041601.