Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.

Supporting Information

Mechanistic understanding of intergranular cracking in NCM cathode material: Mesoscale simulation with three-dimensional microstructure

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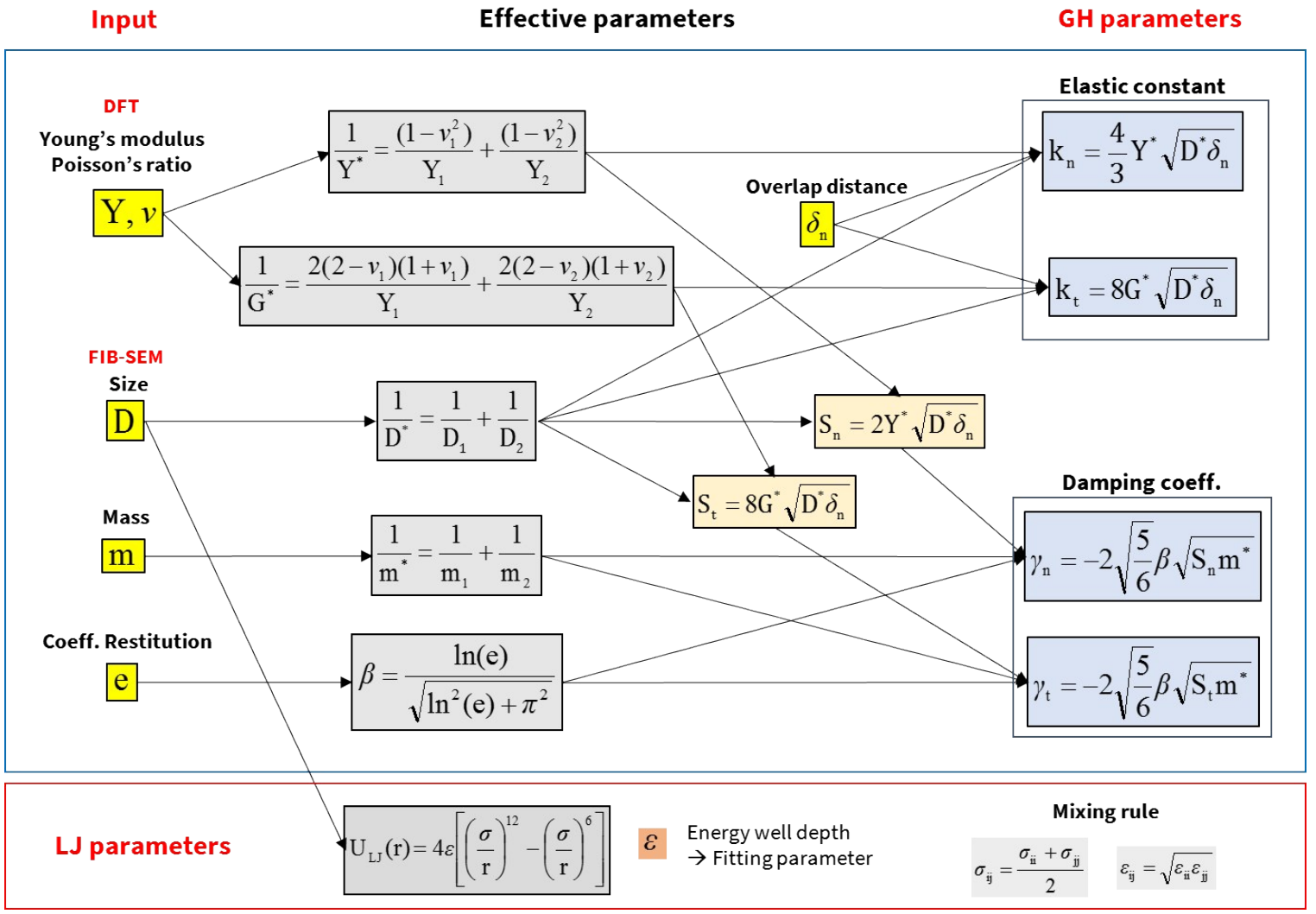

Figure S.1. Derivation of input parameters for LJ-SF potential and GH model

Derivation process for simulation parameters

For LJ-SF parameters, the first step is to obtain the particle size distribution from the actual NCM image. We note that since we implement two models (LJ-SF + GH), we use two notations for the particle size (D for the GH and σ for LJ-SF) for distinguish the model. From the SEM image, we can determine the attractive energy ε, σ (or D), and the cutoff distance r_c. To define the interaction between particles with different sizes, the mixing rule is implemented. In addition, we chose the range of ε which can represent the Young's modulus value of NCM. The weighting factor of 1.2 for the cutoff distance is arbitrary chosen.

For GH model, the particle size D is already obtained from the procedure above. Then it is important to determine the elastic constant of K_n and K_t and the damping coefficient of R_n and R_t. First for the elastic constant, since the K is the function of effective parameters of Young's modulus (Y*), shear modulus (G*), size (D*), and the overlap distance (δ), it is important to obtain the actual Y and Poisson's ratio (v) from the DFT calculations. [1] In addition, δ is arbitrarily chosen as 25 nm, which is 1/10 of the average particle diameter. For the damping coefficient, it is a function of coefficient of restitution (e), effective mass (m*), and the effective damping parameters of S_n and S_t. We assume that the coefficient of restitution (e) is almost fully-elastic (0.95) because the amount of the dissipative energy after the particle collision is not critical concern for the current work. The mass of NCM is obtained from the density (p_p) of NCM from previous DFT calculation by multiplying it with the volume. Finally, S_n and S_t is the function of Y*, G*, D*, and δ, which are already described.

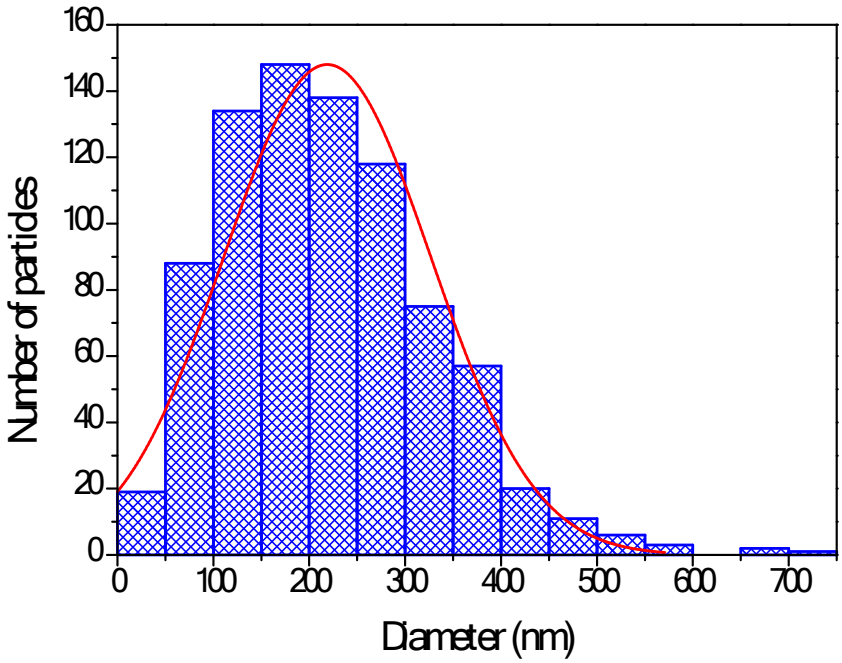

Figure S.2. The size distribution of primary particles from the FIB-SEM image

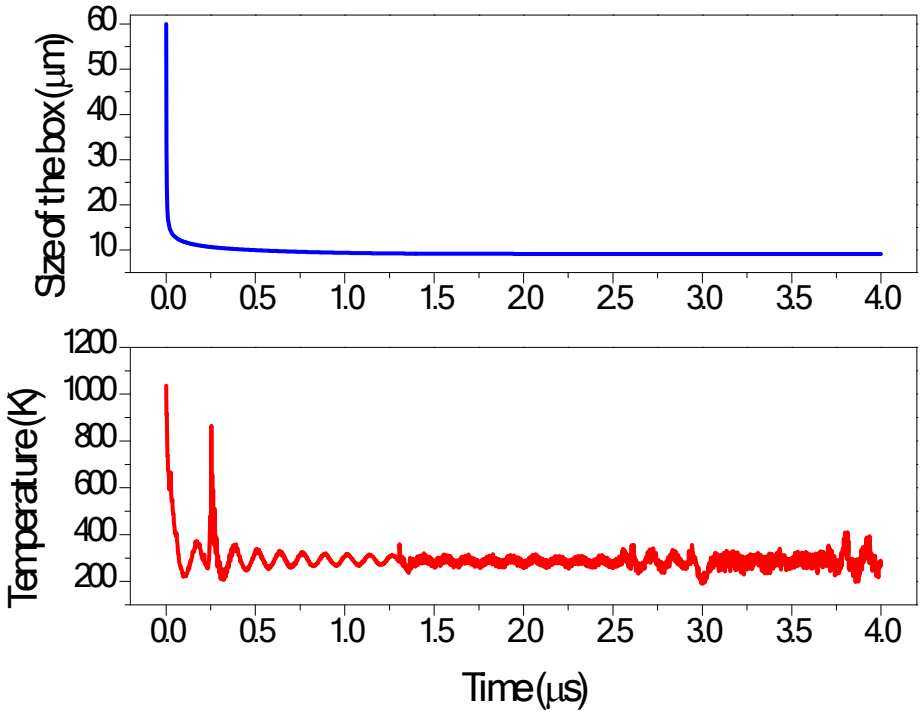

Figure S.3. Simulation output during NPT simulation for (a) the size of the simulation box and (b) the temperature.

Epsilon (ε) [pg/μm·μs^2]	10	10^2	10^3	10^4	10^5	10^6
Max. Stress [MPa]	0.83	6.11	46.09	328.84	2540.95	22005.85
Stress at 0.60 [MPa]	0.15	1.16	9.00	90.88	1212.37	12718.53
Reduction [%]	18.46	19.04	19.54	27.64	47.71	57.80

Table S.I. The percent reduction of the stress at strain of 0.6 after maximum stress value during tensile test for the structure with ε =1e1 to 1e6 pg/μm·μs^2.

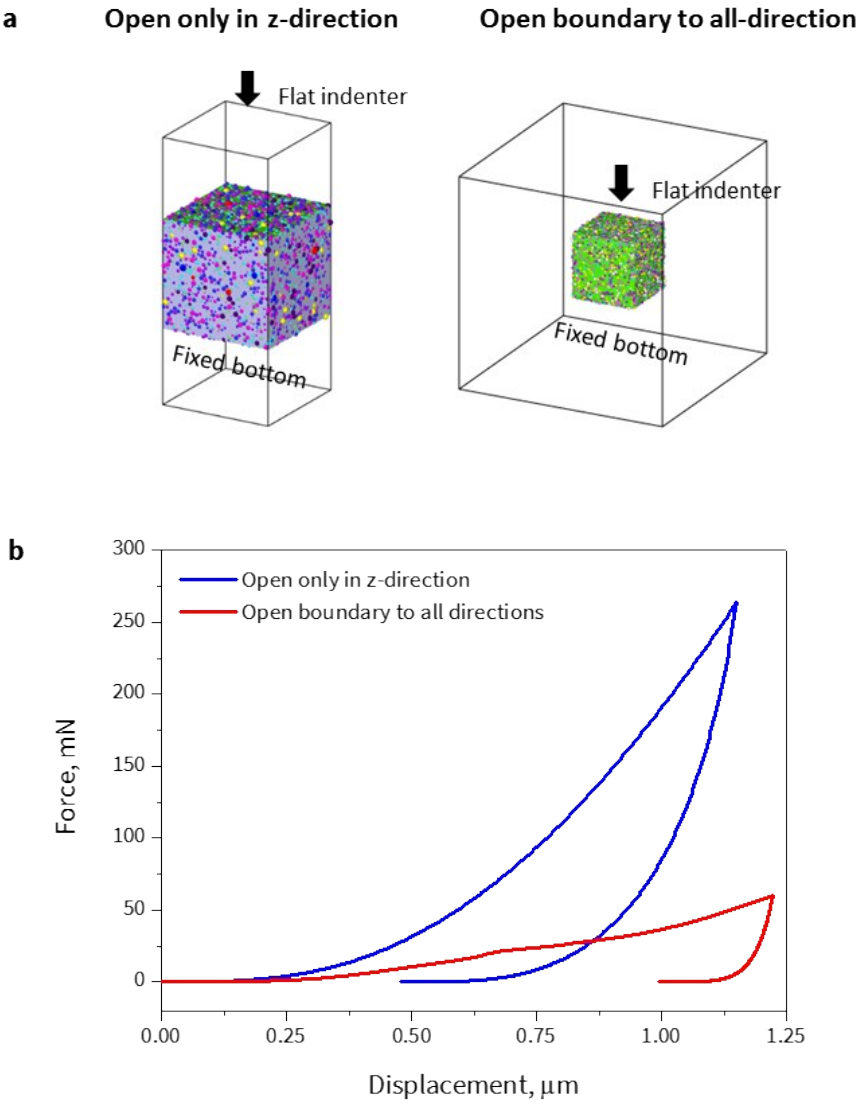

Figure S.4. (a) The boundary condition for NCM structure for nano-indentation test and (b) Force vs displacement curve for each case

References

1. Min, K. & Cho, E. Intrinsic origin of intra-granular cracking in Ni-rich layered oxide cathode materials. *Phys. Chem. Chem. Phys.* **20,** 9045–9052 (2018).