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Supplementary Information

Alleviation of concentration polarization using density-controlled duplex electrode architecture to improve quick-charging performance of lithium-ion batteries

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Experimental

1 Electrode fabrication and pouch-cell characterization

The positive electrode was fabricated using lithium nickel manganese cobalt oxide ($LiNi_{0.8}Co_{0.1}Mn_{0.1}O_2$, NCM811, EcoPro BM) with carbon black (Super P, Timcal) and poly(vinylidene fluoride) as binders at a weight ratio of 95:2.5:2.5. The resulting powder was mixed and dispersed using *N*-methyl-2-pyrrolidone (Sigma–Aldrich). Subsequently, 15-μm Al foil was coated with the slurry. The coated electrode underwent 12-h heat treatment at 120 °C in vacuum after being pre-dried at 80 °C for 5 h. SiO (DMSO, Daejoo Electronic Materials), flake graphite (SFG-6, Timcal), and a poly(acrylic acid)/poly(vinyl alcohol) (PAA, Sigma–Aldrich, Mw = 250000/PVA, Sigma–Aldrich, Mw = 19000–23000; 9:1 ratio) binder in deionized water with a weight ratio of 7:2:1 ratio were employed to fabricate the negative electrode. This mixture was then applied to 20-μm-thick Cu foil, and the product was vacuum-dried for 12 h at 80 °C. After the first half of the electrode mass was casted, an identical mass of the manufactured slurry was casted onto the bottom layer. The areal mass of electrodes was identical for single and double layered electrodes. The electrode was subsequently dried in a vacuum oven for 1 h at 150 °C to crosslink the PVA and PAA binders. A pouch-type cell was constructed by incorporating a PE (NH616, Asahi Kasei) separator and negative electrode. The cell had an N/P ratio of 1.04 and a negative electrode charge (delithiation) areal capacity of 4.16 mA h cm^{-2}. After thermal sealing, the cells were filled with electrolyte (Dongwha Electrolyte) and aged at room temperature for 24 h before electrochemical characterization.

2 Electrode analysis

The horizontal load of the casted electrode on the current collector was measured using a surface and interfacial cutting analysis system (SAICAS, Daipla Wintes, SAICAS EN-EX). The velocity of the diamond blade was 4.0 and 0.4 μm s^{-1} in the horizontal and vertical directions, respectively. The morphology of the cycled electrode was analyzed using scanning electron microscopy (SEM, JEOL JSM-IT200) in a dry room. The porosity of the fabricated electrode was investigated via mercury porosimetry (Autopore 9605, Micromeritics), and X-ray microscopy (XRM) was used to construct three-dimensional images of the pore and active-material distributions in the fabricated electrode.

3 Electrochemical modeling

For computational studies, a two-dimensional Li-ion battery model was constructed using COMSOL Multiphysics software. To predict the electrochemical behavior, a DLE with different interparticle pore sizes was considered (Figure SX). The state-of-charge of the cell was assumed to be 30% for predicting the Li insertion current density at the interface. The charge transport in the electrolyte domain can be expressed as

$$i_l = -\sigma_l \nabla \phi_l + \frac{2\sigma_l RT}{F}\left(1 + \frac{\partial \ln f}{\partial \ln c_l}\right)\left(1 - t_{Li^+}\right)\nabla \ln c_l,$$

where i_l represents the current density in the electrolyte domain, σ_l represents the ionic conductivity of the electrolyte, ϕ_l represents the electrolyte potential, R is the molar gas constant, T represents the absolute temperature, F is the Faraday constant, f is the activity coefficient for the salt, and c_l represents the Li-ion concentration in the electrolyte. The local current density (i_{loc}) for the interfacial reaction between the electrolyte and active materials was defined as

$$i_{loc} = i_0\left(\exp\left(\frac{\alpha F\eta}{RT}\right) - \exp\left(\frac{(1-\alpha)F\eta}{RT}\right)\right),$$

where i_0 represents the exchange current density, α is the anodic transfer coefficient, and η represents the overpotential.

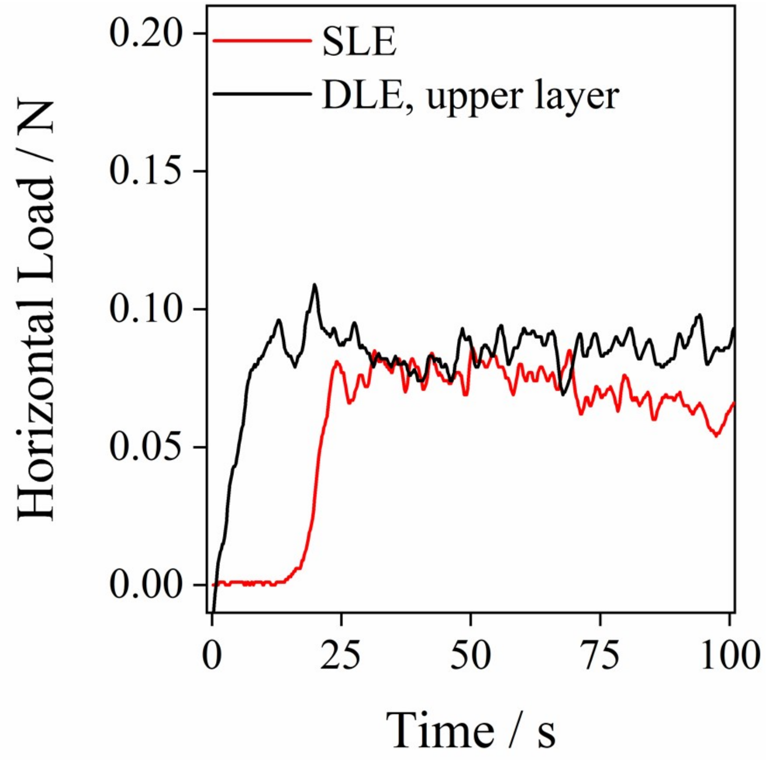


Figure S1. Horizontal load obtained from the SLE and DLE, respectively.

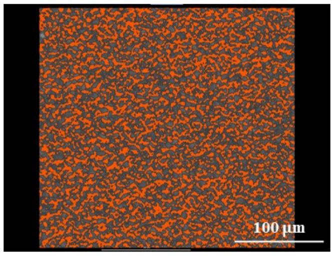
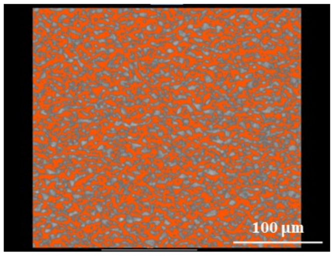
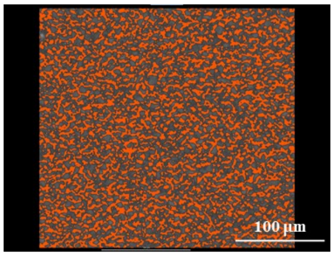
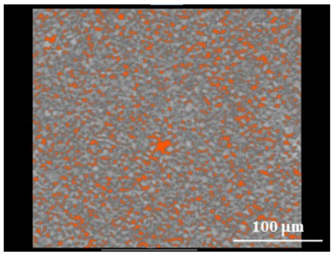
	SLE	**DLE**
Upper		
Bottom		

Figure S2. Layer-by-layer XRM images obtained from SLE and DLE structure, respectively

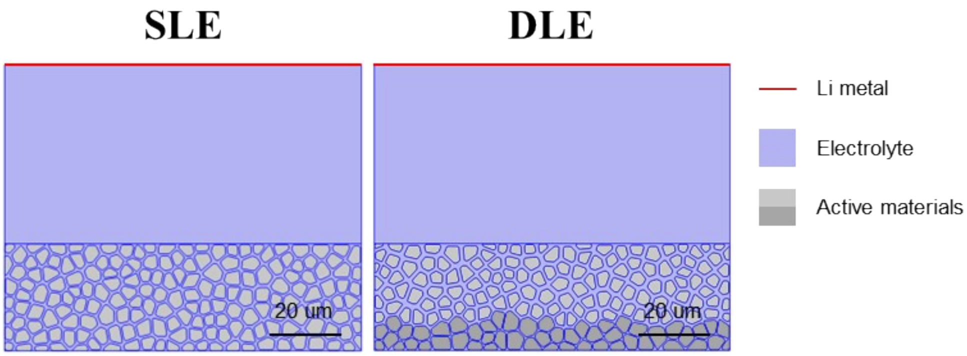


Figure S3. Half-cell geometry for Li-ion modeling with SLE and DLE.

Table S1. Used parameters for electrochemical modeling

	R_{ion} / Ω	A / cm^2	κ / S cm^{-1}	d / cm	ε	Tortuosity
SLE	6.52	1.1304	0.0073	0.004142	0.30896	4.01
DLE	4.00			0.004356	0.327362	2.48

R_{ion} = $ionic\ resistance$

ε = $porosity$

d = $thickness$ of electrode

A = $cross\text{–}sectional\ area$

κ = $conductivity\ of\ electrolyte$