



Supplementary Material

# Ultrafast-Laser Micro-Structuring of $\text{LiNi}_{0.8}\text{Mn}_{0.1}\text{Co}_{0.1}\text{O}_2$ Cathode for High-Rate Capability of Three-Dimensional Li-ion Batteries

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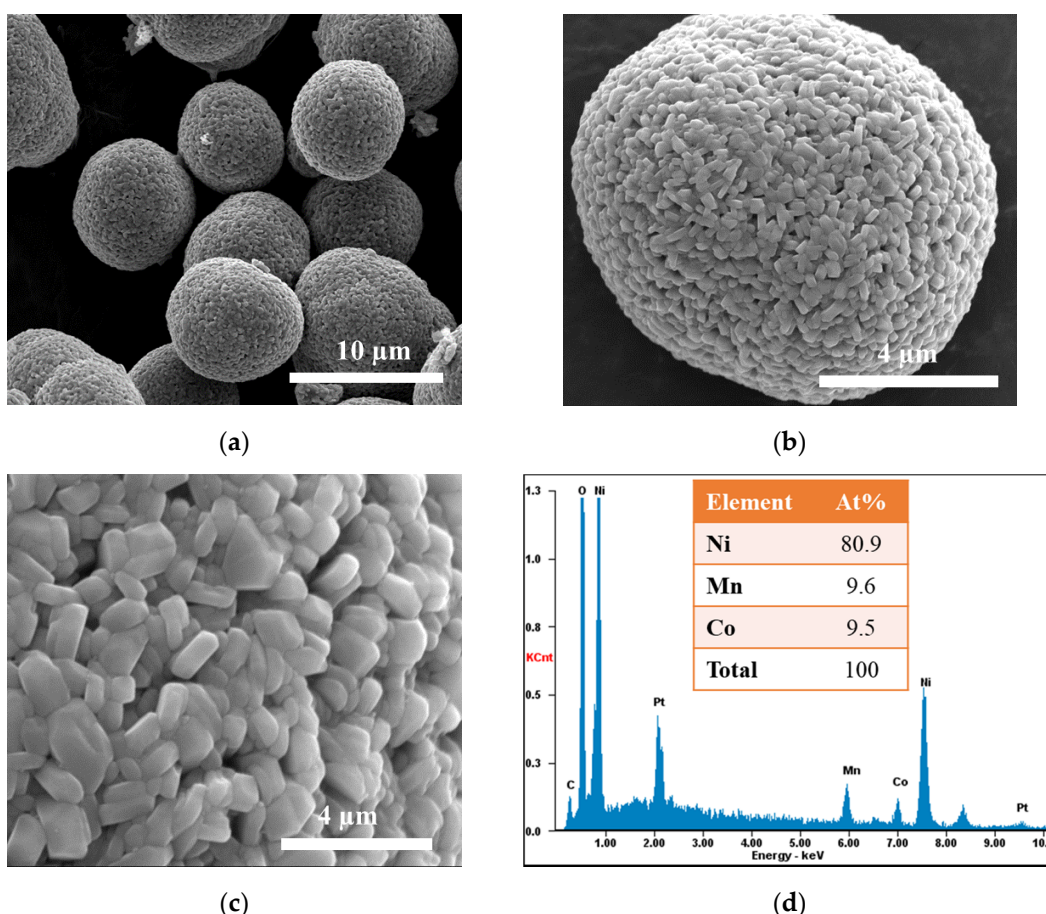
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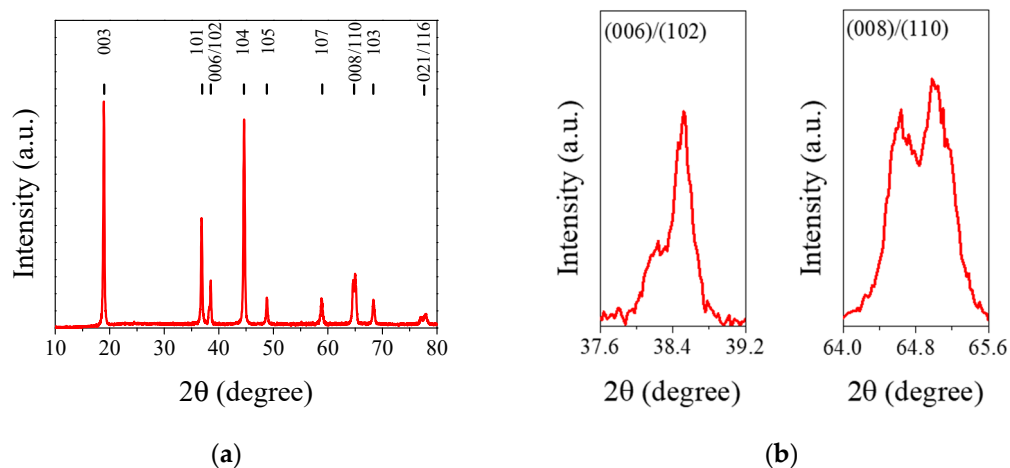
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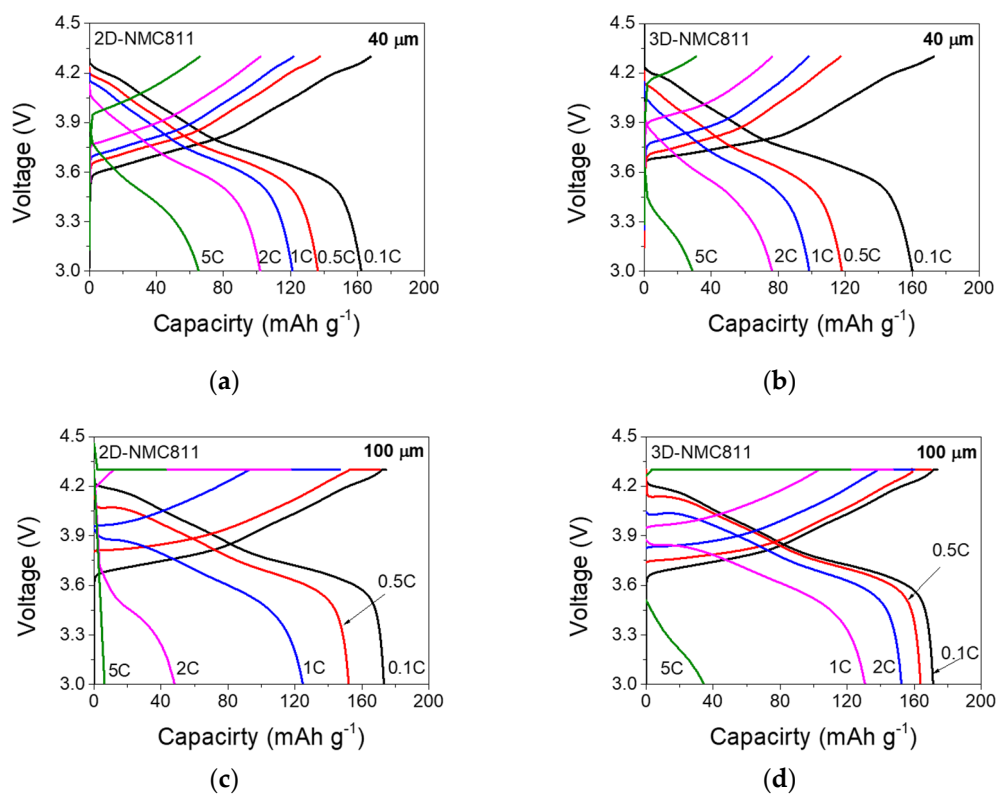
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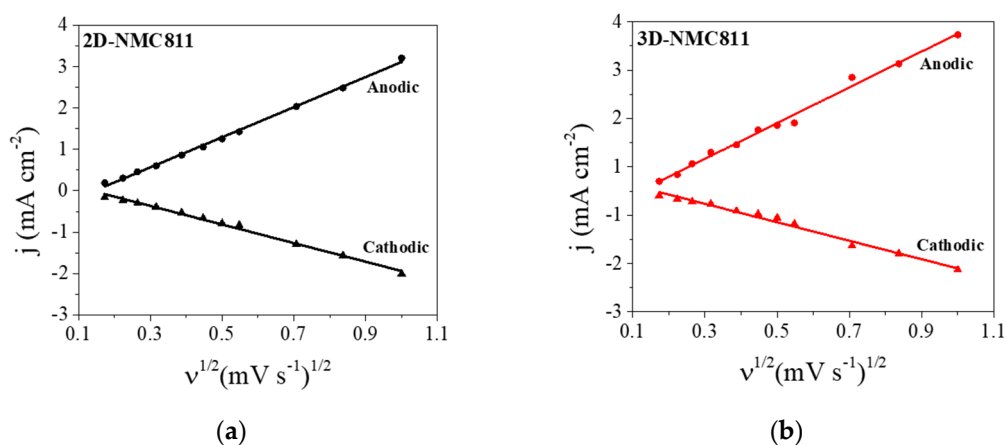
**Figure S1.** Morphology of NMC811 particles. ((a, b, c) SEM images of NMC811 particles and (d) EDX spectrum and element composition of selected area from SEM image (Figure c).



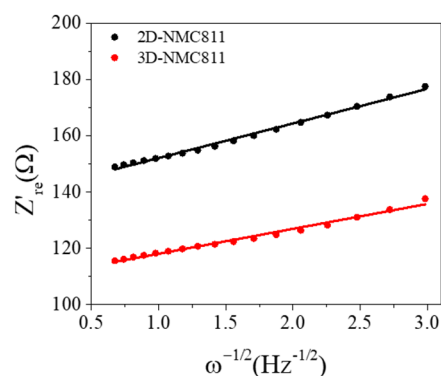
**Figure S2.** (a) XRD patterns of NMC811 and (b) the enlarged diffraction of (006)/(102) and (008)/(110) XRD peaks.



**Figure S3.** Galvanostatic charge/ discharge profiles of (a) 40-μm-2D-NMC811, (b) 40-μm-3D-NMC811, (c) 100-μm-2D-NMC811, and (d) 100-μm-3D-NMC811.



**Figure S4.** Diffusivity coefficient calculation. Normalized peak current obtained from CV scans plotted as a function of the square root of the scan rate for (a) 2D-NMC811 and (b) 3D-NMC811.



**Figure S5.** Linear fit lines demonstrating correlation between  $Z_{re}$  and  $\omega^{-1/2}$  extracted from EIS data.

**Table S1.** Summary of effect of laser structuring on electrodes at various thickness. .

Characteristics	Electrode thickness ( $\mu\text{m}$ )	
	40	100
Increase of interfacial area (%)	20	78
Aspect ratio (AR)	0.96	3.72
Loss of material (%)	10	6.4
Effective area ( $\text{cm}^2$ )	1.13	2.01

**Table S2.** Fitting results for impedance spectra and calculated  $D_{\text{Li}^+}$  from CVs and electrochemical impedance spectroscopy.

Electrodes	$D_{\text{Li}^+}$ ( $\text{cm s}^{-2}$ ) by CVs		EIS		
	Anodic	Cathodic	$R_s$ ( $\Omega$ )	$R_{CT}$ ( $\Omega$ )	$D_{\text{Li}^+}$ ( $\text{cm s}^{-2}$ )
2D-NMC811	$16.6 \times 10^{-11}$	$2.0 \times 10^{-11}$	3.2	136.6	$1.6 \times 10^{-11}$
3D-NMC811	$18.9 \times 10^{-11}$	$1.9 \times 10^{-11}$	1.4	107.7	$3.5 \times 10^{-11}$