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Nanoscale electrochemical characterization of solid-state electrolyte using manganese-based thin-film probe (Electronic Supplementary Information)

DOI: 10.1039/x0xx00000x	Yi-Hung	Liu,ª	Sin-Syu	Liao ^b	and	Bernard	Haochih	Liu* ^b

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Fig. S1 Optical microscope images of the (a) original AFM probe, and thin-film probes after the (b) electrodeposition and (c) hydrothermal reaction.

The images were obtained by an optical microscopy (BX51, OLYMPUS). The AFM probe (a) can be observed with a clean surface in light brown color, except for the probe tip. After the electrodeposition (b), the color of the probe becomes darker. The probe (c) further changes its color to red brown/black (cantilever/chip) after the hydrothermal treatment. Despite no obvious difference in probe shape can be visually observed among the three probes, the change in probe color provides a visual evidence for the sequent thin-film formation during the electrodeposition and hydrothermal reaction.

^{a.} Department of Greenergy, National University of Tainan, 33, Sec. 2, Shu-Lin St., Tainan 70005, Taiwan.

^{b.} Department of Materials Science and Engineering, National Cheng Kung University, No.1, University Road, Tainan 70101, Taiwan. E-mail: hcliu@mail.ncku.edu.tw; Tel.: +886-6-2757575 ext. 62926; Fax: +886-6-2346290

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Fig. S2 Macroscale impedance spectrum for the LiPON thin film and its corresponding equivalent circuit together with best fitting results.

A CR2032 coin cell with a MIM structure of Au/LiPON/Pt was used to perform the macroscale EIS measurement, in which a sinusoidal voltage of 10 mV over the frequency range from 0.1 Hz to 100 kHz was applied. Re, CPEe (Ye) and CPEblock (Yb) in the equivalent circuit represent the electrolyte resistance, constant phase elements for electrolyte and blocking electrode, respectively.

Table S1. Summary of the best fitting results for the impedance spectra shown in Fig. 4a.

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Bias	0 V	0.5 V	1 V	1.5 V	2 V
$R_{\rm e}$ (ohm)	1.03×10 ⁹	1.34×10 ⁹	7.53×10 ⁸	5.12×10 ⁸	4.10×10 ⁸
$Y^{a}_{w}(S^{*\sqrt[a]{S}})$	1.34×10^{-10}	1.27×10^{-10}	Unused		
a_w	0.57	0.56			
$Y_{\rm e} \left({\rm S}^{*a} \sqrt{S} \right)$	5.05×10 ⁻¹²	4.42×10 ⁻¹²	5.40×10 ⁻¹²	5.32×10 ⁻¹²	3.26×10 ⁻¹²
a _e	0.90	0.92	0.91	0.91	0.99
$Y_{\rm ct}({\rm S}^*\sqrt[a]{S})$	0	4.04×10 ⁻¹¹	8.92×10 ⁻¹⁰	2.03×10 ⁻⁹	3.18×10 ⁻⁹
a _{ct}	0	0	0.13	0.08	0.28
$C_{b}(\mathbf{F})$	4.52×10 ⁻¹⁰	2.95×10 ⁻⁹	1.60×10 ⁻¹¹	1.16×10 ⁻¹¹	4.18×10 ⁻¹²

a) $(Y(j\omega) = \sqrt[a]{\frac{1}{Z_{CPE}(\omega)}}$ (Z_{CPE}: impedance for constant phase element))

Table S2. Summary of the best fitting results for the impedance	spectra shown in Fig. 4b.
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Tuble 52. Summary of the best mang results for the impedance spectra shown in Fig. 10.					
Bias	0 V	0.5 V	1 V	1.5 V	2 V
$R_{\rm e}(\rm ohm)$	3.31×10 ⁹	2.98×10 ⁹	2.26×109	1.60×10 ⁹	6.50×10 ⁸
$Y^{a}_{e}(S^{*}\sqrt[a]{S})$	2.83×10 ⁻¹¹	9.51×10 ⁻¹⁰	4.72×10 ⁻¹⁰	4.47×10 ⁻⁹	6.04×10 ⁻⁹
a _e	0.83	0.74	0.80	0.80	0.77
$R_{\rm ct}$ (ohm)	1.06×10^{10}	9.38×10 ⁹	4.68×10 ⁹	2.57×10 ⁹	1.36×10 ⁹
$C_{\text{interface}}(\mathbf{F})$	1.02×10^{-12}	1.03×10 ⁻¹²	9.47×10 ⁻¹³	9.65×10 ⁻¹³	9.56×10 ⁻¹³

 $\sqrt[a]{\frac{1}{Z_{CPE}(\omega)}}$ (Z_{CPE}: impedance for constant phase element))

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