Supporting Information

Scalable, Large-Area Printing of Pore-Array Electrodes for Ultra-High Power Electrochemical Energy Storage

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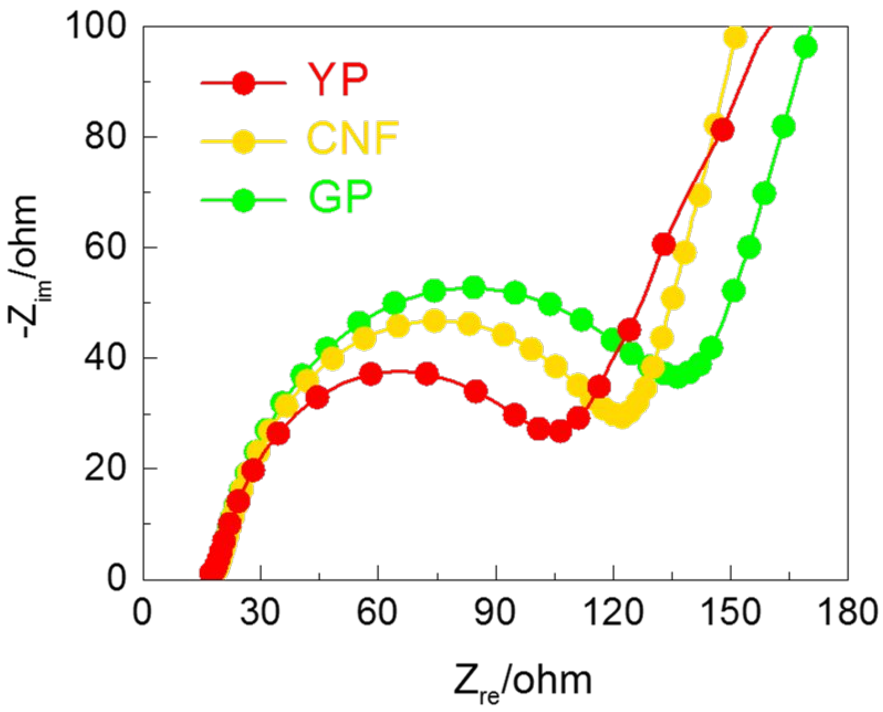

Figure S1. Comparative Nyquist plots of pore-array electrodes as a function of the active material types (YP, CNF and GP). The EIS tests were performed in the range of 1 MHz to 100 mHz after the first three charge/discharge cycles.

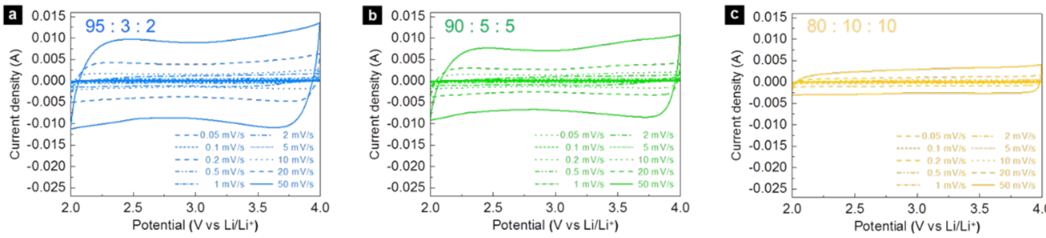

Figure S2. CV plots at increasing scan rates in the potential range of 2.0 to 4.0 V (*vs.* Li/Li^+) for pore-array YP-based electrodes with fractions of YP:SP:CMC in the mass ratio of (a) 95:3:2, (b) 90:5:5 and (c) 80:10:10.

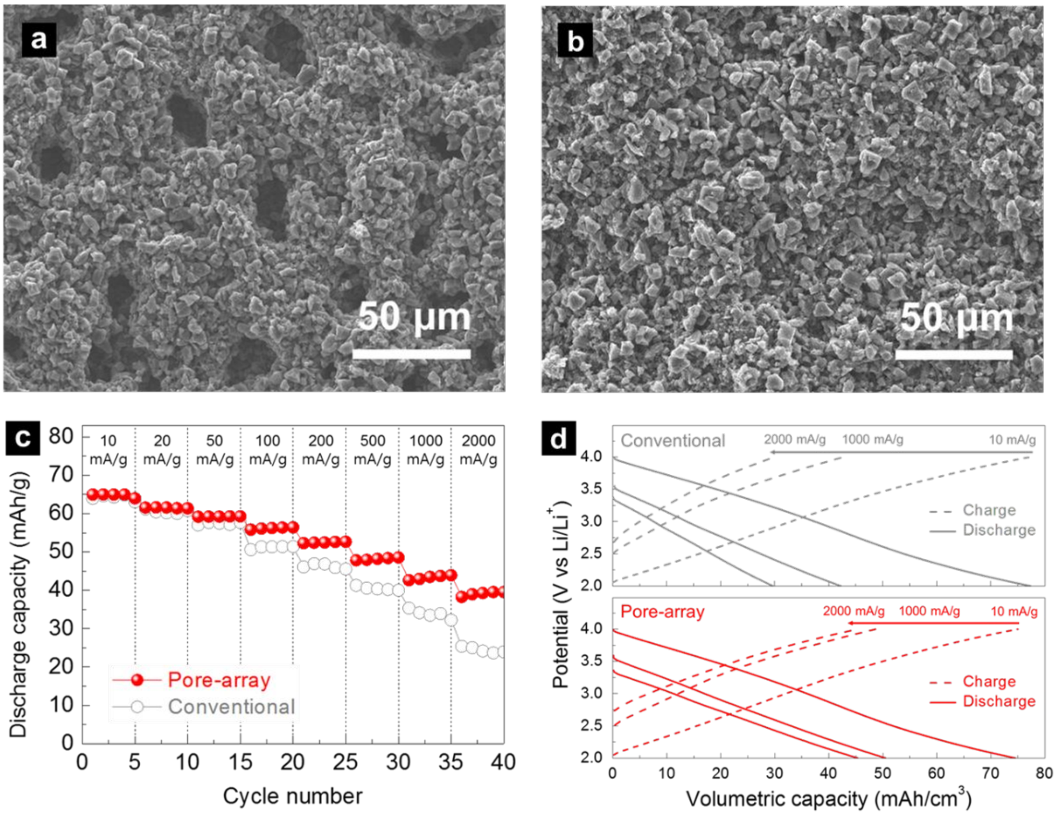

Figure S3. SEM images of the surface of (a) a spray printed pore-array YP electrode and (b) a conventional slurry cast YP electrode. (c) Comparative gravimetric discharge capacity plots for the YP-based electrodes with and without the pore-array at various charge/discharge current densities in the potential range of 2.0 to 4.0 V (*vs.* Li/Li^+). (d) Corresponding volumetric charge/discharge capacity plots at 10, 1000 and 2000 mA/g.

Table S1. Summary of the YP-based pore-array and conventional electrodes.

YP electrode	Formulation (YP : SP : CMC)	Thickness (μm)	Mass loading (mg/cm^2)
Honeycomb	95 : 3 : 2	20 ± 4	1.28 ± 0.03
Conventional	95 : 3 : 2	21 ± 3	1.37 ± 0.04

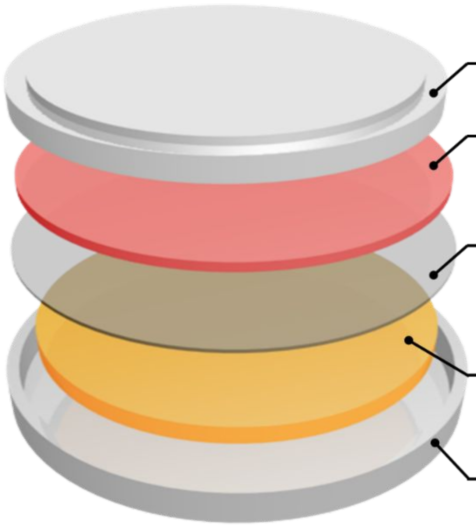

Figure S4. Schematic diagram of the coin-type LIC cell based on a spray printed LTO-based anode and a spray printed pore-array YP-based cathode.

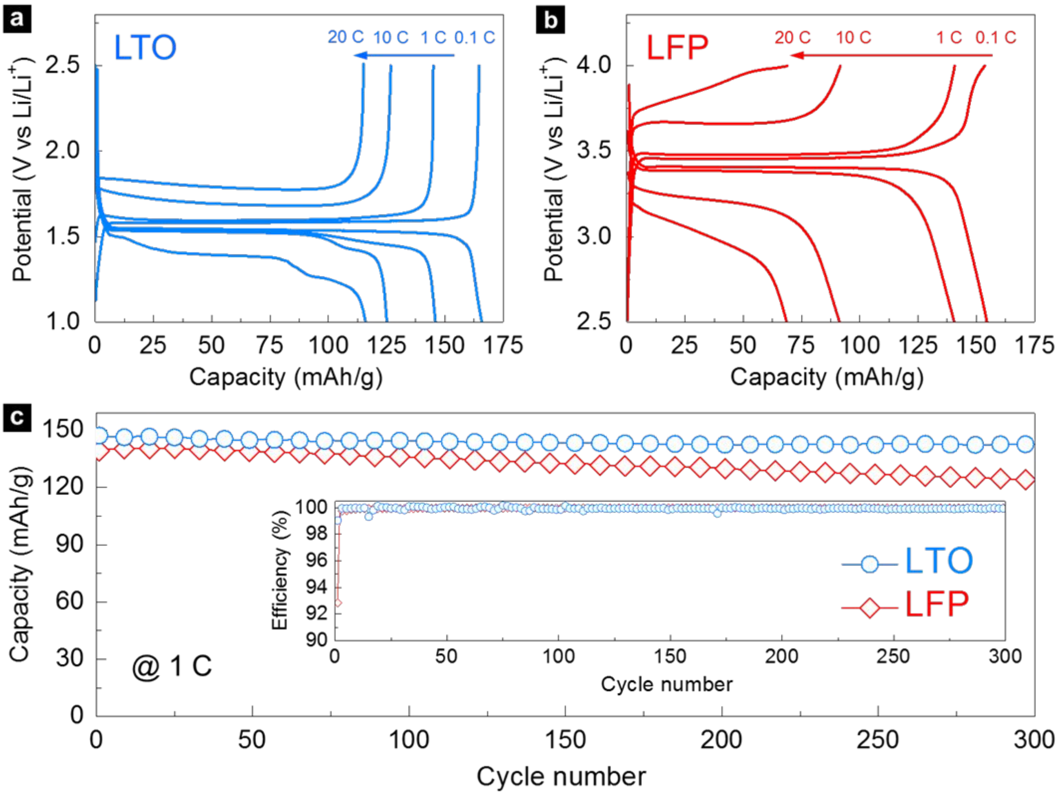

Figure S5. (a) Gravimetric charge/discharge plots of a spray printed LTO electrode at various C-rates in the potential range of 1.0 to 2.5 V (*vs.* Li/Li^+). (b) Gravimetric charge/discharge plots of a spray printed LFP-based electrode at various C-rates in the potential range of 2.5 to 4.0 V (*vs.* Li/Li^+). (c) Galvanostatic discharge profiles of the spray printed LTO and LFP electrodes at 1 C and the corresponding coulombic efficiency plots (inset).

Table S2. Summary of the spray printed LTO-based and LFP-based electrodes.

Electrode	Thickness (μm)	Mass loading (mg/cm^2)	Discharge capacity (mAh/g)			
			0.1 C	1 C	10 C	20 C
95:3:2 LTO:SP:CMC	10 ± 1	1.51 ± 0.03	153	145	127	116
95:3:2 LFP:SP:CMC	10 ± 1	1.53 ± 0.02	153	140	91	68

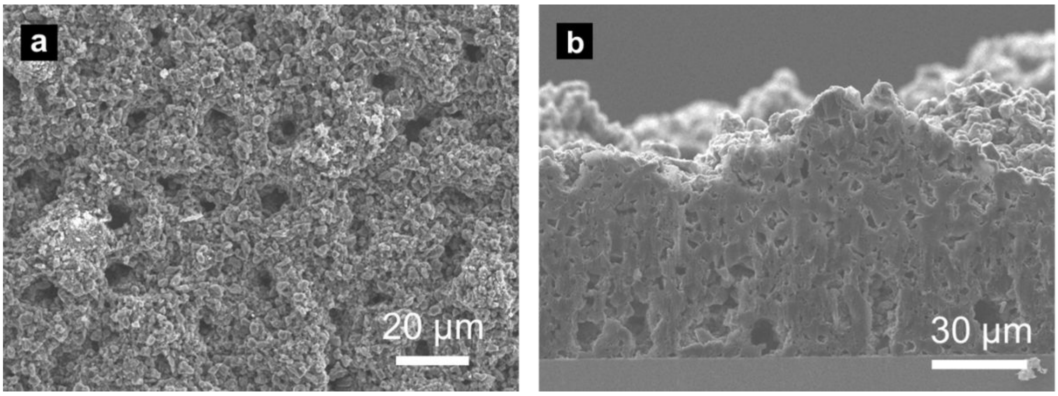

Figure S6. SEM images for (a) the plane view and (b) the cross-section of an 80 μm spray printed pore-array YP electrode.

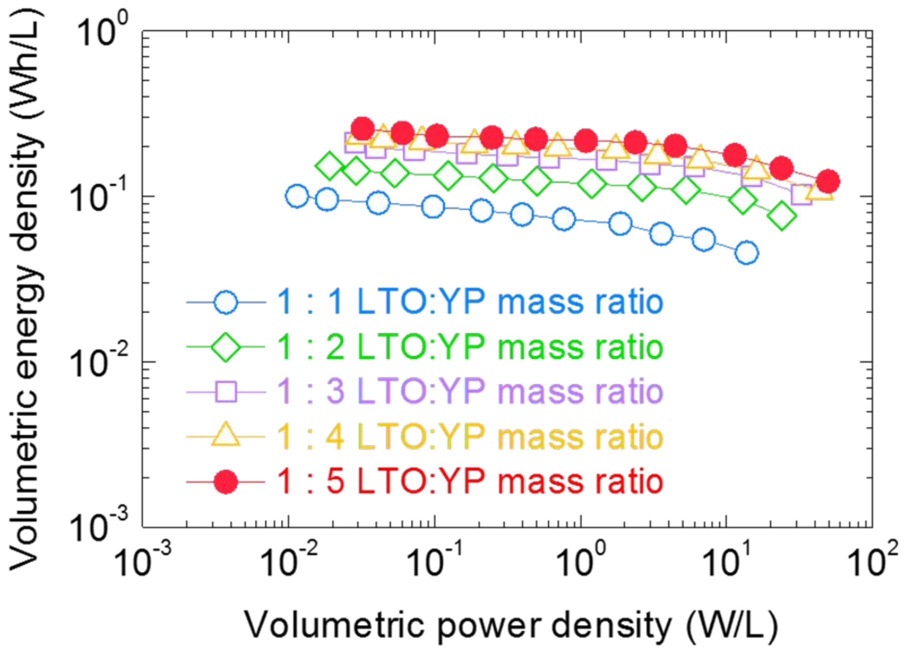

Figure S7. Volumetric Ragone plots of pore-array LICs as LTO:YP mass ratios increased from 1:1 to 1:5. Here, volumetric performances were re-estimated by normalizing the gravimetric values (Figure 6a) by the electrode loading mass and thickness of both anodes and cathodes, as given in Table 3.

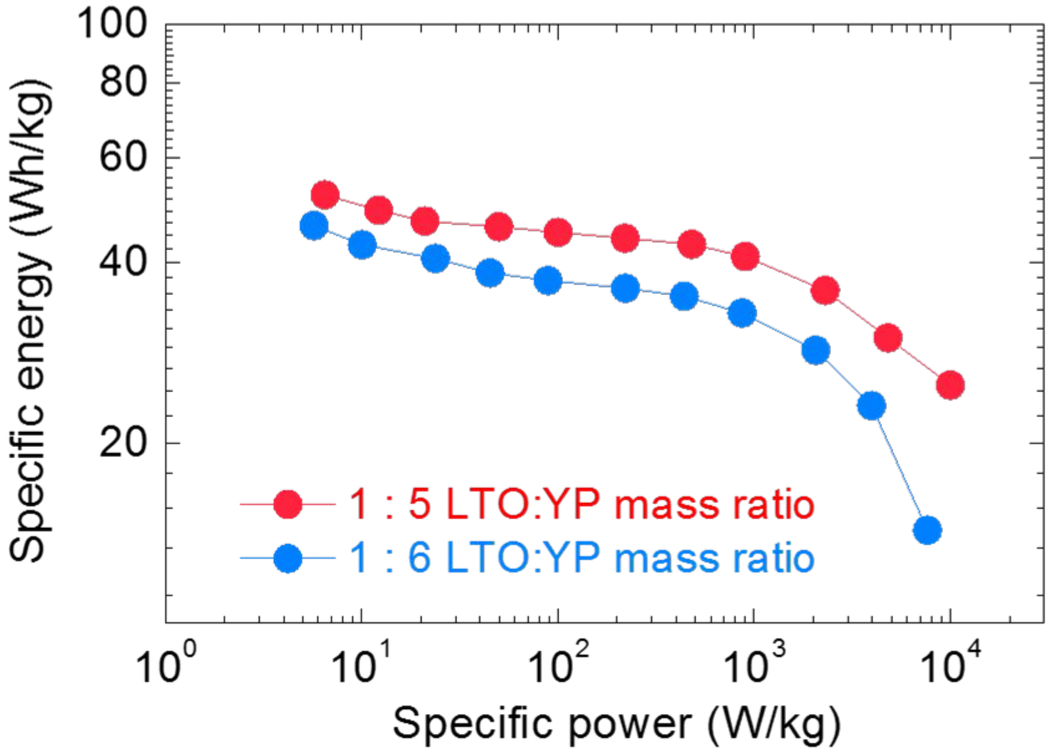

Figure S8. Ragone plots of the pore-array YP-based electrode LICs with LTO:YP mass ratios of 1:5 and 1:6.

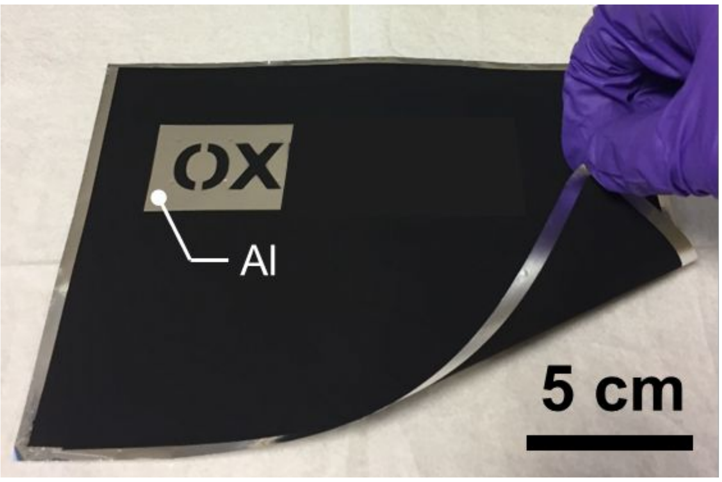

Figure S9. A photograph shows an A5-scale double-sided pore-array YP electrode and selective patterning of the letters "OX". For patterning, a patterned negative shadow mask was placed on a desired region of the current collector before the spray printing process. A pore-array YP-based layer was then spray printed through the negative part of the mask.

Table S3. Comparative performances of LTO/activated carbon-based LICs.

Reference	Materials combination		Performance
	Anode (wt%[a])	Cathode (wt%[b])	
[28]	LTO (80)	Graphene (80)	Energy: 45 Wh/kg @ 6.7 C Power: 3300 W/kg@ 110 C
[29]	LTO-C (80)	Graphene (70)	Energy: 72 Wh/kg @ 8.6 C Power: 8300 W/kg @ 208 C
[33]	LTO-C (70)	Carbon nanofiber (70)	Energy: 50.3 Wh/kg @ 4.4 C Power: 3000 W/kg @ 109 C
[34]	LTO-Graphene (80)	Activated carbon (90)	Energy: 50 Wh/kg @ 0.4 C Power: 2500 W/kg@ 167 C
[38]	LTO (80)	Graphite (80)	Energy: 55 Wh/kg @ 1.8 C Power: 6500 W/kg @ 176 C
This work	LTO (95)	Honeycomb YP (95)	Energy: 53Wh/kg @ 0.1 C Power: 9900 W/kg @ 450 C

[a] Weight fractions of active materials within the anode.

[b] Weight fractions of active materials within the cathode.