## **Electronic Supplementary Information**

## Spray Printing of Self-Assembled Porous Structures for High Power Battery Electrodes

Sang Ho Lee\*, Amoghavarsha Mahadevegowda, Chun Huang, Jack D. Evans and

Patrick S. Grant

Department of Materials, University of Oxford, Oxford OX1 3PH, UK.

<sup>\*</sup>Address correspondence to sangho.lee@materials.ox.ac.uk



**Fig.S1** A series of SEM images of the morphology change of the sprayed electrodes according to a change in the volume ratio of DI water to IPA in the liquid spray suspension: (a) 100:0, (b) 70:30, (c) 50:50, (d) 30:70 and (e) 0:100.



**Fig.S2** (a) SEM image of the pore structure of the  $TiO_2(B)$ -based electrode. (b) A magnified view around the pore region.



**Fig.S3** Schematic diagram of a proposed, simple model of how to fabricate throughthickness pore channels in a battery electrode based on layer-by-layer spray printing and bi-solvent drying.



**Fig.S4** A photograph of an as-spray printed, through-thickness pore electrode robustly withstanding severe bending and flexing by hand.



**Fig.S5** (a) Schematic diagram of the conventional slurry casting process using a doctor blade. (b) Plan view and (c) cross-sectional view of the slurry cast  $TiO_2(B)$ -NT-based electrodes. (d) Plan view and (e) cross-sectional view of the spray printed  $TiO_2(B)$ -NT-based electrodes.

Thickness (µm)	Sprayed (mg/cm²)	Slurry-cast (mg/cm²)
10	$0.53\pm0.02$	$0.68\pm0.03$
20	$0.98\pm0.03$	$1.38\pm0.04$
40	$\textbf{2.01} \pm \textbf{0.02}$	$2.65\pm0.05$
80	$3.99\pm0.04$	$5.53\pm0.06$

 Table S1 Summary of the electrode mass loading according to a change in electrode thickness.



**Fig.S6** Volumetric discharge capacities of 80  $\mu$ m thick spray printed and slurry cast electrodes at various C-rates in the potential range of 1.0 to 3.0 V (*vs.* Li/Li<sup>+</sup>).



**Fig.S7** SEM images of the 80  $\mu$ m thick TiO<sub>2</sub>(B)-NT-based electrodes (a) before and (b) after 200 cycles in the potential range of 1.0 to 3.0 V (*vs.* Li/Li<sup>+</sup>) at 1 C.



**Fig.S8** (a) Plan view and (b) cross-sectional SEM images of through-thickness porous electrodes, manufactured by spray printing of graphene bi-solvent suspensions, where graphene flakes of  $\sim$  5-7 µm diameter were exploited.