Supporting Information

Low-tortuosity and graded lithium ion battery cathodes by ice templating

Chun Huang [a*], Martin Dontigny [b], Karim Zaghib [b] and Patrick S. Grant [a]

[a] Department of Materials, University of Oxford, Parks Road, Oxford, OX1 3PH, UK

[b] Center of Excellence in Transportation Electrification and Energy Storage HydroQuebec, 1806 Lionel Boulet, Varennes, QC J3X1S1, Canada

E-mail: ann.huang@materials.ox.ac.uk

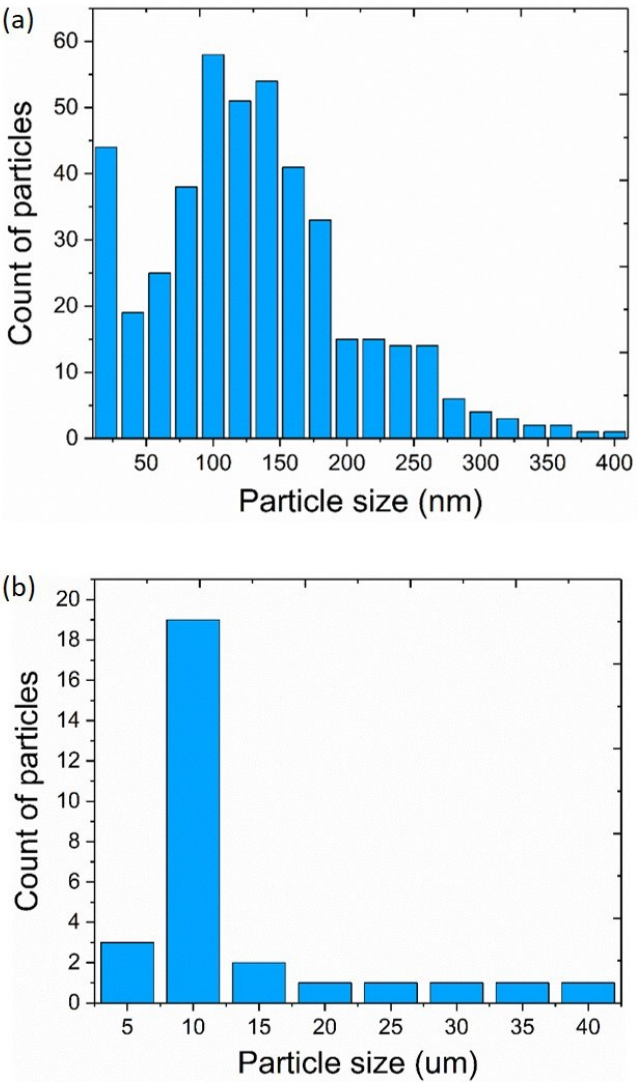


Figure S1: Size distributions of the LFP (a) primary and (b) secondary particles.

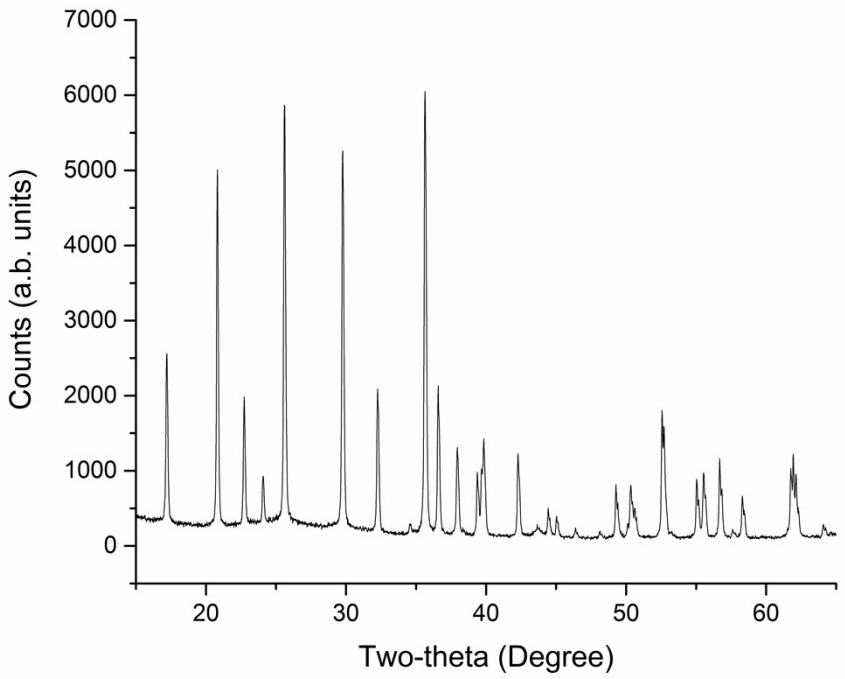
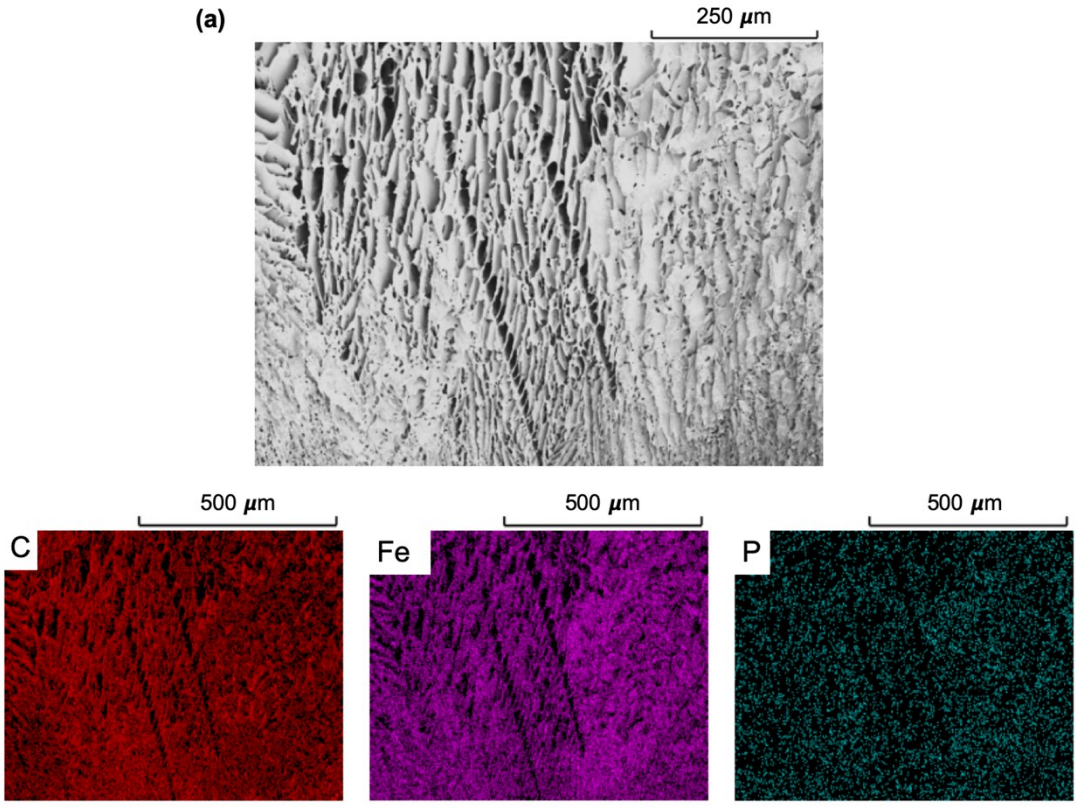


Figure S2: XRD pattern for the LFP electrode made by DIT.



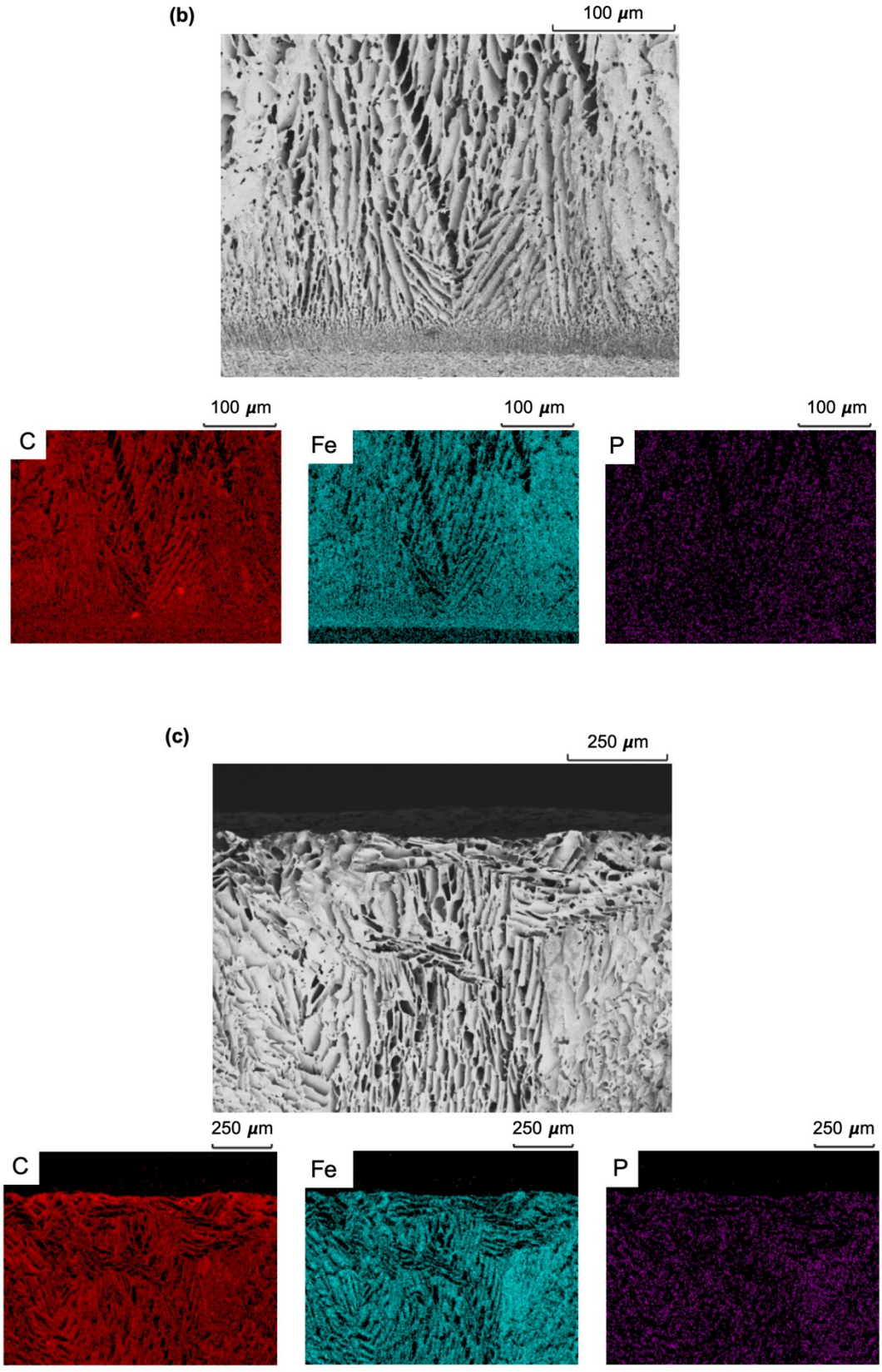


Figure S3: EDX analysis of the DIT LFP electrode cross sections (a) over the entire electrode thickness; (b) in the bottom region of the electrode; and (c) in the top region of the electrode.

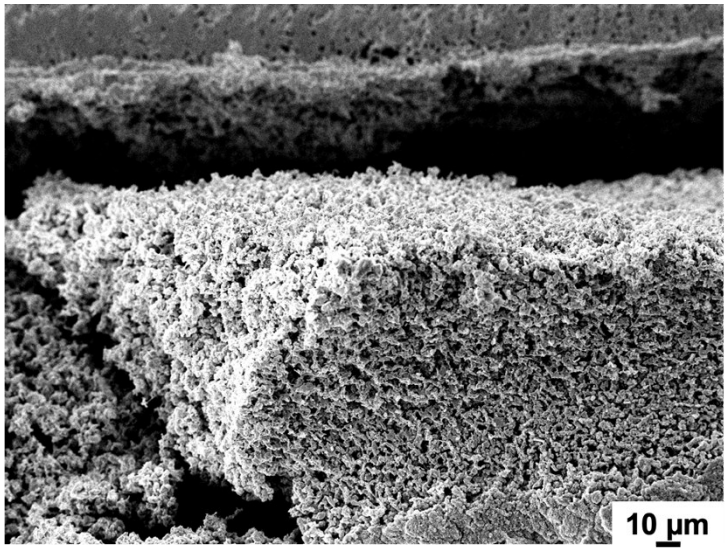


Figure S4: Cross-sectional SEM image of an IIT electrode.

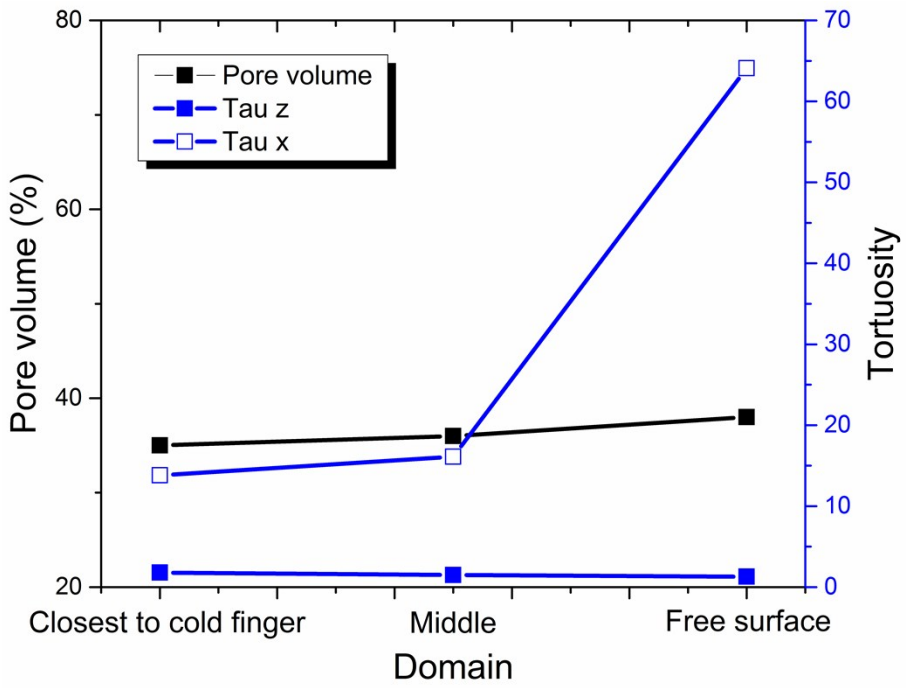


Figure S5: A plot showing pore volume, tortuosity along z direction (through electrode thickness) and tortuosity along x direction (within electrode plane) against the sub-domain of the electrode.

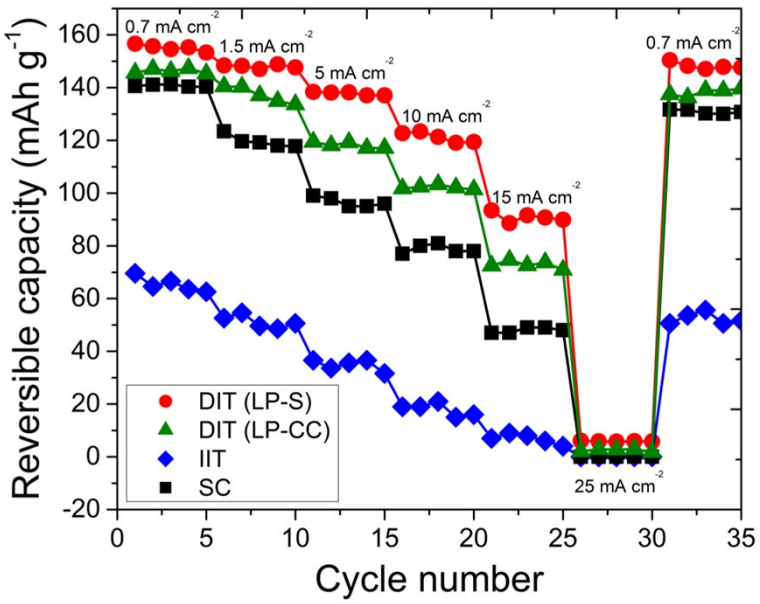


Figure S6: Reversible gravimetric capacities of the DIT (LP-S), DIT (LP-CC), IIT and SC electrodes at different current densities.

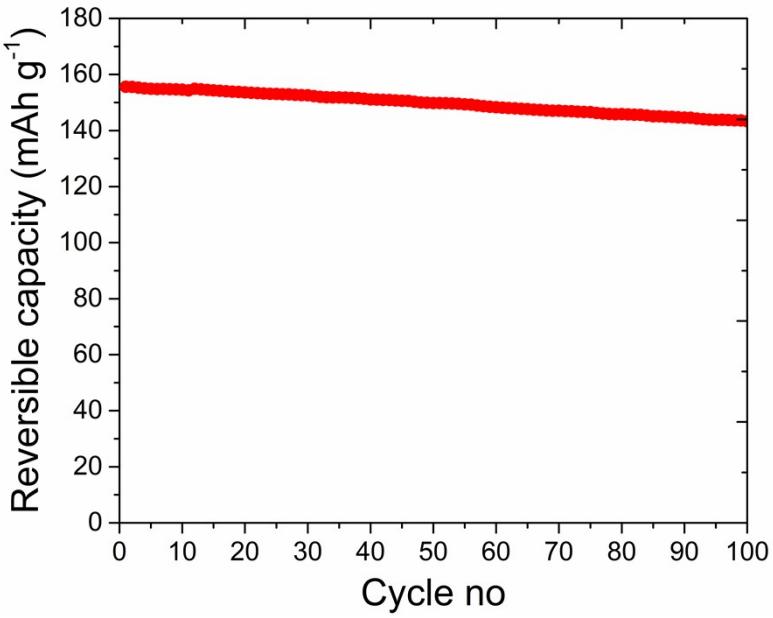


Figure S7: Cycling performance of the DIT (LP-S) electrode at 0.1 C.

Calculation of gravimetric and volumetric energy densities

Following the method in [R1], because the weight and volume of packaging as well as the inactive edge of a cell depend on cell dimensions and configurations, we only consider elementary building blocks or units of LIBs. One unit of LIB comprises a cathode, a separator, a Li foil, cathode current collector (Al) and anode current collector (Cu). For the cell-stack containing SC electrodes, the case that the electrode is coated on both sides of the current collectors is considered, consistent with commercial LIBs. The parameters of the components used for the estimation are also consistent with the relevant parameters in commercial LIBs [R1, R2]. The ½ thickness current collectors was 9 μm, the separator thickness was 25 μm and the Li foil thickness was 600 μm.

In the same total volume, the proportions of inactive components in the LIB unit(s) are:

LIB unit(s) of the same total volume	vol.% inactive components	wt.% inactive components
1 LIB unit containing a DIT (LP-S) cathode	2.8	9.2
1 LIB unit containing a DIT (LP-CC) cathode	2.8	9.2
1 LIB unit containing an IIT cathode	2.8	9.2
LIB units containing calendered SC cathodes	6.2	23.2

Table S1: vol.% and wt.% inactive components for a LIB cell unit comprising a thick, DIT (LP-S) LFP cathode, a separator, a Li counter electrode and current collectors, a LIB cell unit comprising the same components except for a DIT (LP-CC) LFP cathode, a LIB cell unit comprising the same components except for an IIT LFP cathode, and a stack of LIB cell units comprising conventional calendered SC LFP cathodes, separators, Li counter electrodes and current collectors, all four types are of same total volume.

The gravimetric energy density E_g of the LIB unit(s) is calculated as [R3, R4]:

$$E_g = Q_g V_{cell} M \qquad (1)$$

where Q_g is the gravimetric capacity of the LIB, V_{cell} is the mid-point voltage in discharge and M is the wt% of active components in LIB unit(s) of the same total volume.

The volumetric energy density E_v of the LIB unit(s) is calculated as:

$$E_v = Q_v V_{cell} N \qquad (2)$$

where Q_v is the volumetric capacity of the LIB, V_{cell} is the mid-point voltage in discharge and N is the vol.% of active components in LIB unit(s) of the same total volume.

References

[R1] F. Wu and G. Yushin, Conversion cathodes for rechargeable lithium and lithium-ion batteries, Energy Environ. Sci. 10 (2017) 435-459.

[R2] D. L. Wood III, J. Li and C. Daniel, Prospects for reducing the processing cost of lithium ion batteries, J. Power Sources 275 (2015) 234-242.

[R3] L.-L. Lu, Y.-Y. Lu, Z.-J. Xiao, T.-W. Zhang, F. Zhou, T. Ma, Y. Ni, H.-B. Yao, S.-H. Yu and Y. Cui, Wood-inspired high-performance ultrathick bulk battery electrodes, Adv. Mater. 1706745 (2018) 1-9.

[R4] J. Liu, Z. Bao, Y. Cui, E. J. Dufek, J. B. Goodenough, P. Khalifah, Q. Li, B. Y. Liaw, P. Liu, A. Manthiram, Y. S. Meng, V. R. Subramanian, M. F. Toney, V. V. Viswanathan, M. S. Whittingham, J. Xiao, W. Xu, J. Yang, X.-Q. Yang and J.-G. Zhang, Pathways for practical high-energy long-cycling lithium metal batteries, Nat. Energy 4 (2019) 180-186.