

Electronic Supplementary Information

Rechargeable lithium/iodine battery with superior high-rate capability by using iodine-carbon composite as cathode†

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The Brunauer, Emmett and Taylor (BET) surface areas and pore size distribution of the conductive carbon black (CCB) and the as-prepared iodine-CCB composite were obtained from nitrogen adsorption/desorption curves with a JW-BK instrument (Beijing, China). All blank samples were preprocessed at 200 °C for 1h before adsorption–desorption measurements. BET surface areas were calculated from the linear part of the BET equation over the relative pressure range of 0.05–0.50. The micropore and mesopore size distributions (MMSD) were calculated using Horvath–Kawazoe (HK) and Barrett–Joyner–Halenda (BJH) methods, respectively.

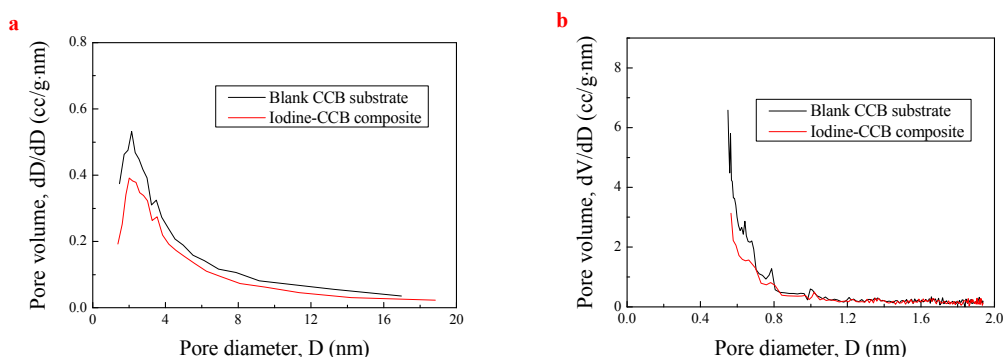


Fig. S1. BJH mesopore distribution (a), and HK micropore distributions (b) of blank CCB substrate and as-prepared iodine-CCB composite.

Fig. S1 shows the pore distribution of both micropore and mesopore of the as-prepared iodine-CCB composite and blank CCB substrate, respectively. For the blank CCB substrate, the most frequent pore diameter is 4.3 nm, and the BET surface area is 951.7 m²/g. In the as-prepared iodine-CCB composite, the BET surface area decreases to 635.6 cm²/g, due to the active iodine loading in the mesopores and micropores of CCB. The pore structure parameters of CCB and iodine-CCB composite are list in Table. S1.

Table. S1 Surface area and pore-structure parameters of blank CCB and iodine-CCB composite.

Sample	S (m^2/g)			V (cm^3/g)		Pore Size (nm)	
	S_{BET}^a	S_{meso}^b	S_{micro}^c	V_{meso}^d	V_{micro}^e	D_{meso}^f	D_{micro}^g
Blank CCB	951.7	621.1	330.6	0.81	0.44	4.3	0.56
Iodine-CCB	635.6	512.7	122.9	0.67	0.30	4.2	0.57
Subtracted value ^h	316.1	108.4	207.7	0.14	0.14	-	-

^a BET surface area, ^b BJH desorption cumulative surface area of pores between 1.7 ~ 300 nm, ^c Micropore surface area, $S_{\text{BET}} - S_{\text{meso}}$, ^d BJH desorption cumulative volume of pores between 1.70~300.00 nm, ^e HK adsorption cumulative volume of micropores, ^fBJH most frequent pore diameter, ^g HK most frequent pore diameter. ^h The reduced value of surface area and pore volume between the blank CCB and the iodine-CCB composite.

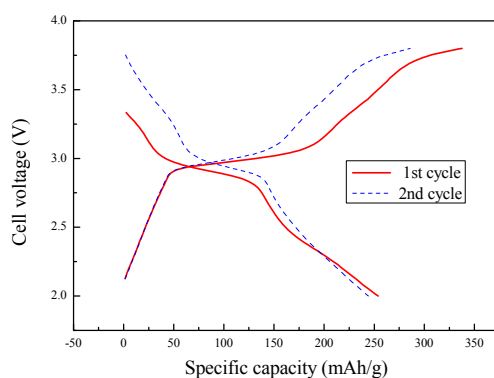


Fig. S2. The discharge and charge profiles in the initial two cycles. The capacity is calculated based on the mass of iodine in cathode electrode. The cathode is prepared by mixing the iodine-CCB composite, poly(tetrafluoroethylene), and acetylene-black with the weight ratio of 80:10:10.

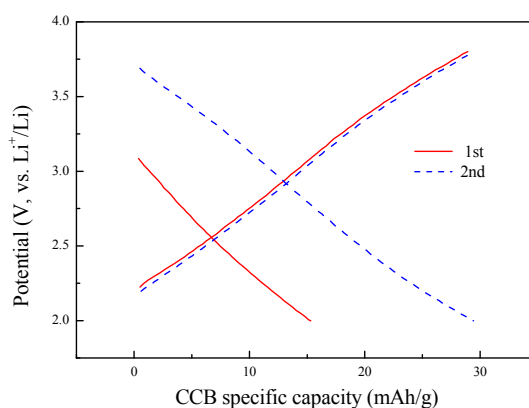


Fig. S3. The initial two discharge and charge profiles of CCB and AB under the same current density. The cathode is prepared by mixing the CCB, poly(tetrafluoroethylene), and acetylene-black with the weight ratio of 60.8:10:10, in accord with the ratio in iodine-CCB cathode. The specific capacity is calculated based on the total mass of CCB and AB.

The obtained initial discharge capacity of 250 mAh/g (Fig. S2), calculated based on the mass of active iodine in the composite, is higher than the theoretical capacity of iodine (211 mAh/g), indicating an extra capacity contribution of CCB substrate and AB binder during the discharge process. In the blank experiment at the same discharge current density (Fig. S3), the typical charge and discharge profiles of the CCB and AB are nearly linear with a stable capacity of 30 mAh/g (60 F/g or 4.87 $\mu\text{F}/\text{m}^2$), in agreement with the capacitance (vs. pore size of 1.2 or 2.8 nm) previously reported by Chmiola.¹ After a deduction of the capacity contribution of CCB and AB on the basis of the linear profile, the charge and discharge profile of active iodine in the composite is obtained and shown in Fig. 2.

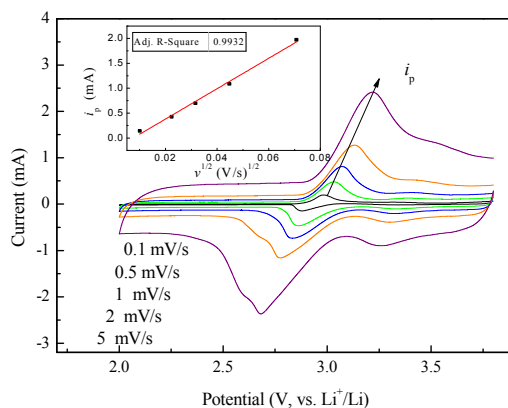


Figure S4. CVs of the iodine-carbon composite at various scan rates of 0.1, 0.5, 1, 2 and 5 mV/s. The linear relationship of i_p (oxidation peak current of iodide ion) vs. $v^{1/2}$ (the square root of scan rates) is inserted.

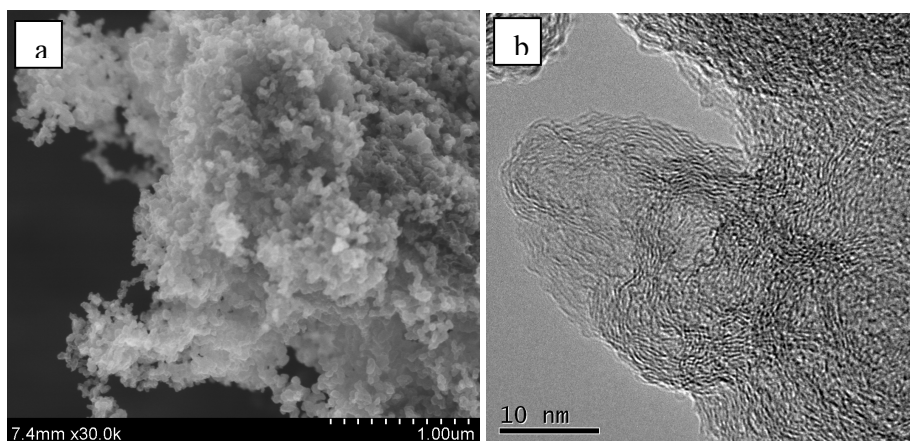


Fig. S5. SEM (a) and TEM (b) images of conductive carbon black (CCB) substrate.

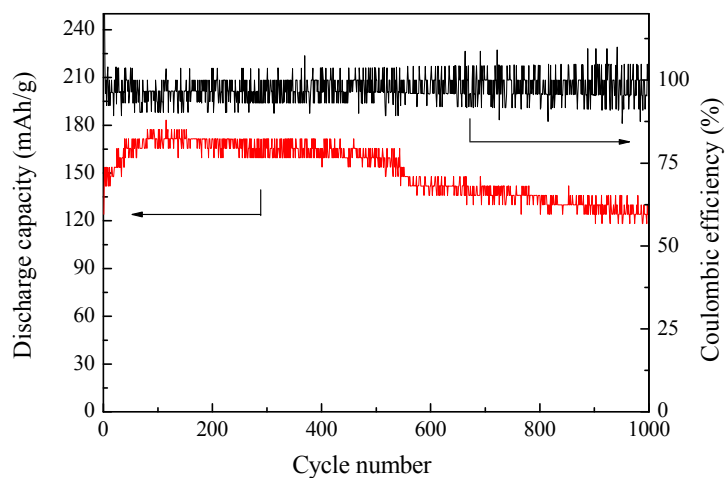


Figure S6. Cycle performance of the lithium/iodine cell at 100C rate

At the high rate (100C), the capacity profile shows an activation process before 100 cycles, and the discharge capacity stabilizes at 175 mAh/g before 560 cycles. Subsequently, the discharge capacity decreases gradually to 125 mAh/g after 1000 cycles. In adjacent several cycles, the coulombic efficiency change sharply in the range of 90~110%, respectively, probably due to the shuttle reaction mechanism, like in lithium/sulfur cell.² The iodine-CCB composite shows excellent high rate performance.

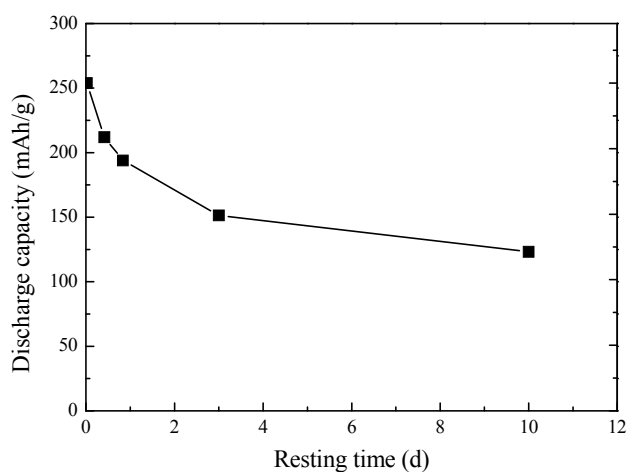


Fig. S7 The corresponding capacity retention after various resting time.

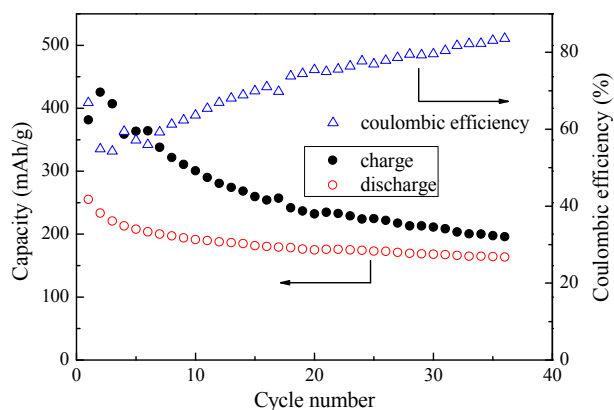


Fig. S8 Cycle performance and coulombic efficiency of lithium/iodine cell at 1C rate. The capacity is calculated based on the mass of active iodine in the composite.

The coulombic efficiency is obviously lower at 1C, indicating the dissolution of active iodine of the composite in electrolyte is more serious due to the shuttle reaction.

References

1. J. Chmiola, G. Yushin, Y. Gogotsi, C. Portet, P. Simon and P. L. Taberna, *Science*, 2006, **313**, 1760-1763.
2. Y. V. Mikhaylik and J. R. Akridge, *J. Electrochem. Soc.*, 2004, **151**, A1969-A1976.