

Electronic Supplementary Information

Encapsulating high content of iodine into active graphene substrate as cathode material for high-rate lithium–iodine batteries

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BET analysis of blank AG and I₂-AG composites

The Brunauer Emmett and Taylor (BET) surface areas and pore size distribution of the blank active graphene (AG) and the I₂-AG composites with 45 wt%, 56 wt% and 72 wt% I₂ were obtained from nitrogen adsorption/desorption curves, as shown in Fig. S1. Fig. S1a and 1b show HK micropore distributions BJH and mesoporous distribution of blank AG and I₂-AG composites, respectively. The AG has a large surface area of 1846 m² g⁻¹ and a large pore volume of 1.96 cm³ g⁻¹, with the predominant pore size around 0.5–5 nm. This results indicate that the AG has abundant microporous and mesoporous. The most frequent micropores and mesopores size are 0.7 nm and 2.5 nm, respectively. For I₂-AG composites, the pore volume decrease with increasing the content of iodine in composites.

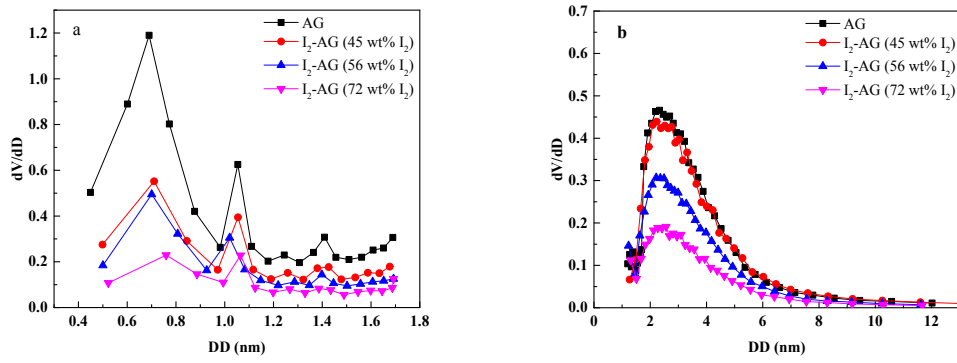


Fig. S1 HK micropores distributions (a) and BJH mesopores distribution (b) of blank AG substrate and the as-prepared I₂-AG composites.

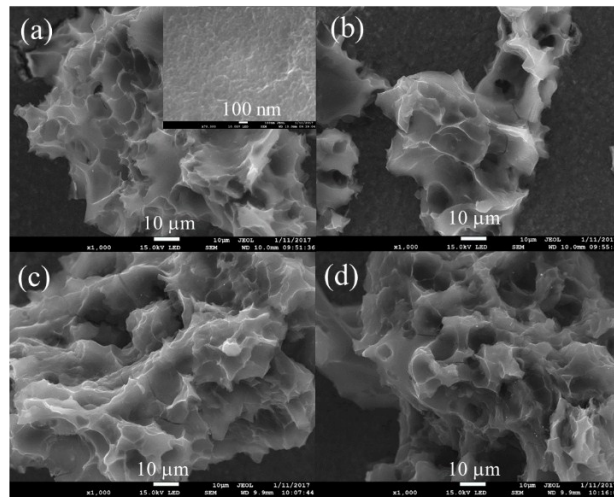


Fig. S2 SEM images of the blank AG (a) and the I₂-AG composites with 45 wt% I₂ (b), 56 wt % I₂ (c) and 72 wt% I₂ (d).

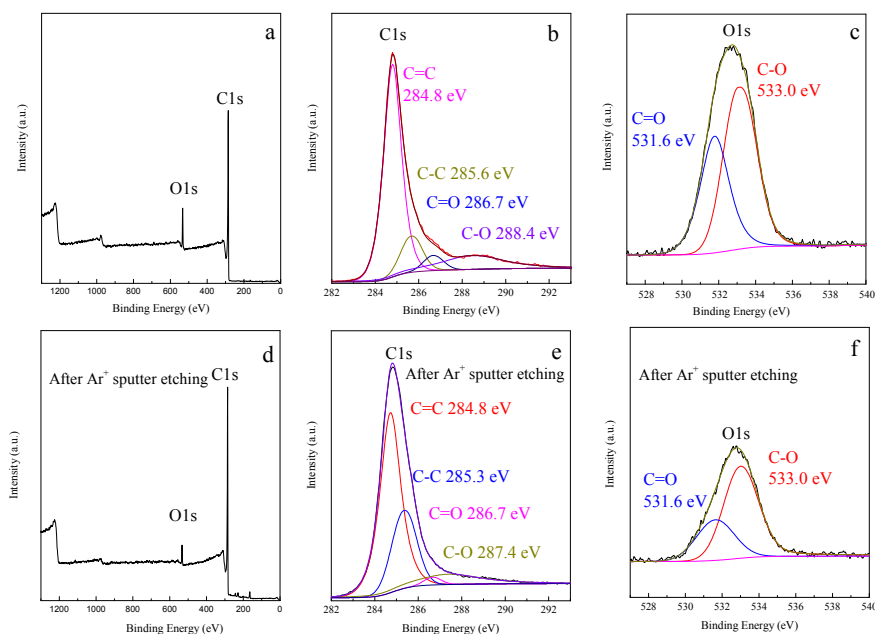


Fig. S3 XPS survey spectra of the AG (a), the XPS C1s (b) and O1s peaks (c) of the AG; XPS survey spectra of the AG (d), the XPS C1s (e) and O1s peaks (f) of the AG after Ar⁺ sputter etching with a 2 keV Ar⁺ beam for 100 s.

XPS analysis of blank AG

Fig. S3a, b and c show that the surface of AG is mainly composed of C and O elements. The high resolution of C1s reveals a narrow main peak at 284.8 eV that is assigned to C=C, and three minor peaks at 285.3 eV, 286.7 eV and 287.4 eV assigned to C-C, C=O and C-O banding. Two peaks of C=O and C-O in the O1s spectra indicate that AG is composed of a small part of oxygen-containing functional groups. After etching the AG with a 2 keV Ar⁺ beam for 100 s, the percentage of surface oxygen groups in AG decreases obviously, which can be seen clearly in Fig. S3d–f. This result confirms that the AG is a typical reduced graphene oxide, which is mainly composed of carbon element.¹

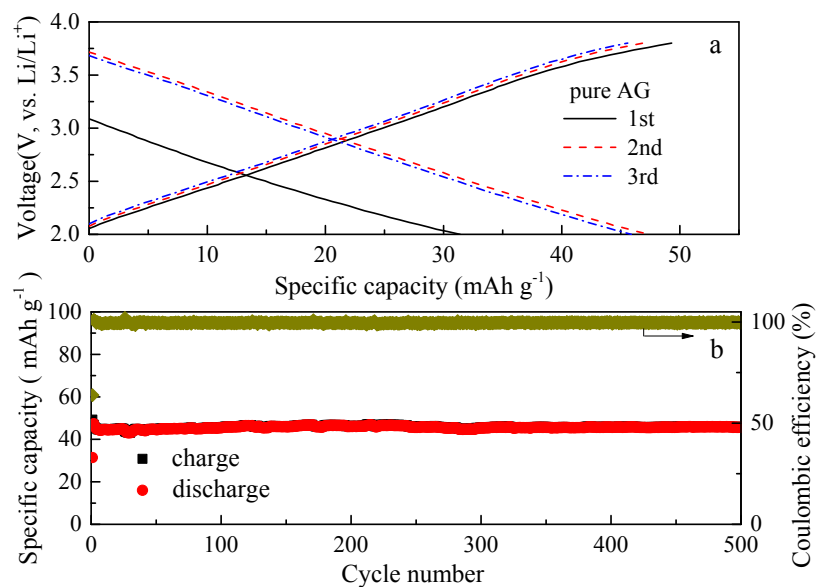


Fig. S4 The initial three charge-discharge (a) and cycle performance (b) curves of the AG at 1 C rate. The specific capacity is calculated based on total mass of cathode electrode. The cathode was prepared by compressing a mixture of the AG, conductive carbon (Super P) and poly-tetrafluoroethylene (PTFE) in a weight ratio of 70:20:10 with ethanol as dispersant.

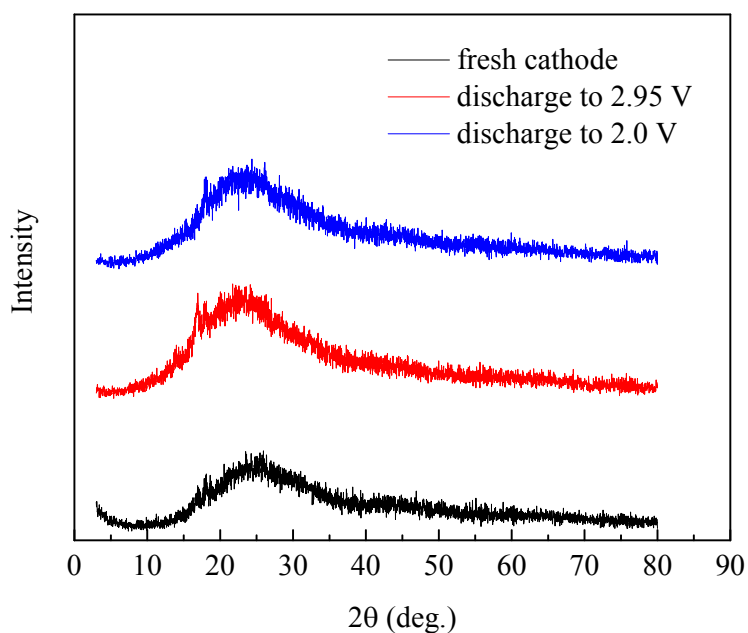


Fig. S5. Ex-situ XRD patterns of the I₂-AG composite with 56 wt% I₂ in different discharge depth.

Self-discharge Performance

The self-discharge performance of the battery after full charge with different resting time are shown in Fig. S6. The specific discharge capacities retain 186, 149, and 146 mAh g⁻¹, with the resting time of 24, 48 and 72 h, respectively. The degree of self-discharge increases along with extending resting time, indicating the dissolution of I₂. However, as the resting time increases to 48 h, the self-discharge capacity shows a relatively stable value, which means the solubility of iodine reaches an equilibrium in the electrolyte. In addition, after different resting times, the lithium iodine battery still exhibits appreciable discharge capacity and cycle performance, indicating the dissolved iodine in the electrolyte can be utilized in the electrochemical process.

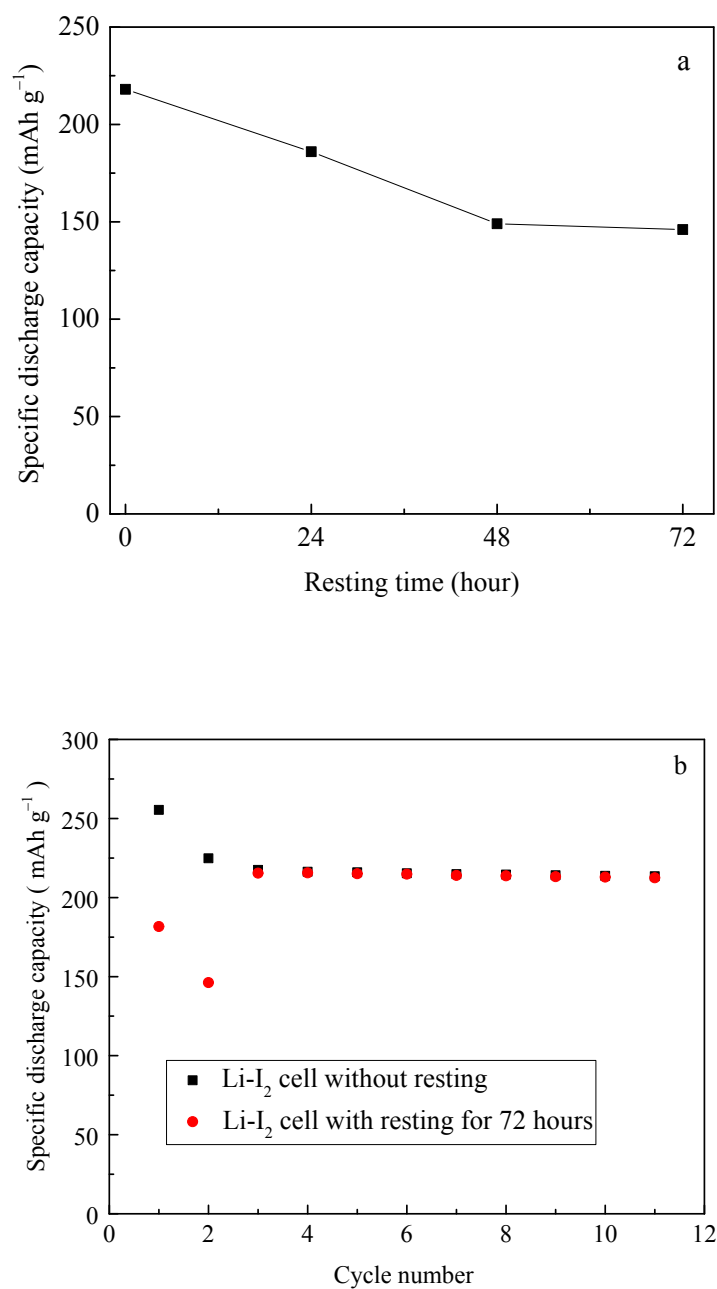


Fig. S6 Self-discharge performance of the I₂-AG composite with 56 wt% I₂. (a) Specific discharge capacity of the cell after resting for different time. (b) Comparison of cycling performance of the cells with and without resting after an initial activation discharge-charge process.

Table S1 A detail comparison of the performances of the recently reported Li-I₂ batteries.

Cathode	Electrolyte	Mass loading	Specific capacity	CE	Ref.
I ₂ -LiI(HPN) ₂	LiI(HPN) ₂	50%(molar ratio)	0.4 mAh cm ⁻² (1 th)	100%	2
I ₂ (aqueous cathode)	I ₂ aqueous electrolyte; 1M LiPF ₆ /EC+DMC		207 mAh g ⁻¹ (2.5 mA cm ⁻² , 1 th)	100%	3
I ₂ /CCB	1M LiPF ₆ /EC +EMC+DMC	24 wt% (I ₂)	61.2 mAh g ⁻¹ (1 C, 1 th) 36 mAh g ⁻¹ (1 C, after 30 th) (calculated by composite)	50-85%	4
I ₂ /CC	1 M LiTFSI/DOL+ TEGDME+1wt% LiNO ₃	22 wt% (I ₂)	65.8 mAh g ⁻¹ (0.5 C, 1 th); 42.9 mAh g ⁻¹ (0.5 C, after 300 th); (calculated by composite)	> 90%	5
I ₂ cathode with a B ₂ O ₃ /carbon interlayer	1 M LiPF ₆ /PC+EC+ DEC		176.9 mA h g ⁻¹ (20 C, after 500 th) (calculated by iodine)	> 90%	6
I₂-AG (this work)	1 M LiTFSI/DOL+ TEGDME+2wt% LiNO ₃	56 wt% (I ₂)	122.1 mAh g ⁻¹ (1 C, 2 th) 90.2 mAh g ⁻¹ (1 C, after 500 th) (calculated by composite)	> 94%	this work

References

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