Electronic Supporting Information (ESI)

A Long-Life Lithium Ion Oxygen Battery Based on the Commercial Silicon Particles as Anode

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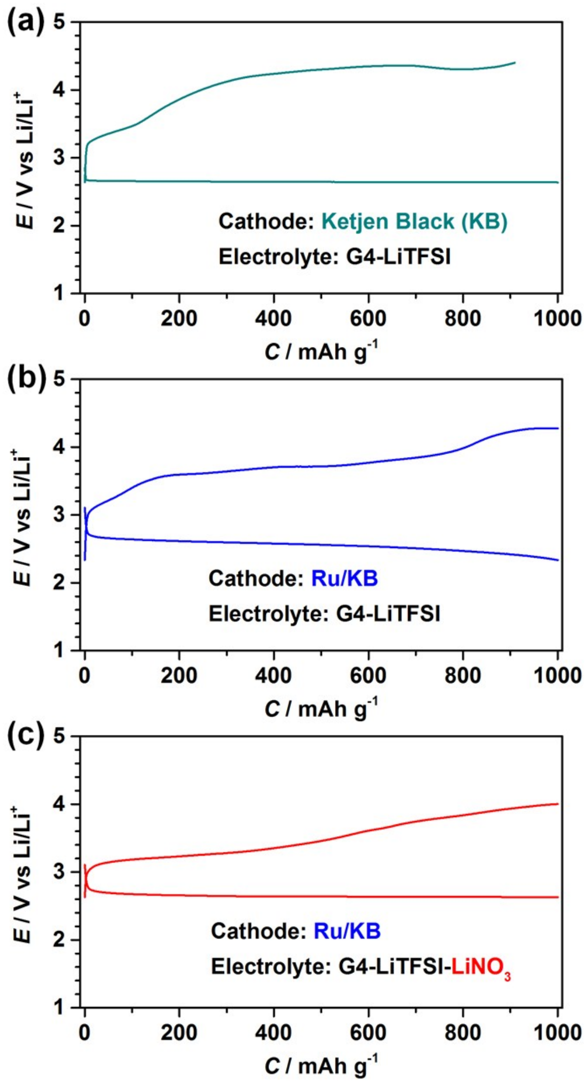

Fig. S1 Discharge-charge profiles of Li-O_2 half cells (Li metal anodes) with (a) KB cathode and (b) Ru/KB cathode in the G4-LiTFSI electrolyte and Ru/KB cathode in G4-LiTFSI-$LiNO_3$ (c) electrolyte, respectively. Current density: 500 mA g^{-1}.

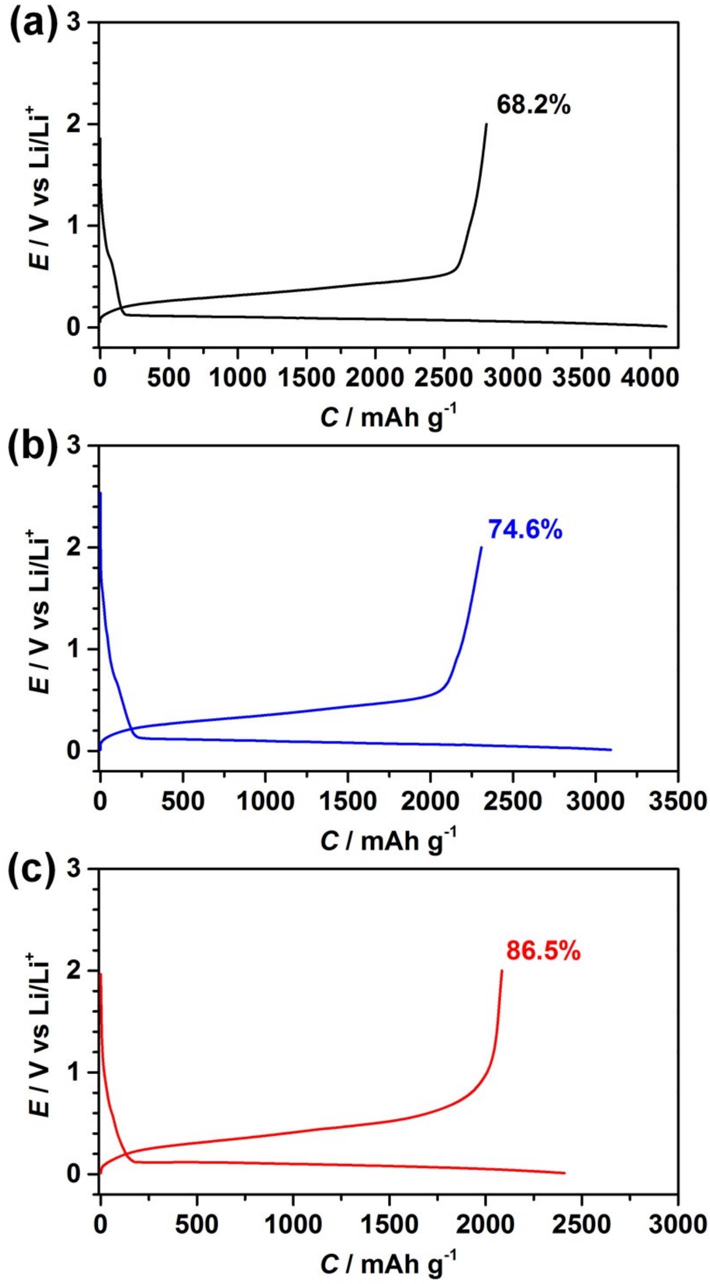

Fig. S2 The initial discharge-charge profiles of Li ion half cells with Si electrodes fabricated using PVDF1100 (a), CMC/SBR (b) and PAA (c) as the binders, respectively. The electrolyte is 1.0 M $LiPF_6$ in EC/DEC solvent. The current density is 50 mA g^{-1}. The columbic efficiencies of the first discharge-charge cycle are 68.2%, 74.6% and 86.5%, respectively. The high value of 86.5% is approaching the commercial level for Si materials, indicating the important role for the improvement of the reversibility by introducing PAA as the binder for Si electrodes. Therefore, we selected PAA binder to prepare the Si electrodes.

The calculation of specific area capacity ratio of anode/cathode

For Li-O_2 batteries with Li metal anodes

The calculation of specific area capacity ratio of Li metal anode/cathode: (3680 mAh g^{-1}*0.534 g cm^{-3}*0.05 cm)/(1000 mAh g^{-1}*0.5 mg cm^{-2}*0.85) = 230

Capacity of Li metal = 3680 mAh g^{-1}, Density of Li metal = 0.534 g cm^{-3}, Thickness of Li metal = 0.05 cm, Capacity of cathode = 1000 mAh g^{-1}, Mass loading of cathode = 0.5 mg cm^{-2}, active material percentage = 0.85.

For Li ion O_2 batteries with lithiated Si anodes

The calculation of specific area capacity ratio of lithiated Si anode/cathode: ~ (2400 mAh g^{-1}*1.0 mg cm^{-2}*0.7) /(1000 mAh g^{-1}*0.5 mg cm^{-2}*0.85) = ~3.9

The calculation of specific area capacity ratio of lithiated Si anode/cathode: ~ (2400 mAh g^{-1}*0.65 mg cm^{-2}*0.7) /(1000 mAh g^{-1}*0.5 mg cm^{-2}*0.85) = ~2.5

Capacity of Si anode = 2400 mAh g^{-1}, Mass loading of Si anode = 1.0 or 0.65 mg cm^{-2}, Si percentage = 0.7, Capacity of cathode = 1000 mAh g^{-1}, Mass loading of cathode = 0.5 mg cm^{-2}, active material percentage = 0.85.

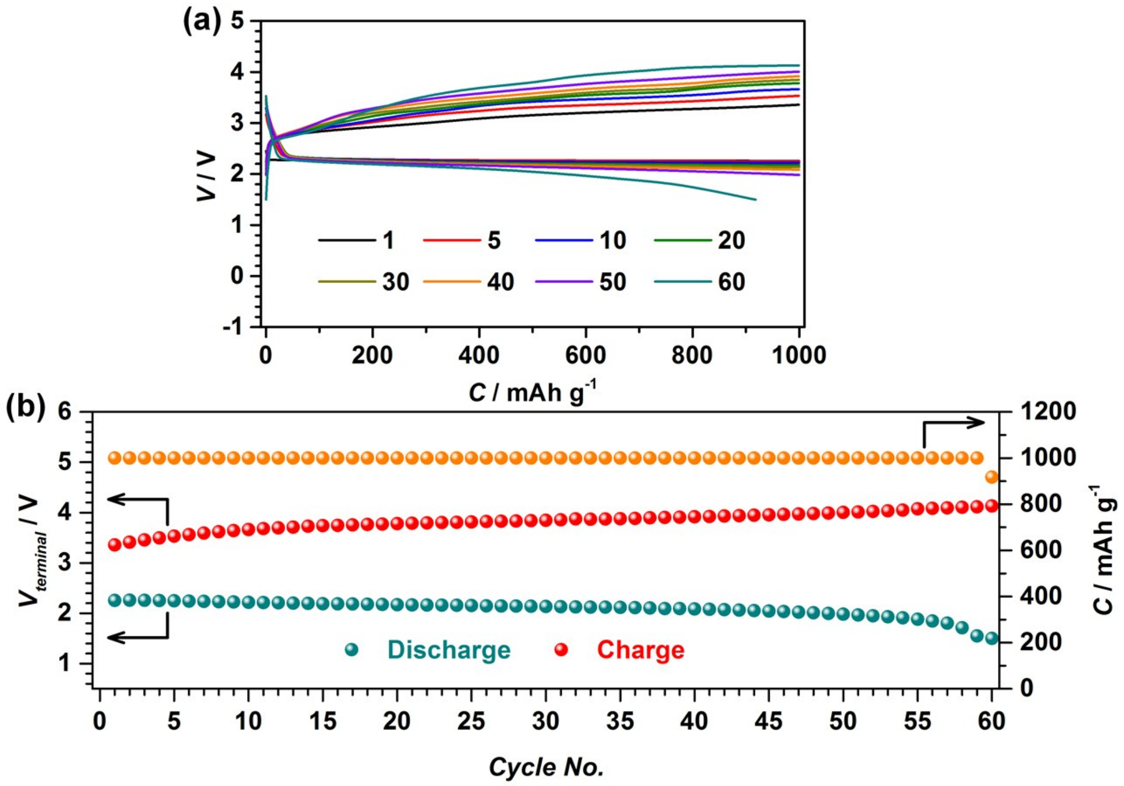

Fig. S3 The performance of Li ion O_2 batteries with F-L-Si anodes at 500 mA g^{-1}. The mass loadings of cathode and Si anode are 0.5 mg cm^{-2} and 0.65 mg cm^{-2}, respectively. The mass ratio of cathode/anode (0.93) is ~1.6 times as that in the former battery (0.61) and the specific area capacity ratio of anode/cathode is ~2.5. (a) The selected discharge-charge profiles of Li ion O_2 batteries with F-L-Si anodes. (b) The specific capacities along with discharge and charge terminal voltages against cycle number of Li ion O_2 batteries with F-L-Si anodes.

When the mass loading of cathode kept at 0.5 mg cm^{-2} and the mass loading of Si anode was decreased to be 0.65 mg cm^{-2}, the mass ratio of cathode/anode (0.93) is ~1.6 times as that in the former battery and the specific area capacity ratio of anode/cathode is decreased to be ~2.5. The corresponding cycling performance is presented in Fig. S3. In the initial cycle, the battery shows discharge potential of 2.3 V and charge potential of ~3.0 V (Fig. S3a), respectively. The low overpotentials indicate the high reversibility. This is similar to the former Li ion O_2 battery. For the cycling stability, the battery can run for ~60 discharge-charge cycles. In the 60^{th} cycle, the discharge capacity fades to less than 1000 mAh g^{-1}, possibly indicating the insufficient amount of Li^+ in the battery. This is a little less than the former battery with higher mass loading of anode. In the operation of Li ion O_2 battery, the discharge and charge capacities are fixed to be 1000 mAh g^{-1} based on the weight of the active materials in cathode. The lower mass loading of anode means more Li^+ from the lithiated Si anode has to be extracted/inserted and then involves the discharging and charging processes. This leads to the larger volume change of Si anode and increases the crack possibility of the built SEI film on F-L-Si anode. Partial Li^+ will be consumed to form new SEI film. The Li^+ in the Li ion O_2 battery derives from the pre-lithiated Si anode and its amount is fixed. During the long-term operation, the reduced amount of Li^+ results in the increased overpotentials of cathode and anode and then limits the cycling performance.