Supporting Information

A reversible Lithium-CO_2 battery with Ru nanoparticle as cathode catalyst

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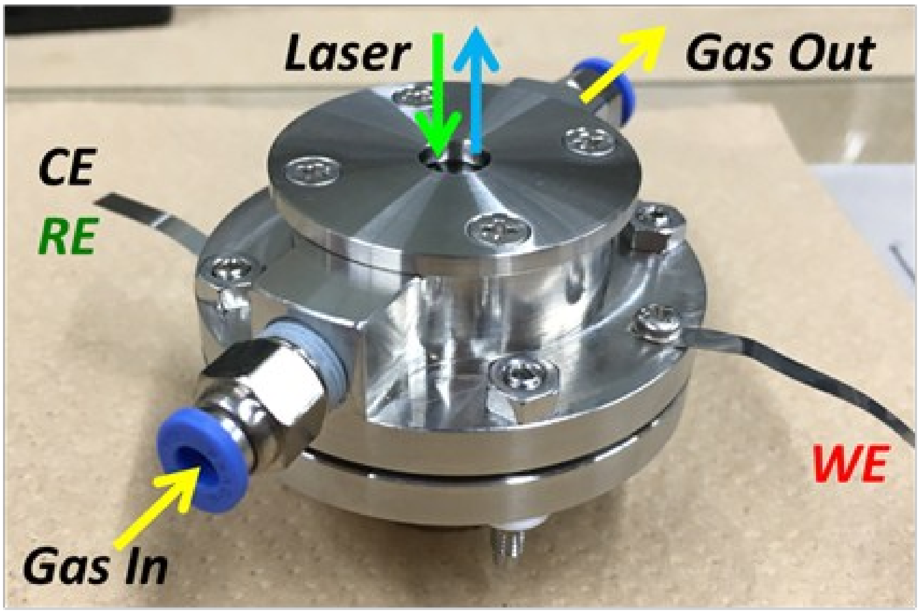
Content:



Scheme S1. Photograph of the homemade cell for in situ Raman spectra test.

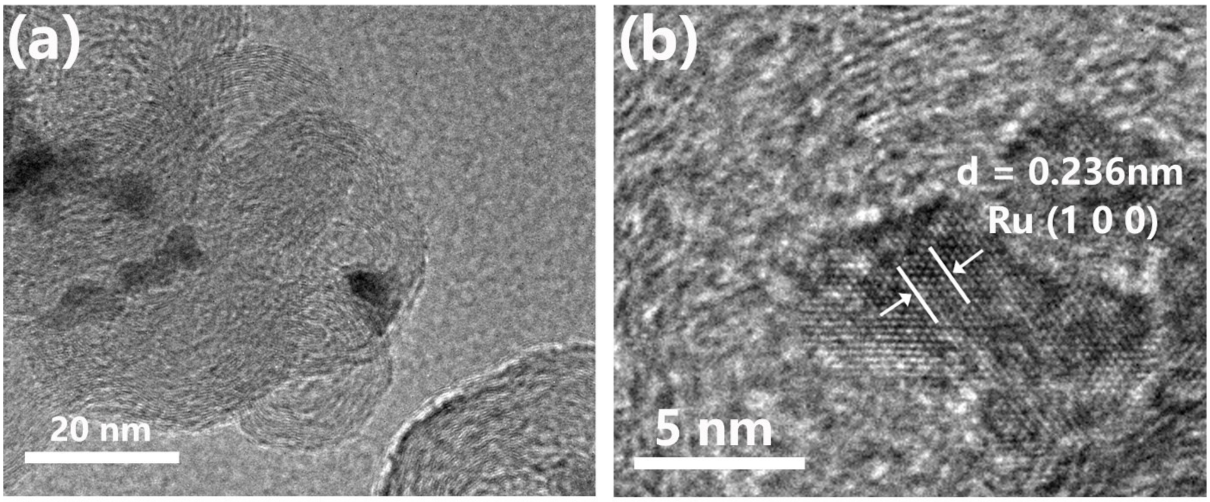


Fig S1. TEM image (a and b) of the Ru@Super P material.

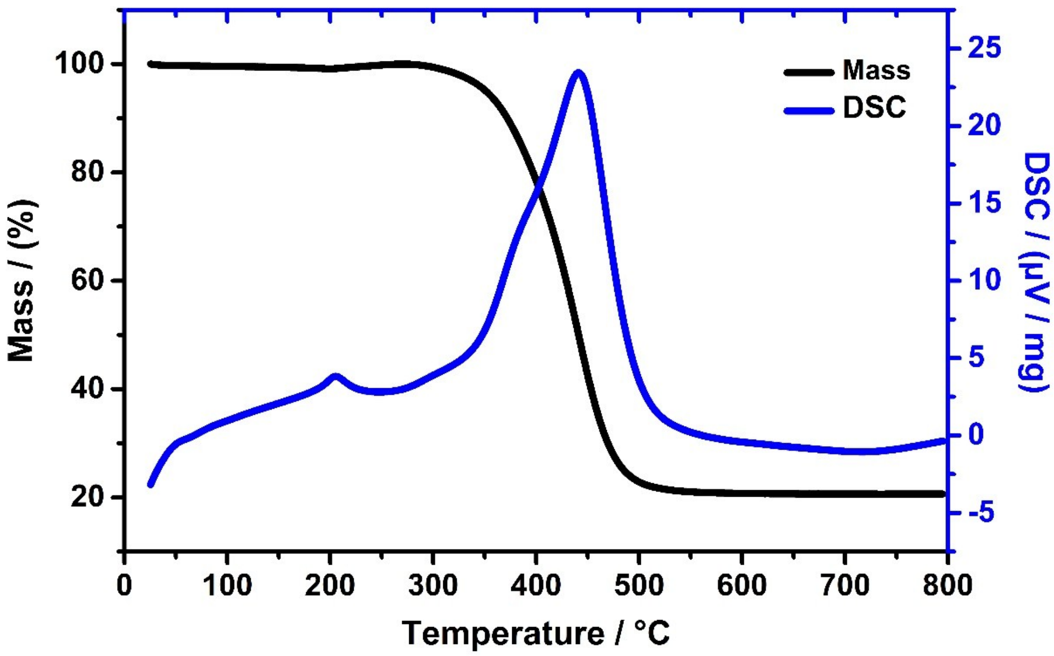


Fig S2. TG-DSC result of the Ru@Super P material. Temperature range: 25 ~ 800 ℃. Heating rate: 5 ℃ / min.

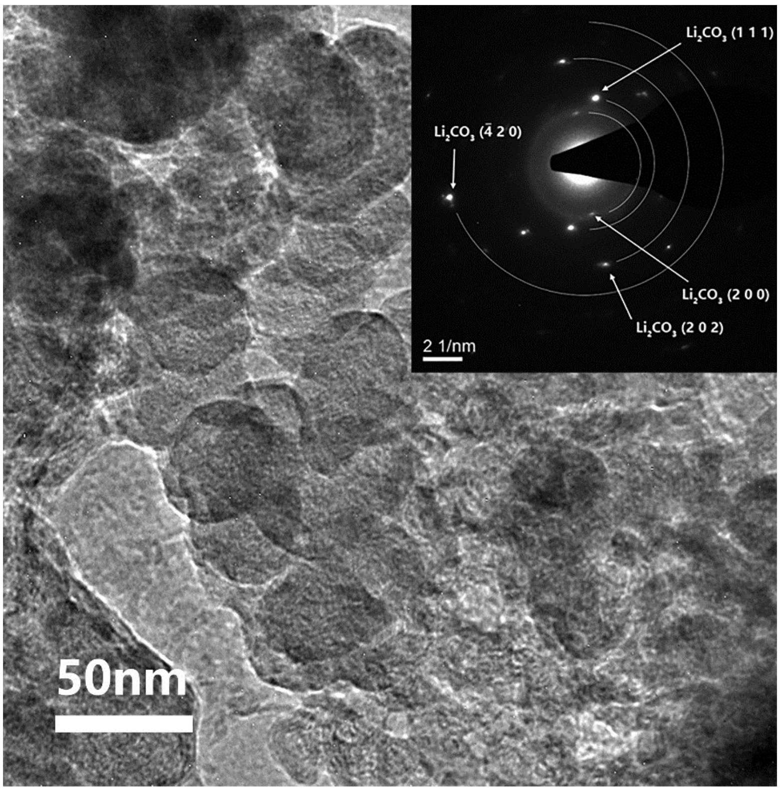


Fig S3. TEM image and SAED pattern (inset) of the discharged electrode

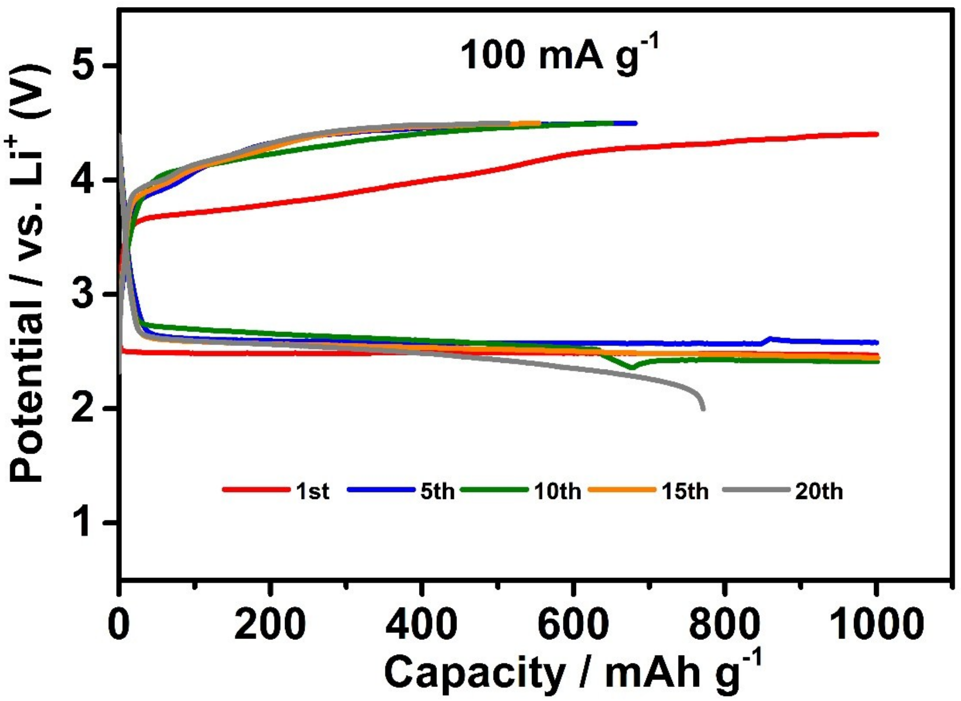


Fig S4 Cycle performance of Super P in $LiCF_3SO_3$-TEGDME (mole ratio of 1: 4) at the current density of 100 mA g^{-1}.

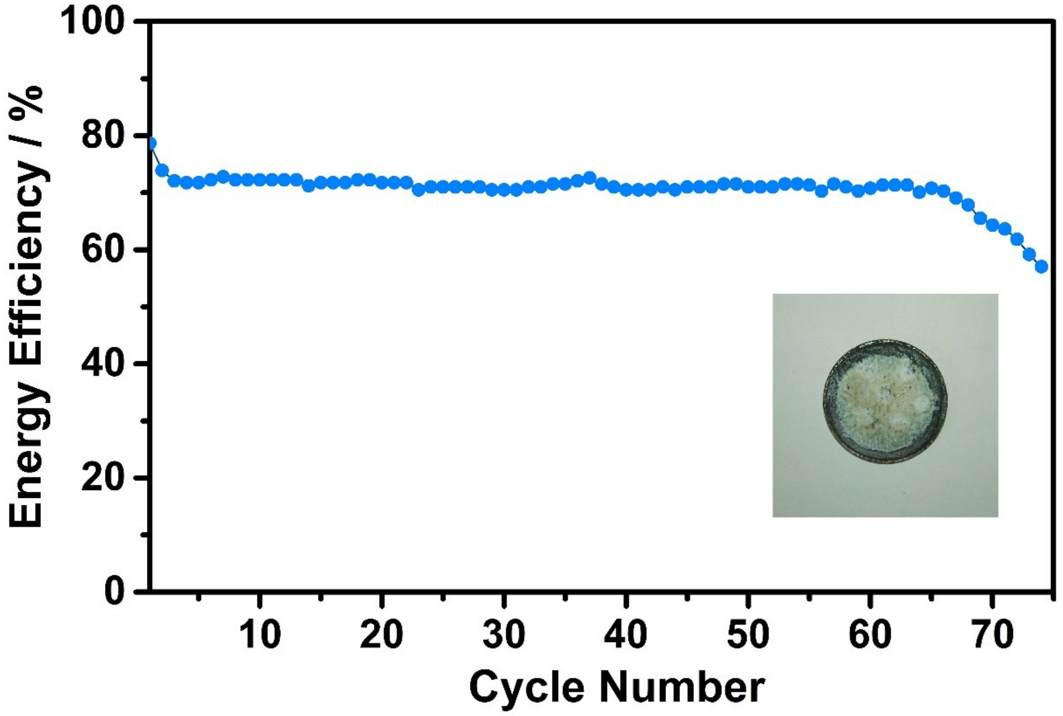


Fig S5. Energy efficiency of Ru@Super P in $LiCF_3SO_3$-TEGDME (mole ratio of 1: 4) at the current density of 100 mA g^{-1}.

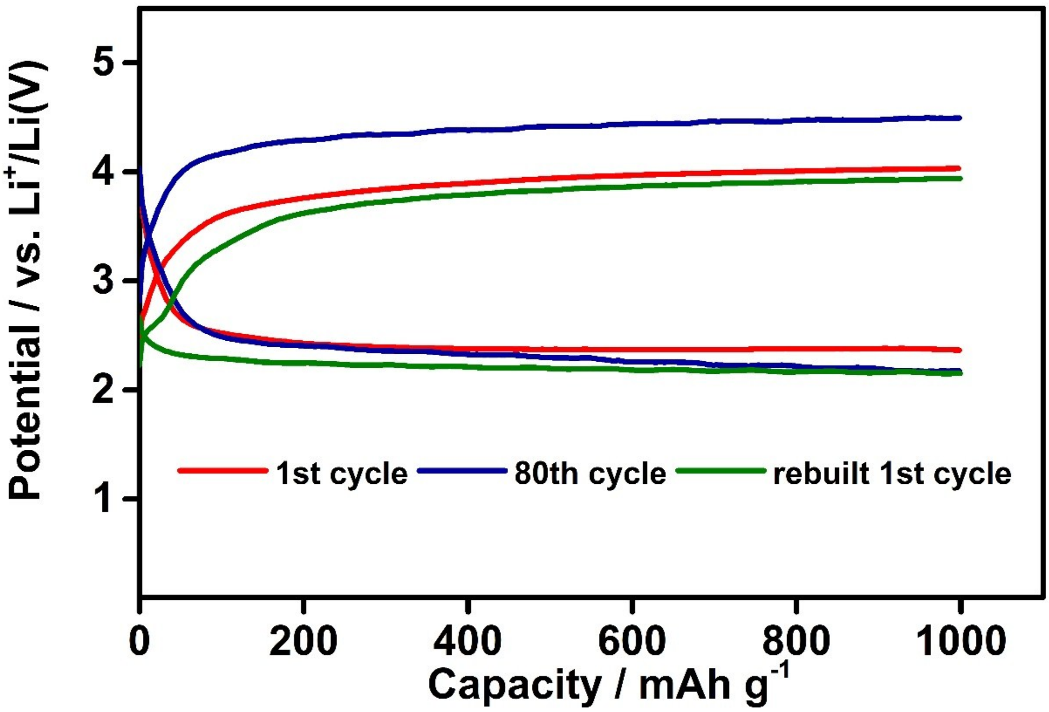


Fig S6. Discharge-recharge profile of a Li-CO_2 cell at the 1st cycle, 80th cycle and the 1st cycle after refilling electrolyte.

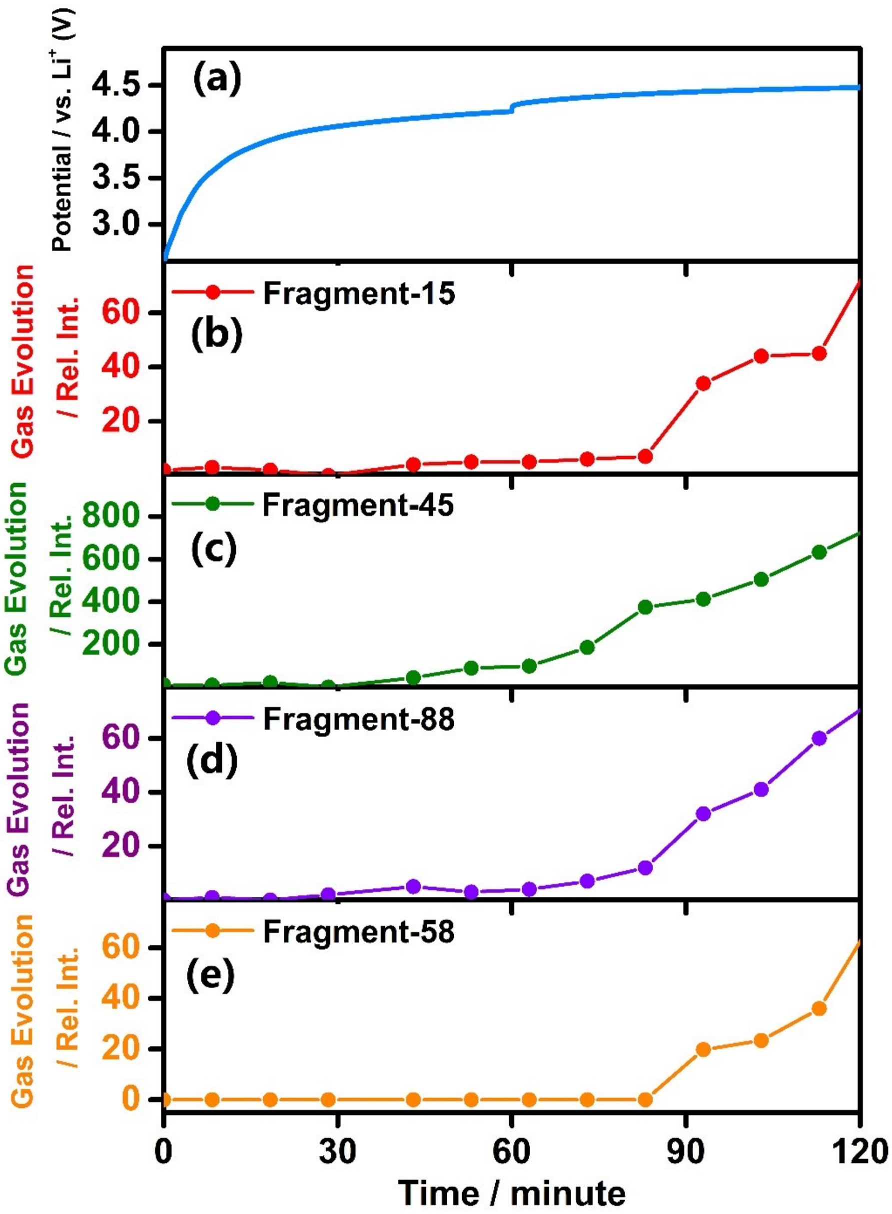


Fig S7. In situ GC-MS characterization of the charging process of a Li-CO_2 cell with Ru@Super P cathode at incremental current densities. **(a)** Charge profile of the Li-CO_2 cell. **(b) ~ (e):** Evolution of fragment-58 (represents the decomposition product of electrolyte solvent).

Calculation of Electrolyte Evaporation:

Electrolyte Evaporation Rate:

The evaporation rate of electrolyte can be estimated according to the equation proposed in Donald Mackay's work[1]:

$\boldsymbol{E_{mass}}$ (μg m^{-2} h^{-1}) = 1464 × $\boldsymbol{P}$ (Pa) × $\boldsymbol{M}$ (g mol^{-1})

$\boldsymbol{P}$ and $\boldsymbol{M}$ represents the vapor pressure of electrolyte and the molecular weight of electrolyte, repectively. For tetraglyme, $\boldsymbol{P} \approx 1$ Pa [2], $\boldsymbol{M} = 222.28$ g mol^{-1} and the evaporation rate can be obtained as 32.54 mg m^{-2} h^{-1}.

The coin cell we used was drilled with 7 holes with a diameter of 4 mm each on the cathode side, thus the exposure area of electrolyte can be estimated to be at least 0.88×10^{-4} m^{-2}. Hence the actual evaporation rate of electrolyte in our cell was 2.86×10^{-3} mg h^{-1}. Considering that about 50 µL (50.45 mg) of electrolyte was added in each cell, we found that at least 73.7 % of the electrolyte solvent evaporated after 65 cycles of battery operation at 100 mA g^{-1} (operating time: 1300 hours). This result may lead to a decrease of the ionic conductivity of the Li-CO_2 cell.

References:

[1] Mackay D and van Wesenbeeck I, Correlation of chemical evaporation rate with vapor pressure, *Environ. Sci. Technol.*, 2014, **48**, 10259-10263

[2] Balaish M, Kraytsberg A and Ein-Eli Y, A critical review on lithium-air battery electrolytes, *Phys. Chem. Chem. Phys.*, 2014, **16**, 2801-2822