Supporting Information

A Rechargeable All-Solid-State Li-CO_2 Battery Using $Li_{1.5}Al_{0.5}Ge_{1.5}(PO_4)_3$ Ceramic Electrolyte and Nanoscale RuO_2 Catalyst

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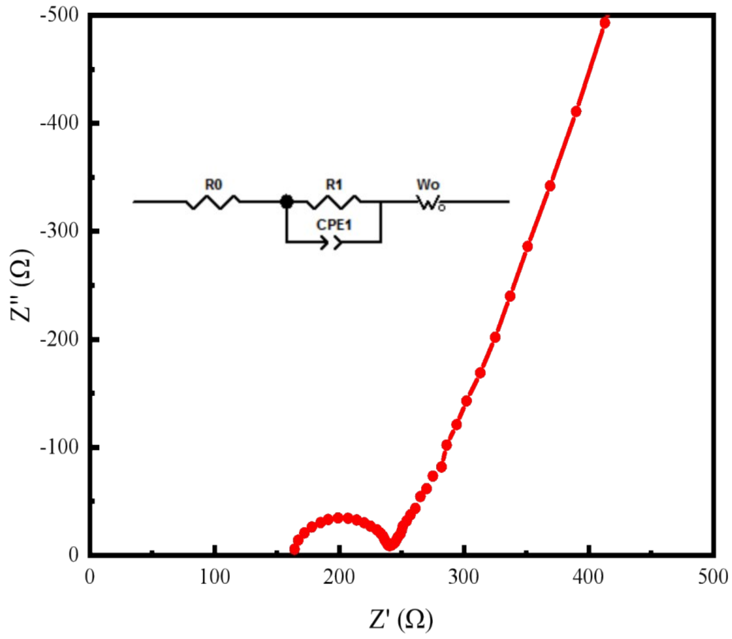

Figure S1. The impedance plots of LAGP pellet measured by an Au/LAGP/Au symmetric cell at room temperature

Electrochemical impedance spectroscopy (EIS) was tested with frequency ranging from 10^6 to 10^{-1} Hz at AC amplitude of 10 mV. The result of EIS was analyzed by Ziew software and then the ionic conductivity σ was calculated by the following equation:

$$\sigma = \frac{l}{R \times S}$$

(l: the thickness of electrolyte, R: resistance, S: the area of electrolyte)

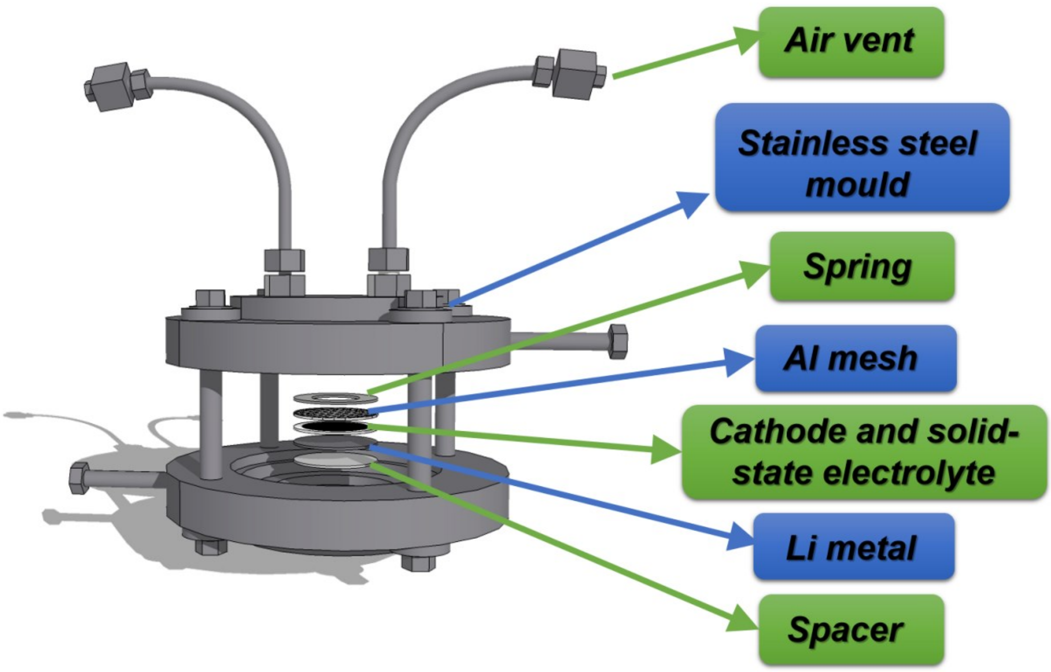

Figure S2. The illustration of the mould used in in-situ GC-MS measurement

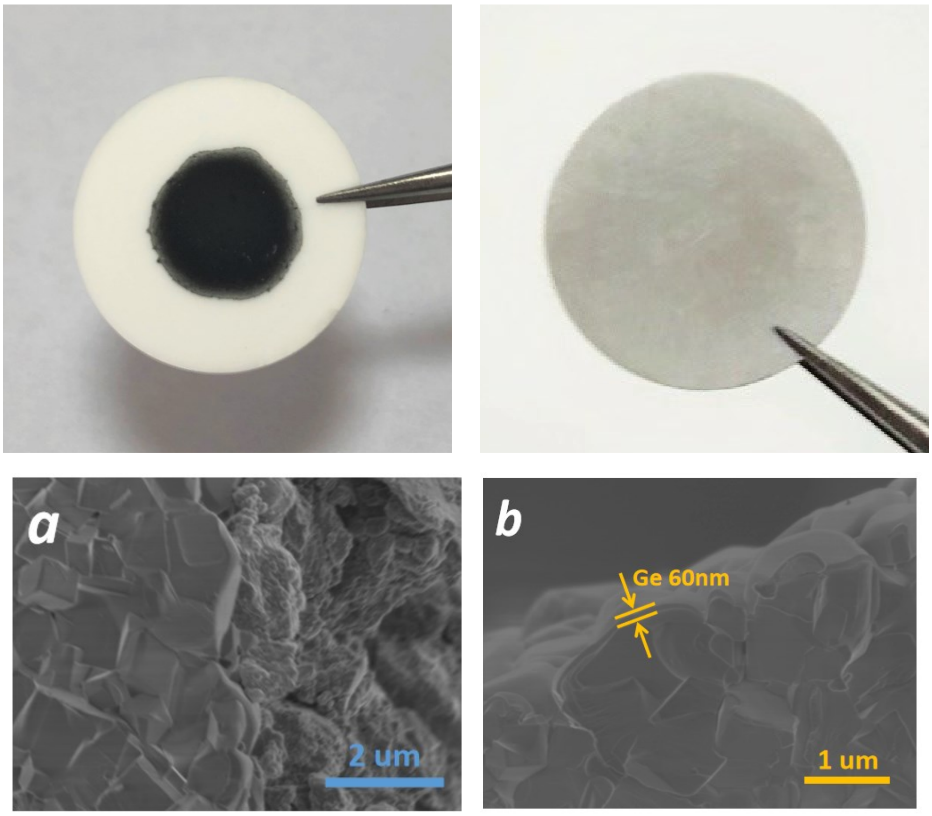

Figure S3. Snapshot and SEM images of (a) the cross-section part of the LAGP-cathode and (b) LAGP-Ge

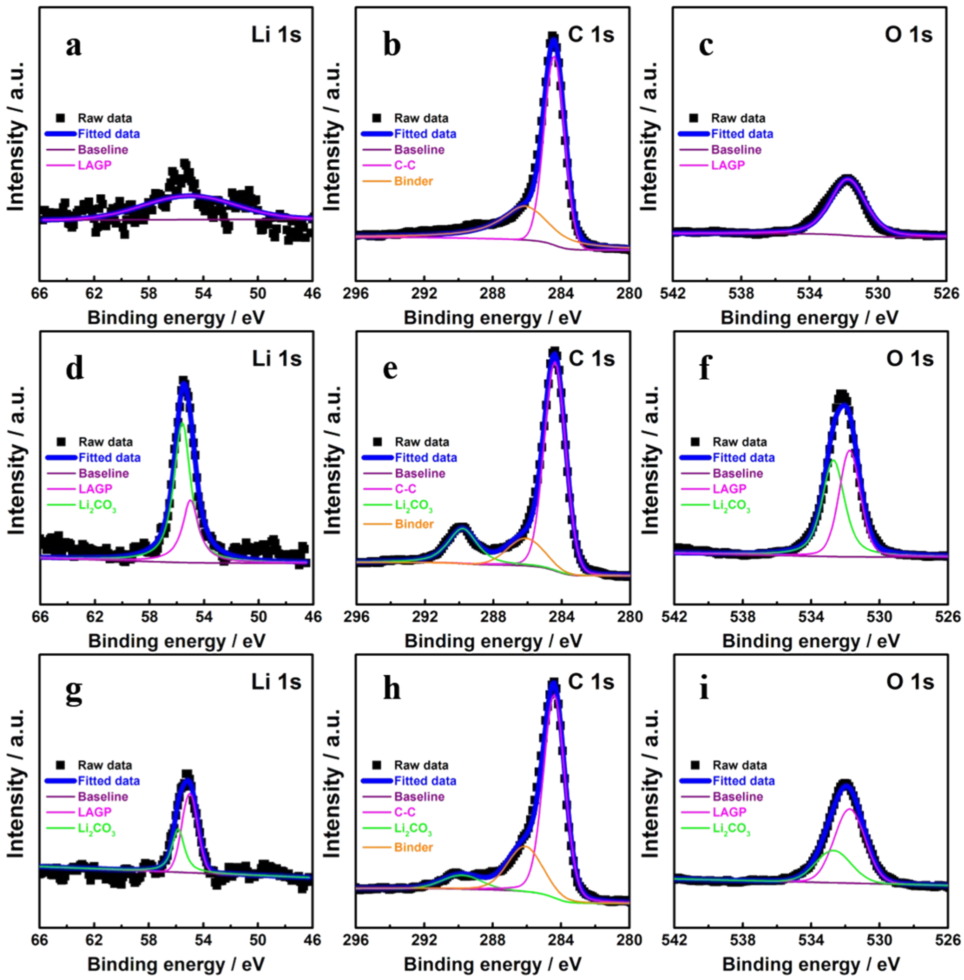

Figure S4 XPS spectra of the cathode materials (a-c)before discharge, (d-f) after discharge and (g-i) after recharge of the all-solid-state Li-CO_2 battery.

XPS analysis was also employed to characterize the discharge product. The peaks at 55.0 and 55.6 were assigned to LAGP and Li_2CO_3 in Figure S4 (a), (d) and (g). It can be clearly seen that the peak at 55.6 appeared and weakened when Li_2CO_3 was produced and deformed at discharge and charge state in Figure S4 (d) and (g), respectively. More obviously, the peak at 289.8 eV can be attributed to the C=O bond in Li_2CO_3 and those peaks at 284.4 eV and 286.0 can be ascribed to carbon nanotubes and binders, respectively. The Li_2CO_3 peaks notably increased in Figure S4 (e) and then reduced in Figure S4 (h). Besides, the peaks at 531.7 eV and 532.7 eV in Figure S4 (c), (f) and (i) were LAGP and Li_2CO_3, respectively. On the whole, the peaks assigned to Li_2CO_3 at 55.6 eV, 289.8 eV and 532.7 eV were simultaneously appeared and weakened corresponding to the formation and decomposition of Li_2CO_3 during the electrochemical process in the all-solid-state Li-CO_2 cell.

The current used in GC-MS measurement was 2.4 μA. After integration, it is known that 446 nmol CO_2 gas was generated after 468 minutes. The chemical reaction of the charge process was $2Li_2CO_3 + C \rightarrow 3CO_2 + 4Li^+ + 4e^-$, so the quantity of electric charge can be calculated by the following equation.

$$Q_{power} = It$$

$$Q_{co_2} = \frac{4}{3} \times n_{co_2} \times N_A \times e$$

After calculating, the efficiency of power is

$$\eta = \frac{Q_{co_2}}{Q_{power}} = \frac{\frac{4}{3} \times 446 \times 10^{-9} \times 6.02 \times 10^{23} \times 1.6 \times 10^{-19}}{2.4 \times 10^{-6} \times 468 \times 60} = 84.99\%$$

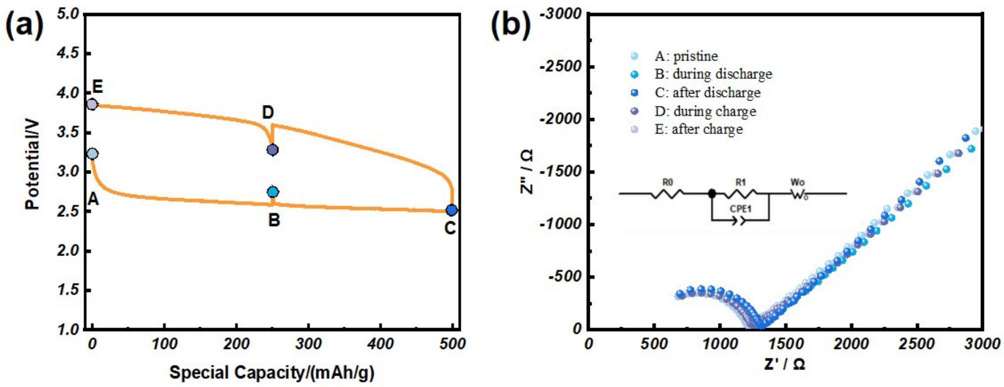

Figure S5. (a) charge/discharge voltage profiles and (b) electrochemical impedance plots at different stages of the Li-CO_2 cell

State	A	B	C	D	E
R0	381.3	369.4	406.3	392.6	375.1
R1	829.6	900.06	900.3	874.3	865.1
CPE1-T	1.804E-9	2.055E-9	1.398E-9	1.942E-9	2.149E-9
CPE1-P	0.87783	0.86289	0.8955	0.87114	0.86316
W0-R	39369	41197	22016	26835	47719
W0-T	14.65	24.88	4.635	7.285	21.11
W0-P	0.52148	0.51491	0.54291	0.51351	0.51809

Table S1. Equivalent Circuits of the Li-CO_2 cell at different stages

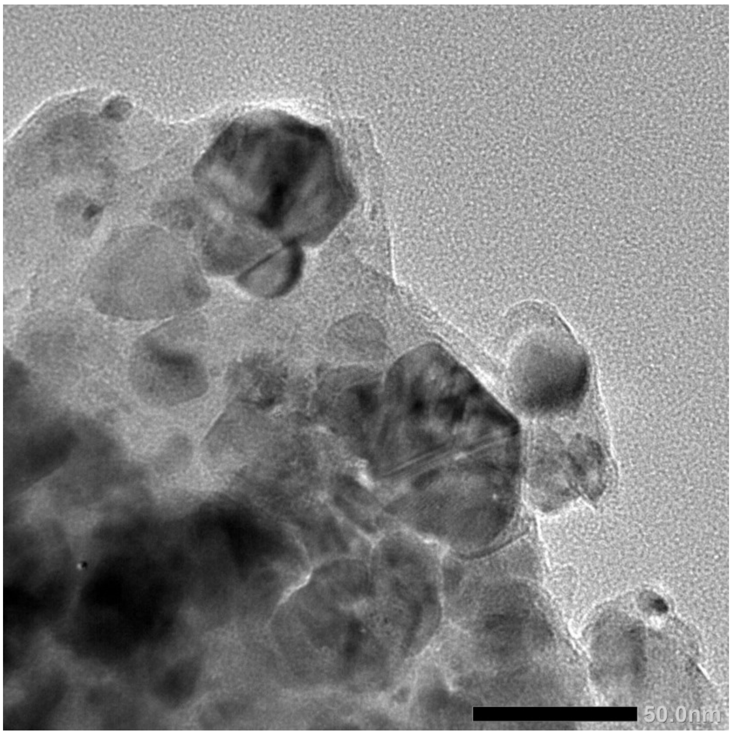

Figure S6. A partial enlarged picture of TEM image of the composite cathode material (the mixture of RuO_2, SWCNTs and LAGP)

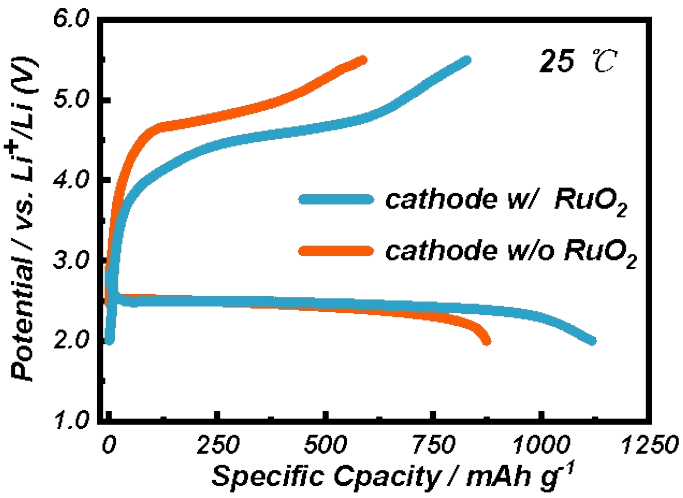

Figure S7. Discharge and charge curves of the Li-CO_2 batteries with/without RuO_2 catalyst at 25 ℃