Unveiling the mysteries of operating voltages of lithium-carbon dioxide batteries

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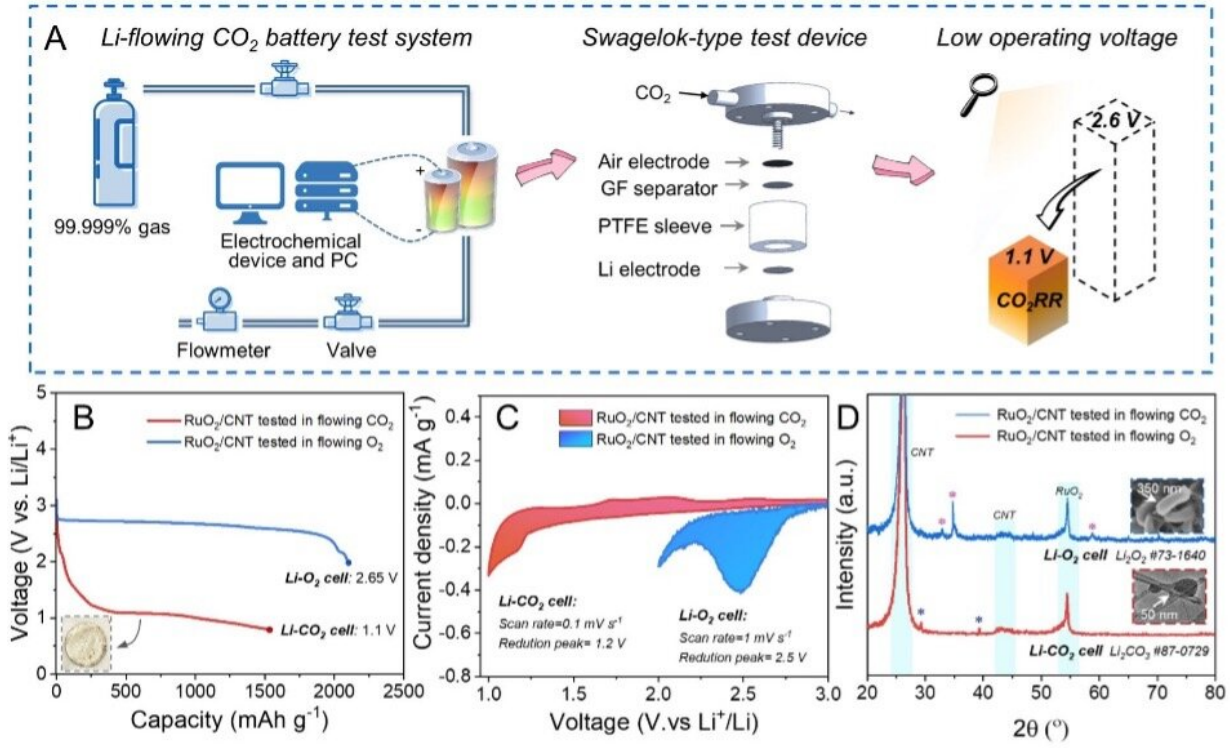

The schematics and performance of the test system. Credit: Xiao Xu et al.

A team led by Prof. Tan Peng from the University of Science and Technology of China (USTC) has widened our understanding of the operating voltages of lithium-carbon dioxide (Li-CO_2) batteries, providing a new strategy for the next generation of Li-CO_2 batteries. Their work was published in *Proceedings of the National Academy of*

Sciences.

Li-CO_2 batteries can turn CO_2 into carbonate and carbon while outputting electric energy, therefore possessing the advantage of both energy storage and CO_2 utilization. Previous studies generally reported that the operating voltage of Li-CO_2 batteries is about 2.6 V, which is similar to that of Li-O_2 batteries. However, this assumption has been facing increasing questions about whether slow CO_2 reduction reaction (CO_2RR) can generate such high voltages.

To cast light on the above question, Prof. Tan Peng's team built an electrochemical test system for the Li-flowing CO_2 battery, ensuring a pure CO_2 environment. The carbon nanotube (CNT) electrode, catalyst-loaded carbon nanotube (RuO_2/CNT) and non-carbon nanotube (RuO_2/NiO) all indicated that the Li- CO_2 battery operates at about 1.1 V and that the CO_2RR rate is much lower than the oxygen reduction reaction. The team determined the equilibrium potential to be about 2.82 V using galvanostatic current intermittent titration technique.

After analyzing the product, the team proposed that the discharge products at 1.1 V is a mixture of crystalline Li_2CO_3, amorphous Li_2CO_3 and amorphous C, verifying the four-electron transfer mechanism ($Li^+ + CO_2 + 4e^- \rightarrow Li_2CO_3 + C$). This mechanism theoretically predicts an equilibrium potential of 2.8 V, which is consistent with the test results.

The products analysis showed that the four-electron transfer proceeds slowly, compliant with the characteristics of low voltage system and inert CO_2.

Moreover, using a transmission electron microscope (TEM), the team found that under electron beam irradiation, small particles in the products began to grow through the phagocytosis of amorphous materials and started merging with other particles. In this process, amorphous

substance gradually transformed into a crystalline state. Therefore, the TEM image in some previous studies was probably not the natural discharge products but the products of electron beam irradiation.

To find out the source of the high voltage, the research team further investigated the effects of decoupled air components and operating conditions on the battery performance. Raising the voltage plateaus to 1.8-2.0 V by 1% O_2 and 500 ppm H_2O, the team didn't detect byproducts like LiOH and Li_2O_2 in the discharge products.

However, the morphology and crystallinity of Li_2CO_3 showed significant difference. O_2 and H_2O lowered the potential energy barrier and alleviated electrode passivation by changing the generation path of Li_2CO_3, thus accelerating the reaction and raising the discharge voltage plateaus. Based on the decoupling analysis, the slight air residue or leakage in the test device could lead to higher voltage plateaus and is extremely difficult to detect.

This work suggested that for the development of the next generation Li-CO_2 batteries, researchers need to conduct mechanism study in a pure CO_2 environment and develop compatible components like catalysts, electrolytes and electrodes.

More information: Xu Xiao et al, Unveiling the mysteries of operating voltages of lithium-carbon dioxide batteries, *Proceedings of the National Academy of Sciences* (2023). DOI: 10.1073/pnas.2217454120

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