Research on mechanism of electrochemical reaction at interface of Li-O_2 battery in Complex Systems

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Abstract:

The comprehensive performance of Li-air batteries is still far from practical application at present, including the cycle life, rate capability, energy conversion efficiency and so on. Further investigations on fundamental theories concerning Li-air batteries are required to promote these properties. The Li-air battery is a half-open system. During the battery operation, the process of material exchange proceeds between the open cathode and atmosphere. Since dry oxygen is incorporated into the research system, it is set as "battery-oxygen" system. During the process of discharge, the Li anode loses electrons forming Li^+ and oxygen acquires electrons and reacts with Li^+ to form Li_2O_2 at cathode. In the subsequent process of charge, it happens inversely to above reactions that Li_2O_2 decomposes at high voltage, producing O_2 and Li at cathode and anode, respectively.

The "dry oxygen" system has been studied thoroughly, of which the reaction mechanism at the three-phase interface between "electrode/electrolyte/oxygen" has been basically probed and the performance of electrode has been optimized progressively. Nonetheless, the dry oxygen system is only a simplified ideal research object to make the original complex open system accessible. Through the study of this simplified system, it is beneficial to clarify the electrode reaction mechanism of energy conversion and storage, design high performance catalysts and establish suitable research methods for air cells. However, limitations still exist in this system. Firstly, the electrochemical reactions at the solid-liquid-gas three-phase interface of other components in the air are not considered. Secondly, the "dry oxygen" simplified system contains the processes of oxygen reduction and evolution at the catalyst/electrolyte interface, ignoring a series of parasitic reactions resulting from the instability of electrode components. For example, the carbon materials and trace amounts of water in the electrode have been proved to participant in the electrode reactions. The study on the simple "dry oxygen" system has also achieved great achievements currently. And it should be further extended--introducing other constituents like CO_2, H_2O and N_2 from the atmosphere into the research system.

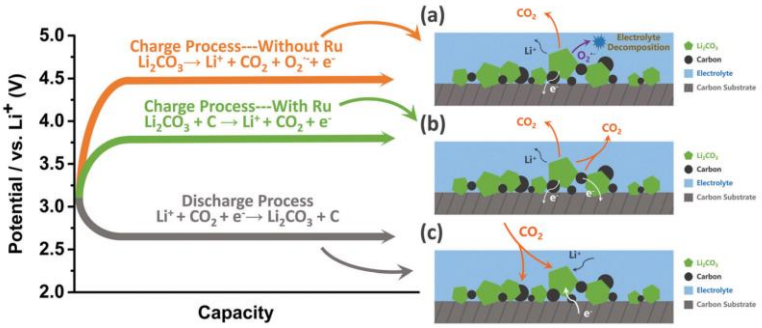

Figure 1. Schematic diagram of **(a)** reaction mechanism of the charging process of Li-CO_2 battery without Ru catalyst, **(b)** possible charging mechanism of Li-CO_2 battery with Ru catalyst and **(c)** the discharging process of Li-CO_2 battery.

In this report, other air components such as CO_2 and H_2O are introduced into the electrode/electrolyte interface system. The processes of CO_2 discharge and charge at electrode will be described in detail and the control of CO_2 reversible and irreversible processes has been

realized by Ru catalysts (as seen in Figure 1). Besides, the effect of H_2O in electrolyte and air on the discharge and charge performance of cell will also be present. And the new reaction mechanism caused by H_2O will be detailed. The great efforts have been made to focus on a series of parasitic reactions at the electrode/electrolyte interface and understand the complex electrode reactions at air electrode/electrolyte interface extensively. In the near future, the research system will be extended to the real atmosphere, and Li-air batteries will be put into practical applications gradually.

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Ping He was born in Nanjing, China, in 1981. He obtained his Ph. D. in Physical Chemistry from Fudan University in 2009, and later worked as a postdoctoral fellow at National Institute of Advanced Industrial Science and Technology (AIST) in Tsukuba, Japan. He joined the faculty of Nanjing University in Nov. 2011, where he is currently a Full Professor and deputy head of the Department of Energy Science and Engineering in College of Engineering and Applied Sciences. Prof. Ping He was awarded National Science Fund for “Outstanding Young Scholars”and Jiangsu Province Natural Science Fund for “Distinguished young Scholars”. In 2019, he was honored as “Young Scholar of Yangtze River Scholar Program” sponsored by Ministry of Education. His research interests focus on Energy storage and conversion materials, Energy chemistry and technology for advanced batteries including Li-ion battery, Li-Sulfur battery and Li-air battery. Prof. Ping He published more than 130 SCI peer-reviewed papers Such as *Joule, Angew. Chem. Int. Ed., Energy Environ. Sci., Adv. Mater., Adv. Energy Mater.*, *Nano Energy,* which were cited more than 7000 times. H-Index is 50. Prof. Ping He has presided over Topic of National key Research and Development Programs of China “New energy vehicle special project”, National Natural Science Foundation of China (3), and Natural Science Foundation of Jiangsu Province of China (3).