Electrocatalytic and Surface Morphology Effects on Carbon Supports for Li-O₂ Batteries

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Li-O₂ batteries attract great interest due to the high theoretical energy storage capacity. However, to approach the high capacity in rechargeable Li-O₂ batteries many challenges have to be solved. The most apparent problems are; (1) to control both the electro-catalysis of oxygen reduction and oxygen storage in desirable Li_xO_y layer (discharge) so that the deposited film can be easily decomposed, (2) to achieve feasible decomposition of the formed layer (charge) and (3) to ensure that conditions during charge and discharge do not lead to deterioration of an electrocatalyst or electrolyte. Up to date, most of the numerous studies in literature focus on an actual Li-O₂ battery consisting of all components. Solution of the above problems, however, may be very challenging without separating individual contributions to the complex system.

Carbon supports represent very important component for most electrodes up till now considered for $\text{Li}_x O_y$ storage in $\text{Li}-O_2$ batteries. The $\text{Li}_x O_y$ deposition likely consists of an electrocatalytic step to activate oxygen and actual $\text{Li}_x O_y$ storage and growth. Since carbon without any additional 'catalyst' seems to be also active in $\text{Li}-O_2$ storage, it is necessary to elucidate the role of carbon in non-aqueous media to understand possible synergetic effects on any mixed carbon-catalyst composites. Indeed, carbon can exhibit both the function of a catalyst and a $\text{Li}_x O_y$ depository. As known for electrocatalysis by carbon in aqueous alkaline media, different carbon surface functional groups or defects significantly influence electrocatalytic activity of carbon. For the storage function of the carbon, it is reasonable to expect that the same groups can act as possible $\text{Li}_x O_y$ growth nucleation sites. Carbon surface morphology is also expected to have an impact on $\text{Li}_x O_y$ growth and could influence its reversibility.

In this paper, various carbon supports are investigated and evaluated in Li-O₂ storage. Electrochemical treatment of carbon to create defects and oxidized functional groups is employed to study their possible catalytic function and impact on Li_xO_y growth. Morphology effects are evaluated by a comparative study on various carbons from highly oriented pyrolytic graphite (HOPG) toward high surface area carbons and 3D nanostructured carbons. Part of the electrochemical experiments is carried out in rotating disc electrode arrangement to allow better control of experimental conditions, but the high surface area carbons are also employed in an actual battery to enable quick comparison with a system with a high loading of carbon. The growth of the Li_xO_y films will be monitored by spectroscopic methods and a comparison with behavior of well defined Au and Pt surfaces will be provided to determine the nature and properties of the Li_xO_y layers. Reversibility of the Li_xO_y storage on high surface area carbon and a stability of solvents employed will be monitored by differential electrochemical mass spectrometry (DEMS).