Investigating the effect of the textural properties of zeolite-templated carbon air electrode on lithium-oxygen battery performance

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Lithium-oxygen (Li-O_2) batteries represent a promising next generation energy storage technology due to their high theoretical energy density (~3500 Wh kg^{-1}), which far surpasses that of conventional lithium-ion batteries (~300 Wh kg^{-1}) [1]. In a non-aqueous Li-O_2 battery, oxygen from the air reacts with lithium ions at the air electrode during discharge, forming lithium peroxide (Li_2O_2) through an oxygen reduction reaction which is reversed during charge. Despite their potential, Li-O_2 batteries face several critical challenges that must be resolved for the technology to gain commercial feasibility, including limited practical energy density, poor round-trip efficiency and cyclability, and capacity fading [2]. In this work, we demonstrate the influence of the porous structure of carbon electrodes on the formation and decomposition of Li_2O_2 during the operation of Li-O_2 batteries. To achieve this, zeolite-templated carbons (ZTCs) with different textural properties—conventional ZTC (ZTC) and House-of-cards ZTC (HZTC) with larger mesopores (assembly of 2D microporous carbon sheets)—were employed, marking, to the best of our knowledge, the first time ZTCs have been tested in Li-O_2 batteries. Using chemically similar carbons allowed us to unambiguously identify the influence of the carbon porosity on battery performance. Both ZTC and HZTC demonstrated significant specific capacities of 4012 mA-h g^{-1} and 3954 mA-h g^{-1}, respectively, at a current density of 100 mA g^{-1}. However, HZTC exhibited enhanced cycling stability and a notable improvement in both discharge and charge overpotentials, with values of -0.43 V and 0.51 V, compared to -0.74 V and 0.90 V for ZTC. Three-electrode EIS was used to systematically analyze the resistances that arise on the air and Li electrodes during discharge. This allowed us to provide valuable insights into the reaction interface and to establish a novel connection between the carbon microstructure and the charge and discharge overpotentials. The oxygen reduction reaction interface for both ZTC and HZTC changed from electrode/electrolyte to Li_2O_2/electrolyte as the discharge progressed and the charge transport resistance through the deposited Li_2O_2 particles was significantly reduced in HZTC (132.4 Ω) compared to ZTC (367.2 Ω) at a 40 % state of charge. This enhancement is attributed to the larger mesopores of HZTC, which facilitate the formation of smaller and less crystalline discharge products [3]. In addition, the batteries' poor rechargeability was primarily attributed to the irreversible discharge products formation. These findings underscore the critical impact of carbon porosity on optimizing air electrode performance and offer a pathway for designing high efficiency Li-O_2 batteries with improved performance and extended cycle life.

References

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Figures

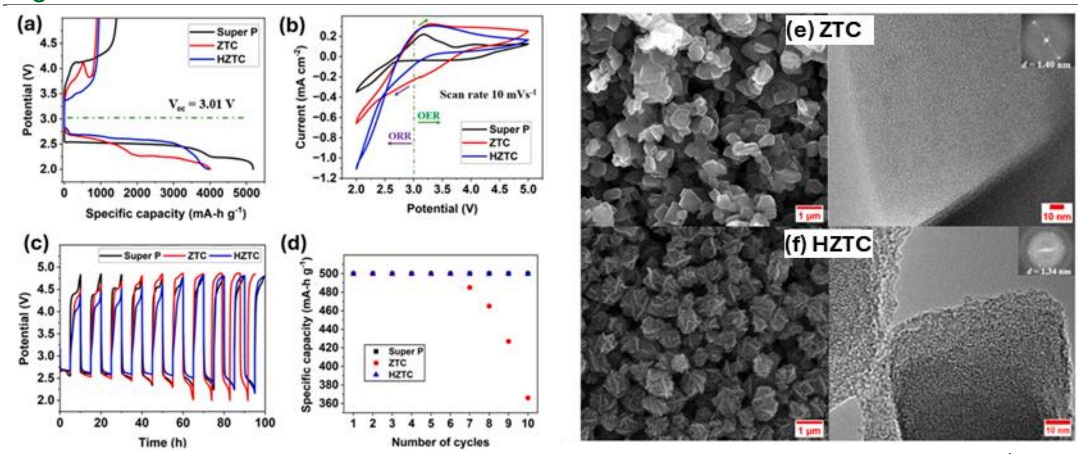

Figure 1: (a) Initial discharge/charge profiles for ZTC, HZTC, and Super P at 100 mA g^{-1} current density, (b) Cyclic voltammetry scans at 10 mV s^{-1}, (c) Discharge/charge profiles over 10 cycles at a fixed specific capacity of 500 mA-h g^{-1} and a current density of 100 mA g^{-1}, and (d) Changes in specific capacity over the 10 cycles. (e and f) SEM (left) and TEM (right) images of ZTC and HZTC, respectively. Insets in TEM images correspond to FFT of the images.