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Title	バイオベース窒素ドープカーボンにCoFe2O4ナノ粒子を担 持させたリチウム空気電池用両機能性電気化学触媒
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Abstract

This research work focuses on the utilization of a bio-based polymer-derived nitrogendoped carbon as a support for CoFe₂O₄ nanoparticles electrocatalyst in Li-air battery application. This work also showcases the electrocatalyst's bifunctional ability to perform robust electrochemical performances. Nowadays lithium-air batteries (LABs) have emerged as a promising prospect due to their unparalleled theoretical energy density of 3,505 Whkg⁻¹. However, despite the promising theoretical advantages, issues such as poor cycling stability, limited lifespan, and unresolved side reactions have hindered their commercial viability and widespread adoption. The major challenges of LABs are sluggish oxygen kinetics from oxygen reduction reaction (ORR) and oxygen evolution reaction (OER) are major issues that hinder their performance. Hence, developing electrocatalysts to promote ORR/OER is essential to reach the theoretical performance of LABs.

In this study, we introduced spinel cobalt iron oxide (CoFe₂O₄) nanoparticles decorated on bio-derived pyrolyzed poly(2,5-benzimidazole) (PYPBI800) as a nitrogen-doped carbon support electrocatalyst for LABs. The electrocatalyst was synthesized through pyrolysis of PBI followed by incipient wetness impregnation calcination on the metal precursors. The Strong Metal-Substrate Interaction (SMSI) between CoFe₂O₄ and PBI800 has been confirmed with X-ray photoelectron spectroscopy. Due to both the intrinsic properties of CoFe₂O₄ and the nitrogen doping effect, these electrocatalysts also modulate the electronic state of metals, facilitating oxygen adsorption and desorption at the electrode.

The electrocatalysts were evaluated as air-breathing electrodes in a CR-2032 coin-type cell LABs compared with other types of carbon supports. The initial discharge capacity for LAB coin cells with CoFe₂O₄ on PBI800 was observed to be 18,356 mAhg⁻¹. Furthermore, the cycling stability of the CoFe₂O₄ on PBI800 cathode was tested through 200 charge-discharge cycles at 400 mAg⁻¹ with 1,000 mAhg⁻¹ cut-off capacity. The resulting cycle-life data revealed that the battery maintains a discharge capacity as high as 100% even after the 200th cycle, demonstrating exceptional stability with overpotential remains consistently low at 140 mV throughout the examined cycle. The extensive surface area with N heteroatom defects of the PBI provides abundant nucleation sites for CoFe₂O₄ nanoparticles which serve as active sites for ORR and OER. The CoFe₂O₄ nanoparticles formed on the support exhibit strong metal-substrates interaction which leads to high exceptional stability and electrocatalytic activity.

Based on these findings, the $CoFe_2O_4$ nanoparticles on bio-based polymer-derived nitrogen-doped carbon suggested through this hold promise as a practical air-breathing electrode for high-performance rechargeable LABs. This work will also benefit future heteroatom-doped carbon support design, specifically nitrogen to enhance LABs performance.

Keywords: Oxygen Reduction Reaction, Oxygen Evolution Reaction, Metal Nanoparticles, Lithium Air Battery, Nitrogen Doped Carbon.