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博士の専攻分野の名称 博士（工学） 氏名 Sim Wei Jian

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Transition Metal-based Electrocatalysts for High Current Density Zinc-Air Battery
(遷移金属を用いた高電流密度亜鉛空気電池用電極触媒)

Rechargeable zinc–air batteries (ZABs) are promising next generation energy storage systems owing to their high theoretical energy density, intrinsic safety, and reliance on earth abundant zinc. Their potential to complement or replace lithium ion batteries in grid scale applications is compelling in the context of renewable energy integration. Yet sluggish oxygen reduction (ORR) and oxygen evolution (OER) kinetics, together with catalyst instability under high current densities, remain critical barriers to deployment. Chapter 1 outlines the history of ZABs, current challenges, and the experimental methods employed throughout this thesis.

To address these limitations, this work systematically explores advanced electrocatalyst design strategies, spanning carbon based composites, spinel oxides, perovskite oxides, and metal–organic framework (MOF) derived nanostructures. Each study builds upon the previous, collectively advancing understanding of structure–property relationships and their implications for high rate ZAB operation.

Chapter 2 investigates FeCo and CoNi oxide catalysts supported on reduced graphene oxide (rGO), tested under 100 mA cm^{-2} . This work demonstrated that nanocrystalline CoFe_2O_4 and CoO phases at optimal loading delivered $\sim 90\%$ capacity retention over 60 cycles, outperforming amorphous counterparts. The results also highlight the role of rGO in activating catalysts for high current density applications. Importantly, it revealed that conventional low-current electrochemical tests may not reliably predict full-cell behaviour, underscoring the need for practical testing conditions. Building on this, a second study investigated $\text{Co}_x\text{Ni}_{1-x}\text{O}-\text{NiCo}_2\text{O}_4/\text{rGO}$ composites, where the Co:Ni ratio and nanoparticle size were shown to critically influence bifunctional activity. At high current density, the equimolar CoNi/rGO catalyst exhibited superior cycling stability and ORR electron transfer efficiency, once again highlighting the synergy between spinel oxides and rGO supports.

In Chapter 3, the effect of noble-metal doping in LaNiO_3 perovskites, with Pt and Pd incorporated at the B-site via sol–gel synthesis is investigated. Pt-doped LaNiO_3 exhibited a 30% improvement in cycling stability at 60 mA cm^{-2} compared with pristine LaNiO_3 . Synchrotron-based X-ray absorption spectroscopy and Cs-corrected scanning transmission electron microscopy (STEM) confirmed Pt^{4+} substitution at the Ni site, inducing structural relaxation toward a cubic phase, enhancing Ni–O ionicity, and strengthening La–O covalency. These modifications collectively stabilised lattice oxygen redox processes, resulting in the improved ZAB cycling performance. This study provided mechanistic insight into noble-metal doping strategies and established a direct correlation between electronic structure and high-current ZAB performance.

Chapter 4 examined MOF-derived NiCo bimetallic nanoparticles encapsulated in nitrogen-doped graphitic carbon. Encapsulation was shown to effectively protect NiCo cores from oxidation, with STEM and EDS confirming uniform dispersion and complete encapsulation. Electrochemical cycling over 5000 cycles demonstrated greater activity retention for NiCo@C, significantly outperforming Co@C, highlighting the benefit of bimetallic nanoparticles. This study highlighted the MOF precursor synthesis method as a robust strategy to reliably synthesise bimetallic nanoparticles while suppressing oxidation and maintaining conductivity by means of N-doped C encapsulation.

In Chapter 5, the general conclusions would describe how these studies illustrate a coherent research trajectory: from carbon-supported transition metal oxides to noble-metal-doped perovskites, and MOF-derived encapsulated nanoparticles. Each study not only advances catalyst design principles but also emphasises the importance of evaluating performance under realistic high-current conditions. Moreover, the collaborative nature of

this work—spanning institutions in Japan, Thailand, and Taiwan— provides a global perspective on future challenges that lie ahead for sustainable energy storage.

By integrating mechanistic insights with practical battery testing, this thesis establishes design strategies for robust bifunctional electrocatalysts and reinforces the promise of zinc–air batteries as cost-effective, safe, and scalable solutions for renewable energy integration.