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Abstract

A plasma-in-liquid process is a versatile approach to synthesize nanomaterials. Sophisticated control of the interactions between the plasma and the surrounding liquid can lead to unique materials through the reactions of various chemical species occurring at the interface between the plasma and liquid. In this thesis, design strategies for carbon-based materials synthesized *via* the plasma-in-liquid process are explored. These strategies include structural optimization for specific applications of the pristine and heteroatom-doped carbon. In addition, advanced applications of the carbon-based materials synthesized through the above strategies, especially sodium-ion batteries (SIBs) and the oxygen reduction reaction (ORR), are examined.

First, this thesis proposes a simple plasma-in-liquid process to synthesize a sulfur-doped disordered carbon anode material for SIBs. SIBs have been heralded as a viable alternative to lithium-ion batteries because of the advantages of sodium over lithium, namely, its natural abundance and low cost. However, the practical deployment of SIBs has long been hindered by the radius of the sodium-ion, which is too large to store in conventional graphite, a widely used anode material. Hence, through the plasma-in-liquid approach, a new harmonized carbon material with a three-dimensional (3D) grapevine-like structure and a sulfur component is simply prepared, which affords a high reversible capacity at an extremely high specific current and long-term cycling stability with synergistic electrochemical performance. To substantiate these exciting findings, the physical, morphological, electrochemical, and electronic properties of the material are characterized.

Furthermore, the predominant 2-electron-transfer process for the ORR on the sulfur-doped disordered carbon is reported. The sulfur content doped in the unique carbon structure promotes oxygen adsorption of end-on types, which is supported by the excellent electrocatalytic activity leading to a 2-electron-transfer process. Although the electrocatalytic 2-electron-transfer process of various heteroatom-doped carbon materials has been reported, noble metals or stepwise syntheses are still required. Furthermore, the electrocatalytic 2-electron-transfer process on sulfur-doped carbon remains challenging. Given that sulfur-doped carbon investigations have focused only on the 4-electron-transfer process, and research on the 2-electron-transfer process has seldom been reported, it is anticipated that these findings will broaden the horizon on electrocatalytic behaviors.

Carbon-based ORR catalysts have been heralded as a viable alternative to noble metals because of their sustainable origin. Even though many studies on the plasma-in-liquid synthesis are focused on new materials and their outstanding performance, and materials such as various heteroatom-doped carbon are extensively reported, there has been a lack of insight into pristine carbon, which is the base material for such processes. We developed a partially crystalline porous carbon material with a high specific surface area, including various pore-size distributions, by discharging plasma through a single solution. This approach yields a pristine carbon material that affords a predominant 2-electron-transfer process and achieves a superior kinetic current density. We also characterized the structural properties of the material to substantiate these findings. It is expected that this work will provide useful references for the development of ORR catalysts that are low-cost and sustainable.

A sulfur-doped carbon material with an effective structure for multifunctional applications is finally demonstrated. Benefiting from enhanced reaction kinetics derived

from the C–S covalent bond, superior sodium-ion exchange was achieved, which resulted in a high reversible capacity, superior cyclability, and stable rate capability, thus demonstrating excellent performance for SIBs applications. In addition, this work further demonstrates the superior performance of the sulfur-doped carbon for the 2-electron-transfer ORR, which is important in a broad range of catalysis applications in many chemical engineering and environmental fields.

Although various heteroatom-doped carbon materials have been studied, insights into the underlying pristine carbon have been debatable; hence, we strived to establish useful references by analyzing structural information and evaluating the corresponding electrochemical performance. Furthermore, the synergistic contributions of the sulfur component with the unique structure constructed *via* the plasma-in-liquid process are theoretically and experimentally demonstrated. Thus, we hope that this thesis can provide novel strategies for the synthesis of carbon-based materials to facilitate their advanced applications with useful references for researchers working in this field.