

Abstract

The hydrogen economy has emerged as a promising solution to address the global energy and environmental challenges, with fuel cells playing a crucial role in enabling clean and efficient energy conversion. However, the widespread adoption of fuel cells, particularly in polymer electrolyte membrane fuel cells (PEMFCs), requires the optimization of catalyst materials to enhance performance, durability, and cost-effectiveness. One of the major challenges in fuel cell technology lies in the cathode oxygen reduction reaction (ORR), where carbon-supported platinum (Pt) nanoparticles are commonly used as the catalyst. However, providing sufficient durability of such catalyst nanocomposites remains a significant challenge. Under operating conditions Pt-based catalysts suffer from degradation due to electrochemically-induced Pt dissolution and carbon support corrosion. To address these challenges, this dissertation explores graphene derivatives as potential alternatives to current carbon black (CB) supports in order to improve the durability of PEMFC catalysts under operating conditions.

In the first part of the thesis we focus on a novel and innovative synthesis procedure to prepare graphene derivative-supported Pt catalysts. First we utilized the Hummers method to produce oxidized versions of graphene derivatives. Subsequently, we employed the pulse combustion method in combination with the double galvanic displacement method to prepare graphene derivative-supported Pt catalysts. This scalable method enables the production of multigram quantities of high-performance catalysts in high quantities, with high metal loading and electrochemically active surface area (ECSA).

Through accelerated degradation tests, these advanced catalysts were thoroughly examined for their electrochemical durability, surpassing the performance of traditional CB-supported catalysts. During the durability testing, we also conducted a thorough X-ray photoelectron spectroscopy (XPS) study to investigate the chemical properties of the carbon supports and correlate them with the observed durability trends. Furthermore, we tested the high current density performance in gas diffusion electrode (GDE) achieving performance comparable to commercial reference and record performance in the field of graphene derivatives.

To gain deeper insights into the durability of our catalysts, we conducted in-situ carbon corrosion measurements using an electrochemical cell coupled with a mass spectrometer (EC-MS). The results confirmed the enhanced durability of the graphene derivative-supported Pt catalysts, further validating their potential for application in fuel cells.

In the last part of the dissertation, we conducted a dopant study by introducing Boron (B) and Nitrogen (N) functionalities to the carbon support. This investigation aimed to enhance the catalyst's stability even further. The doped graphene derivative-supported Pt catalysts were subjected to durability testing. The results reveal unexpected negative effect on the durability of the graphene derivative-supported Pt catalysts. However, this initial observation has opened a new avenue for further investigation. To gain a deeper understanding of the impact of dopants on catalyst durability, additional comprehensive testing will be conducted in future research.

Our research has demonstrated the potential of graphene derivatives as advanced binary platinum catalyst supports in fuel cells. The innovative synthesis procedure based on the pulse combustion method has enabled the production of durable and high-performance catalysts in large quantities. The promising outcomes of this study open new horizons for the practical application of graphene derivatives in fuel cell technology, contributing to the advancement of clean and efficient energy conversion systems.