

## Carbon Nanotube-Based Catalyst Modification to Improve Proton Exchange Membrane Fuel Cell Interlayer Interactions

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**Abstract :** Optimizing the catalyst layer structure is crucial for enhancing the performance of proton exchange membrane fuel cells (PEMFCs) with low Platinum (Pt) loading. Current works focused on the utilization, durability, and site activity of Pt particles on support, and performance enhancement has been achieved by loading Pt onto porous support with different morphology, such as graphene, carbon fiber, and carbon black. Some schemes have also incorporated cost considerations to achieve lower Pt loading. However, the design of the catalyst layer (CL) structure in the membrane electrode assembly (MEA) must consider the interactions between the layers. Addressing the crucial aspects of water management, low contact resistance, and the establishment of effective three-phase boundary for MEA, multi-walled carbon nanotubes (MWCNTs) are promising CL support due to their intrinsically high hydrophobicity, high axial electrical conductivity, and potential for ordered alignment. However, the drawbacks of MWCNTs, such as strong agglomeration, wall surface chemical inertness, and unopened ends, are unfavorable for Pt nanoparticle loading, which is detrimental to MEA processing and leads to inhomogeneous CL surfaces. This further deteriorates the utilization of Pt and increases the contact resistance. Robust chemical oxidation or nitrogen doping can introduce polar functional groups onto the surface of MWCNTs, facilitating the creation of open tube ends and inducing defects in tube walls. This improves dispersibility and load capacity but reduces length and conductivity. Consequently, a trade-off exists between maintaining the intrinsic properties and the degree of functionalization of MWCNTs. In this work, MWCNTs were modified based on the operational requirements of the MEA from the viewpoint of interlayer interactions, including the search for the optimal degree of oxidation, N-doping, and micro-arrangement. MWCNT were functionalized by oxidizing, N-doping, as well as micro-alignment to achieve lower contact resistance between CL and proton exchange membrane (PEM), better hydrophobicity, and enhanced performance. Furthermore, this work expects to construct a more continuously distributed three-phase boundary by aligning MWCNT to form a locally ordered structure, which is essential for the efficient utilization of Pt active sites. Different from other chemical oxidation schemes that used HNO<sub>3</sub>:H<sub>2</sub>SO<sub>4</sub> (1:3) mixed acid to strongly oxidize MWCNT, this scheme adopted pure HNO<sub>3</sub> to partially oxidize MWCNT at a lower reflux temperature (80 °C) and a shorter treatment time (0 to 10 h) to preserve the morphology and intrinsic conductivity of MWCNT. The maximum power density of 979.81 mw cm<sup>-2</sup> was achieved by Pt loading on 6h MWCNT oxidation time (Pt-MWCNT6h). This represented a 59.53% improvement over the commercial Pt/C catalyst of 614.17 (mw cm<sup>-2</sup>). In addition, due to the stronger electrical conductivity, the charge transfer resistance of Pt-MWCNT6h in the electrochemical impedance spectroscopy (EIS) test was 0.09 Ohm cm<sup>-2</sup>, which was 48.86% lower than that of Pt/C. This study will discuss the developed catalysts and their efficacy in a working fuel cell system. This research will validate the impact of low-functionalization modification of MWCNTs on the performance of PEMFC, which simplifies the preparation challenges of CL and contributing for the widespread commercial application of PEMFCs on a larger scale.

**Keywords :** carbon nanotubes, electrocatalyst, membrane electrode assembly, proton exchange membrane fuel cell

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