CHEMICAL ENGINEERING

ENGINEERING OF NOVEL FUEL CELL CATHODE INTERFACES WITH SURFACE-INITIATED IONOMER FILMS BASED ON MODIFIED POLYNORBORNENE

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In this dissertation we describe a strategy to molecularly optimize the cathode side of a proton exchange membrane fuel cell. This new cathode consists of a porous gold membrane with pores that function as gas diffusion conduits while supporting a catalyst layer and ionomer. The ionomer chains are grown from the surface of the pore walls and are designed to encapsulate the catalyst particles to provide a well-defined three-phase boundary for gas, proton, and electron transfer. A new class of ionomer prepared by the surface-initiated ring-opening metathesis polymerization of a functionalized norbornene and subsequent sulfonation with acetyl sulfate is evaluated. These ionomers are grown rapidly and conformally over any surface geometry with nanometer-level control over thickness. The effectiveness of a high-utilization catalyst attachment method is demonstrated through electrochemical monitoring of the oxygen reduction reaction. A platinum submonolayer deposited onto the gold substrate is catalytically active in the oxygen reduction reaction, and the activity of the catalyst is preserved during the deposition of the sulfonated polynorbornene ionomer overlayer.

Nanoporous gold substrates are prepared through the exposure of a ~100 nm alloy sheet of gold and silver to nitric acid followed by binding onto a gold substrate. The nanoporous gold substrate demonstrates a 10x improvement in electrochemically active surface area available and is compatible with the deposition of a platinum monolayer. The incorporation of the sulfonated polynorbornene into the platinum-coated support decreases the oxygen reduction capacity of the electrode, likely due to poor oxygen permeability in the ionomer as well as reduced volume for oxygen transport within the nanoporous support.

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