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Enhanced Activity and Durability for the Oxygen Reduction Reaction on High Surface Area Multimetallic Aerogels

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Improvements of reactivity and durability of cathode electrocatalysts are still necessary for the world-wide implementation of practical polymer electrolyte fuel cells (PEFCs).[1,2] Most of the cathode electrode catalyst concepts are currently based on the utilization of Pt alloys and the increase of catalytically active surface area by using of highly dispersed nanoparticles supported on high surface area carbon. Despite the improved initial ORR activities on supported carbon nano-sized Pt-M catalysts, their long-term durability is largely limited by corrosion resistance of the support material (carbon oxidation and Pt detachment) and by (electro)chemical stability of the metals under operating fuel cell conditions. Considering these critical aspects, unsupported aerogels have emerged as a new promising generation of ORR electrocatalysts with special physical and chemical properties such as low densities, high porosity and large inner surface areas (BET surface areas up to 180 m^2/g) compared to conventional Pt-M nanoparticles.[3,4] Their unique properties are due to the particular linking of the specific properties of the nanomaterials magnified by macroscale self-assembly (network structure).[4,5]

In our study, we present new results for Pt mass- and surface area-based activities of various Pt-based multimetallic aerogel catalysts and compare those with pure Pt catalysts. For the structural and chemical investigations we used TEM, XRD, XPS and EDS techniques. The electrochemical experiments were conducted using Rotating (Ring) Disk Electrode (R(R)DE) technique. Durability investigations were performed on the catalytically active aerogels. The aerogel catalysts were subjected to two different voltage cycling test protocols, the “lifetime” regime (0.5–1.0 V vs. RHE) and the corrosive “start-up” regime (0.5–1.5 V vs. RHE).

Our collected data shows an ORR activity improvement for aerogels compared to Pt catalyst. The resulting ORR activities are correlated with the initial composition, size and BET surface area of the as-prepared aerogels. The electrochemical durability of the aerogels were traced out by monitoring of the catalytically active surface area and reactivity over the time and by the changes of particle size, network structure and chemical composition. Thereby, the aerogels showed an improved durability behavior in both cycling test protocolls compared to Pt/C catalyst. The durability benefits of aerogels are based on their highly specific surface area, long-range particle network structure with high porosity and the lack of support materials. Our work provides a new generation of cathode electrocatalysts with enhanced catalytic activity and durability for PEFCs.

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