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Dealloying and Annealing Optimization of High Mass Activity Pt_3Ni_7/NSTF ORR Cathodes for PEMFCs

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PEM fuel cell systems for automotive traction applications have stringent requirements of cost, durability, efficiency, power density, and operational robustness. While significant progress towards commercial viability continues, the membrane electrode assembly (MEA) cathode electrocatalyst is likely still insufficiently active and durable, requiring high precious metal loadings to achieve performance and durability targets (but not cost).

Membrane electrode assemblies (MEAs) utilizing the ultra-thin (<1µm) 3M nanostructured thin film catalyst (NSTFC) technology have several demonstrated advantages compared to MEAs comprising conventional, relatively thick (~10µm) carbon-supported catalyst, such as increased durability towards start-stop(1) and voltage cycling(2), higher specific activity(3), and high specific rated power (3). We have previously reported (4) a Pt_3Ni_7 alloy NSTF catalyst, which as-made demonstrated ORR mass activities ranging from 0.2 to 0.4A/mg, depending upon fabrication process parameters. Mass activities exceeding 0.6A/mg_{PGM} were obtained by application of a 3M annealing process, which significantly exceeded the current U.S. Department of Energy (DOE) target of 0.44A/mg_{PGM}.

One significant issue with the Pt_3Ni_7 alloy is dissolution of Ni from the alloy results in contamination of the PFSA PEM and reduction in peak power sufficient to exclude its applicability for automotive traction applications. Simple acid washing of the catalyst results in modest improvement in the peak power, but the processing time (hours to days) is economically prohibitive. A second significant issue was that the very high mass activities obtained with the 3M annealing process were with catalyst PGM content which was too low (<0.1mg/cm^2) to provide the high absolute activity and MEA performance required (5).

Here, we report progress on these two issues. Recently-developed proprietary ex-situ dealloying treatments have resulted in significant improvements in processing time (minutes or less), recent advances in annealing process development have allowed demonstration of mass activities exceeding 0.48A/mg (Fig.1) with cathode PGM content of ca. 0.12mg/cm^2, compatible with the DOE 2017 total (anode+cathode) MEA PGM target of 0.125mg/cm^2. When integrated with optimized NSTF anode catalyst and flow fields, these improved Pt_3Ni_7/NSTF catalysts have demonstrated MEA inverse specific power densities ranging from 0.16-0.13g/kW at 0.67V (150-250kPa, respectively), approaching the DOE 2017 target of 0.125g/kW (Fig. 2). Additional characterization, including STEM (Fig. 3), RDE, and XRD, will be discussed.

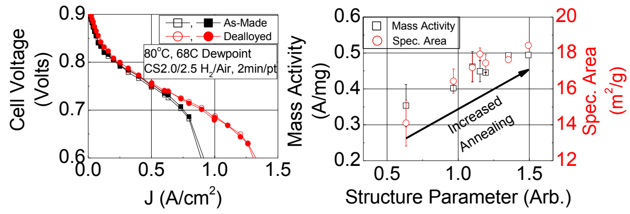

Fig. 1. (Left) Increased Rated Power After Improved Dealloying Process and (Right) Mass Activity, Specific Area Variation After Annealing. Pt_3Ni_7/NSTF Cathode with ca. 0.12mg_{PGM}/cm^2.

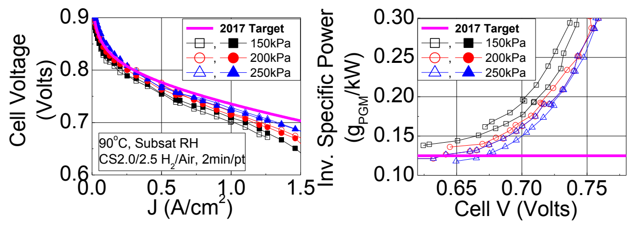

Fig. 2. H_2/Air Polarization and Inverse Specific Power at Various Reactant Pressures with 2013(March) Best of Class NSTF MEA. 0.137mg_{PGM}/cm^2 total (A+C).

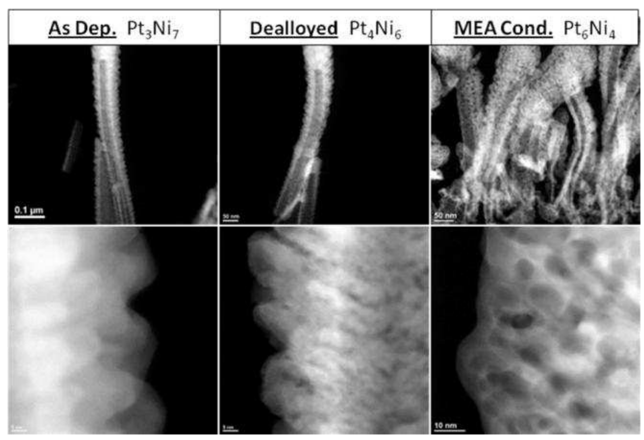

Fig. 3. High-Angle Annular Dark-Field (HAADF) STEM Images of Pt_3Ni_7 Catalyst: As Deposited, After Dealloying, and After MEA Conditioning.

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