

Impact of Catalyst Activity and Loading on Performance and Durability: Progress and Challenges Toward Automotive Targets

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Introduction

Recently, polymer electrolyte fuel cells (PEFC) have generated significant interest for automotive applications. Automotive competitiveness requires cathode (oxygen reduction reaction [ORR]) kinetic mass activities 4-fold higher than those of current state-of-the-art platinum catalysts [1] in order to achieve a cathode loading of 0.1 mgPt/cm^2 or a total of $0.2 \text{ g}_{\text{Pt}}/\text{kW}$ at a full-power potential above $\sim 0.6 \text{ V}$. A catalyst-related cell voltage loss less than $\sim 50 \text{ mV}$ over the entire current density range is sought over a >10 year automotive lifetime including $\sim 300,000$ large load cycles and $\sim 30,000$ start-stop cycles [2].

Experiments with MEAs of varying anode and cathode catalyst loadings in H_2/air demonstrate that the anode catalyst loading in state-of-the-art MEAs operating on pure H_2 can be reduced to 0.05 mgPt/cm^2 without voltage loss or negative impact on durability, while the cell voltage loss upon a reduction of the cathode catalyst loading for conventional Pt catalysts has shown initial voltage losses consistent with oxygen reduction reaction kinetics, with voltage losses significantly increasing throughout run time.

There are two pathways of work ongoing towards meeting automotive catalyst targets. One is towards the viability of a 4x mass activity catalyst. The other path is on understanding the catalyst properties that can affect the rate of electrode degradation with studies in areas such as carbon support type, effect of alloying, and stability of mass and specific activities.

Materials and Methods

All catalyst-coated membranes (CCM) were prepared in-house by an identical procedure [3]. Air or pure O_2 were used as cathode reactants and pure H_2 as anode reactant (all gases of 99.99% purity). For short-stack testing, 400 cm^2 active-area MEAs with different catalyst loadings and catalyst types were tested in the same stack with groups of at least 4 MEAs per type.

Results and Discussion

The area of high mass activity catalysts has been focussed on Pt-alloy binary and ternary compositions. However, conventional methodologies and combinatorial approaches [4] have yielded at best only modest reliable activity improvements beyond the 2.6x Pt activity typically achieved for the most-commonly-used Pt-alloy system, Pt-Co. The degree of activity enhancement of alloys when supported on corrosion resistant supports has been more difficult to ascertain. Some data suggests that some catalysts on corrosion resistant support do not maintain the alloy mass activity improvement (Figure 1).

Is the enhancement of activity by alloying sufficiently durable to be used in automotive applications? Short-stack testing under steady-state and load-cycling protocols have been performed on MEAs prepared with a variety of Pt and Pt alloy catalysts from disparate sources on both conventional and corrosion-resistant carbons. Mass activities at 900 mV were derived

from periodically-acquired O_2 polarization curves and 1 kHz impedance data (for cell resistance correction).

Figure 1 plots the mass activities of catalysts vs. durability run time from several short stacks that ran various protocols. Independent of type of test conditions, alloy catalysts on conventional carbons started with about twice the mass activity of Pt alone and maintained that advantage through 1000hrs of run time (or approximately 50,000 load cycles for load cycling durability tests), but the alloys on corrosion-resistant carbons did not exhibit the expected alloy enhancement factors and only maintained the same activity as pure Pt. What happens to Pt alloy MEAs beyond 1000hrs is still unclear.

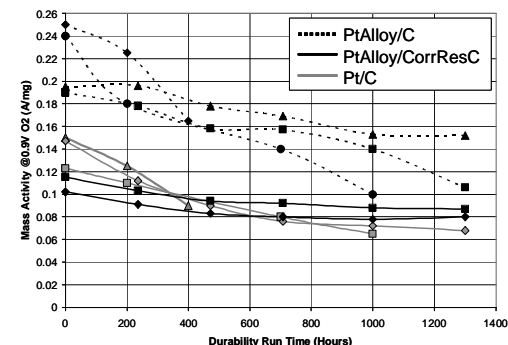


Figure 1. Mass activities of Pt-Alloy vs. Pt Catalysts from various durability short stacks.

Significance

More studies need to be done to understand what drives and limits the activity of catalysts. Can the 4x mass activity goal be attained as suggested by several studies in the literature that give evidence that control of structural, rather than simply compositional, variables can provide more productive pathways towards higher mass activities [5,6]? If high initial mass activities are possible, will that activity maintain throughout an automotive stack life? The answers will help in determining the near term viability of automotive fuel cells.

References

- [1] H.A. Gasteiger, S.S. Kocha, B. Sompalli and F.T. Wagner, *Appl. Catal. B: Env.*, **56**, 9 (2005).
- [2] R. Makharia, S.S. Kocha, P.T. Yu, M.A. Sweikart, W. Gu, F.T. Wagner, and H.A. Gasteiger, *Electrochem. Soc. Transactions (X2 Symposium)*, Los Angeles Meeting (2006).
- [3] H.A. Gasteiger, W. Gu, R. Makharia, M.F. Mathias and B. Sompalli, "Beginning-Of-Life MEA Performance – Efficiency Loss Contributions" in: *Handbook of Fuel Cells: Fundamentals, Technology, and Applications*, Wiley: vol. 3, Chapter 46, 593 (2003).
- [4] Atanassova, P.; Sun, Y., *USDOE Annual Hydrogen and Fuel Cell Review*, (2005).
- [5] V.R. Stamenkovic, T.J. Schmidt, P.N. Ross and N.M. Markovic, *J. Phys. Chem. B*, **106**, 11970 (2002).
- [6] R. Adzic, J. Zhang, K. Sasaki, M. Vukmirovic, J. Wang and M. Shao, *DOE Hydrogen/Fuel Cell Program Annual Review Meeting*, project FC-09 (2006).