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Journal of Power Sources 173 (2007) 644–645
JOURNAL OF POWER SOURCES
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Reply to Letter to the Editor

Reply to D. Gruber's comment on my previous paper

First of all, I appreciate Dr. D. Gruber's comments on our work [1]. He raised the issue of my description of platinum (Pt)'s poor adherence to the substrate in the electrocatalyst fabricated by sputter deposition method in my previous paper [1]. I welcome his criticism and the opportunity it has given me to reassess our work. I would like to reply to his comments.

The sputter deposition method has been widely used for integrated circuit manufacturing, semiconductor and glass industries for decades due to its various advantages. The principles and mechanisms of this method have been well explained by D. Gruber himself. However, the fabrication of the PEMFC electrocatalyst using the sputter deposition method differs substantially from the case of semiconductor production and glass industries. Therefore, our focus on the sputter deposition method is concentrated on the PEMFC electrocatalyst fabrication.

The objective of using the sputter deposition method for PEMFC electrocatalyst fabrication is not the full coverage of the Pt deposition throughout the substrate surface or in localized zones, but rather the formation of efficient and stable active sites with low Pt loadings for good electro-chemical reaction. In addition, the substrate of the PEMFC electrocatalyst such as membrane or carbon cloth is very porous and its surface is geometrically complex, unlike silicon wafer and class. In addition, the PEMFC electrocatalyst is subjected to a lot of pressure due to the stacks and the system compression.

Regarding the poor adhesion of the Pt, I referred to Saha's reports [2]. I partially agree with his description of the poor or low adhesion of the Pt to the electrocatalyst substrate. However, I cannot accept his view on the greater probability of dissolution and sintering of the Pt deposits, because the PEMFC working temperature of approximately 80 °C is insufficient for sintering and dissolution.

As mentioned by Dr. Gruber, most of the energy generated from the sputtering device for deposition is used to release atoms from the target and increase the kinetic energy of the atoms for film deposition on the substrate. However, I think this explanation is insufficient to clarify that the used energy is directly contributed to enhance the adherence force and that the sputter deposition method forms a stronger adhesion of Pt to the substrate. Furthermore, unlikely wet processes such as thin film deposition [3] and electrodeposition [4], there are no additives or chemical agents to help the Pt strongly stick to the substrate in the sputter method. Therefore, in the application of the sputter deposition method for electrocatalyst fabrication, I considered a high possibility of Pt sticking to the substrate with poor or low adhesion force.

Dr. Gruber commented on the "strong adhesion of Pt" reported by O'Hayre et al. [5]. However, O'Hayre et al. did in fact comment on the possibility of low Pt adhesion using the sputter deposition method. Their paper clearly described that a sputter-deposited catalyst layer must satisfy two requirements: strong adherence of the catalyst layer to the membrane, in order to reduce ohmic losses, and the capability of supporting the high mechanical stresses produced during operation. Therefore, O'Hayre et al. conducted a qualitative adhesion test using a variety of adhesive tapes and scratch-tips with the thin sputtered Pt films and they claimed that Pt adhered strongly to Nafion. However, they did not quantitatively measure the thin Pt film adherence. I consider that such a quantitative experiment could have provided very good information on the adherence force of the Pt film fabricated by sputter deposition method.

Based on O'Hayre's description of the strong adherence, I recognized that he also worried about Pt's low adhesion force and I guessed that his expression "strong adhesion" could be explained not by the adhesion force of Pt-sputtered film being stronger than that achieved via other methods but rather being sufficiently strong to allow the PEMFC to work well. The observed physical weakness is directly related to the adhesion force of Pt and the level of Pt loadings.

In conclusion, if the Pt adheres to the substrate with sufficient strength to allow effective PEMFC operation, then the extent of the absolute adhesion force between Pt and the substrate is unimportant. The important point here is whether the adhesion force of Pt fabricated by sputter deposition is relatively stronger than that of other methods, including wet processes. However, few reports and papers have clearly investigated this question. I therefore expect further studies to be conducted in order to elucidate this matter.

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DOI of original article:10.1016/j.jpowsour.2006.12.051.

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4 April 2007

Available online 6 August 2007