

Effect of Micropore Structure of Carbon Support on CO Tolerance of Pt_2Ru_3 Anode Catalyst for PEFC

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1. Introduction

Carbon monoxide impurities in hydrogen produced by reforming of hydrocarbon cause significant degradation of the PEFC performance. Even platinum–ruthenium alloy with the highest CO tolerance [1] cannot accept H_2 with CO contamination higher than 100 ppm. The highly dispersed and high-alloy degree $\text{Pt}_2\text{Ru}_3/\text{C}$ was found to be much higher CO tolerance than that of commercial PtRu/C [2]; however, effect of carbon support was not clear. Carbon xerogel is attracting much attention for its high electrical conductivity, high surface area and mesoporosity; moreover, pore characters are tuneable by vary of synthesis condition [3]. In this work, effect of pore structure of carbon support on CO tolerance was investigated.

2. Experiment

Resorcinol-formaldehyde carbon was prepared at an R/C ratio of 1000, and degrees of activation were 37% and 53%, RC1000 ac37 and ac53, respectively. Pt_2Ru_3 was supported on RC1000 by rapid quenching method (30wt%Pt-25wt%Ru/C) [2]. The catalysts were characterized by XRD, BET surface area, TEM. The anodes were tested in 5 cm^2 membrane electrode assemblies (MEA). The CO tolerance experiments were performed at constant current density mode (0.2 A/cm^2) First, starting with pure H_2 for one hour and then introduce 100 ppm of CO in H_2 for 2 hours, followed by 500 ppm, 1000 ppm, and 2000 ppm in every 2 hours.

3. Results

RC1000 ac37 and ac53 have meso-micro pore structure with bimodal pore size distribution. With an increase in degrees of activation, only micropore increases with maintaining regular mesopore structure of RC1000, resulting in increase in BET surface area. Fig. 1 shows that $\text{Pt}_2\text{Ru}_3/\text{RC1000ac37}$ and $\text{Pt}_2\text{Ru}_3/\text{RC1000ac53}$ catalysts have smaller metal particle size than commercial $\text{Pt}_2\text{Ru}_3/\text{C}$ because of regular mesopore structure of RC1000. For $\text{Pt}_2\text{Ru}_3/\text{RC1000ac53}$ having larger portion of micropore structure, even the Pt_2Ru_3 particles are located very close to others but they are not sintered. RC1000ac53 support has stronger interaction with Pt_2Ru_3 particles than RC1000ac37 support.

As shown in Fig. 2, cell voltage at 0.2 A/cm^2 drops from 0.78 to 0.59 V (24.4%) for $\text{Pt}_2\text{Ru}_3/\text{RC1000ac37}$ while the voltage drops from 0.75 to 0.4 (46.7%) for commercial $\text{Pt}_2\text{Ru}_3/\text{C}$. $\text{Pt}_2\text{Ru}_3/\text{RC1000ac37}$ and $\text{Pt}_2\text{Ru}_3/\text{RC1000ac53}$ catalysts have superior CO tolerance to commercial $\text{Pt}_2\text{Ru}_3/\text{C}$. $\text{Pt}_2\text{Ru}_3/\text{RC1000ac37}$ has higher CO tolerance than $\text{Pt}_2\text{Ru}_3/\text{RC1000ac53}$. These results indicate that small Pt_2Ru_3 particles and lesser density of the micropores is important for CO tolerance.

These preliminary studies could lead to importance of metal support interaction, and carbon xerogel can be a

promising support for future development of CO tolerance catalyst for PEFC.

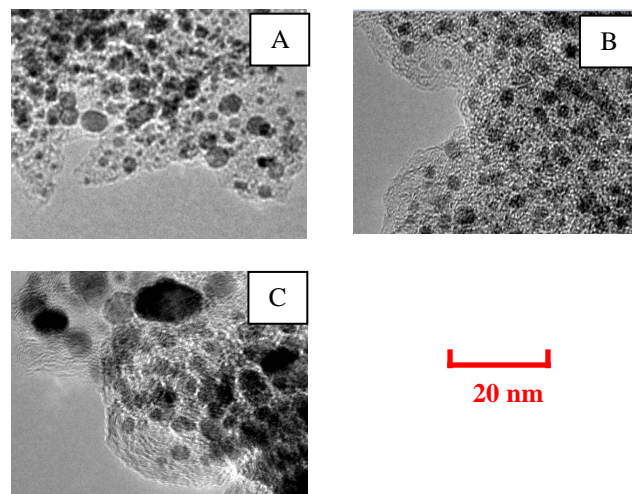


Figure 1 TEM images of (A) $\text{Pt}_2\text{Ru}_3/\text{RC1000 ac37}$, (B) $\text{Pt}_2\text{Ru}_3/\text{RC1000 ac53}$ and (C) Commercial $\text{Pt}_2\text{Ru}_3/\text{C}$

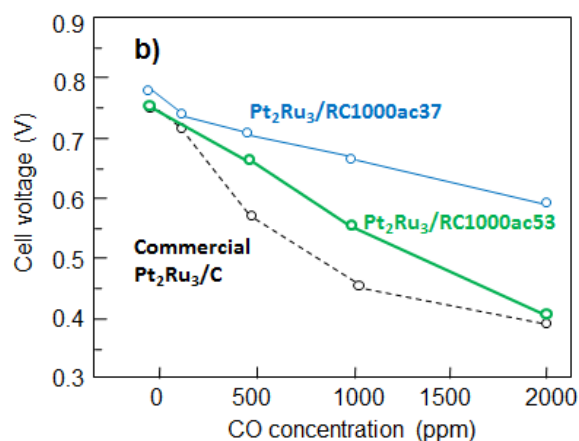


Figure 2 Effect of CO concentration on cell voltage at 0.2 A/cm^2 . Cell temp.: 70°C; Electrolyte: Nafion® NRE 212; Cathode: Pt/C (0.5 mg/cm^2); O_2 humidified at 70°C; Flow rate: 80 mL/min; Anode: $\text{Pt}_2\text{Ru}_3/\text{C}$ (0.5 $\text{mg-PtRu}/\text{cm}^2$); H_2 containing 0-2000 ppm CO humidified at 70°C; Flow rate: 80 mL/min.

References

1. M. Watanabe, S. Motoo, J. Electroanal. Chem., 60 (1975), p. 275-283
2. T. Takeguchi, T. Yamanaka, K. Asakura, E. N. Muhamad, K. Uosaki and W. Ueda, J. Am. Chem. Soc. 134, (2012), 14508–14512
3. K. Kraiwattanawong, H. Tamon, P. Praserttham, Microporous Mesoporous Mater, 138 (2011), p. 8–16

Acknowledgments

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