

Highly active tantalum oxide-based electrocatalysts toward oxygen reduction reaction for PEFC

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Introduction

Development of non-noble metal cathodes is required to commercialize polymer electrolyte fuel cells. We have investigated group 4 and 5 transition metal oxide-based cathodes. We reported that carbonitrides of tantalum, niobium, zirconium and titanium oxidized under low oxygen partial pressure had high catalytic activity for the oxygen reduction reaction (ORR) in acidic solution¹⁻⁴.

In this study, we focused on tantalum whose oxide had high stability in acidic solution. It is hard to prepare the fine particles by using carbonitrides as starting material because of large particle size of carbonitrides. Therefore, we tried to prepare nano-sized oxide-based particles by using new starting materials such as nitrogen-containing organic metal complexes.

Experimental

Tantalum carbonitrides powder (Ta_2CN) was heat-treated for 40 h at 900°C under N_2 containing 2% H_2 + 0.05% O_2 to prepare partially oxidized tantalum carbonitride powders ($\text{Ta-CNO}(\text{CN})$). As a new starting material, we used oxy-tantalum phthalocyanine (TaOPc). TaOPc was dissolved into solvent with multi-walled carbon nanotube (MWCNT) as a conductive material. The solvent was evaporated from the suspension to prepare the TaOPc supported MWCNT powder. The powder was heat-treated for 1 h at 900°C under N_2 containing 2% H_2 + 0.5% O_2 to prepare oxide-based catalyst powder ($\text{Ta-CNO}(\text{Pc})$).

All electrochemical measurements were performed in $0.1 \text{ mol dm}^{-3} \text{H}_2\text{SO}_4$ at 30°C with a 3-electrode cell. A reversible hydrogen electrode (RHE) and a glassy carbon plate were used as a reference and a counter electrode, respectively. A slow scan voltammetry was performed at a scan rate of 5 mV s^{-1} from 0.2 to 1.2 V under O_2 and N_2 atmosphere. A current density was based on the amounts of catalyst powder with carbon support. The catalysts were characterized by an X-ray diffractometer and Transmission Electron Microscope (TEM).

Results and discussion

Figure 1 shows the TEM image of the $\text{Ta-CNO}(\text{Pc})$. The catalyst particles were dispersed on the MWCNT. Compared with the particle of $\text{Ta-CNO}(\text{CN})$ (hundreds of nanometers)¹, the $\text{Ta-CNO}(\text{Pc})$ was one hundred times smaller. These fine particles had Ta_2O_5 structure according to the XRD pattern. These meant that the TaOPc , new starting material, was useful to prepare the oxide-based fine particles ($\leq 10 \text{ nm}$).

Figure 2 shows the potential - current curves for the oxygen reduction reaction of the $\text{Ta-CNO}(\text{CN})$ and the $\text{Ta-CNO}(\text{Pc})$ in $0.1 \text{ mol dm}^{-3} \text{H}_2\text{SO}_4$ at 30°C . The ORR current density, i_{ORR} , of the $\text{Ta-CNO}(\text{Pc})$ was approximately ten times larger than that of the $\text{Ta-CNO}(\text{CN})$ at 0.8 V vs. RHE. Therefore, the oxide-based fine particles of the $\text{Ta-CNO}(\text{Pc})$ had some ORR activity and probably the increase in the surface area of the oxide-

based compounds caused the increase in the ORR current.

From these results, we successfully prepared the oxide-based fine particles with high ORR activity by new starting material. It indicated that the TaOPc was better than the Ta_2CN as a starting material to prepare the high active ORR catalyst.

Acknowledgements

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Reference

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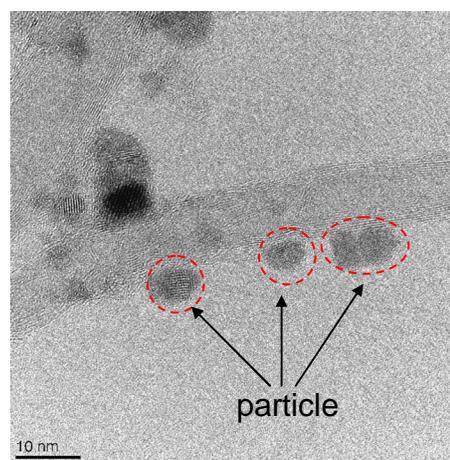


Fig.1 TEM image of $\text{Ta-CNO}(\text{Pc})$.

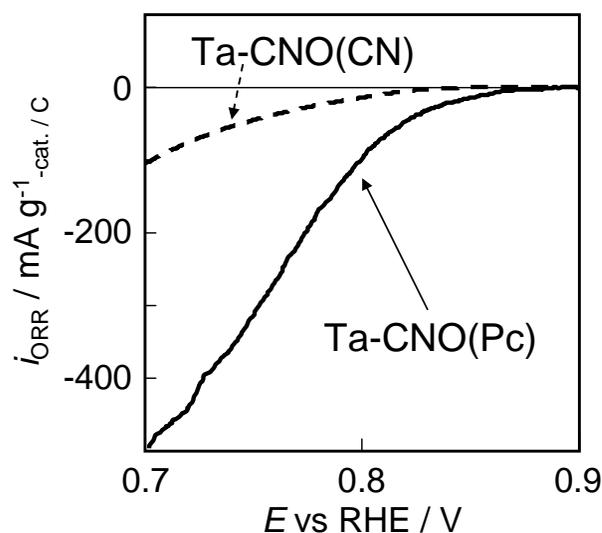


Fig2. Potential-ORR current curves of $\text{Ta-CNO}(\text{CN})$ and $\text{Ta-CNO}(\text{Pc})$.