Transition Metal Oxynitrides for New Cathode of Polymer Electrolyte Fuel Cell

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Introduction

Polymer electrolyte fuel cells (PEFCs) are expected to be used in the portable applications, especially to the automobile use, due to its high power density and low operating temperature. Although the theoretical efficiency is high, the actual efficiency is not so high due to the potential cathode overpotential. The cathode overpotential causes the large energy loss. An improvement of cathode materials should be needed to increase the energy conversion efficiency and to commercialize the fuel cell systems widely.

Highly dispersed platinum or platinum alloy on carbon powder are used as a conventional cathode catalyst at present. Though platinum is generally used as catalyst in many fields, its catalytic activity for ORR is not enough to obtain the sufficient energy efficiency. Furthermore, the estimated amount of the Pt resources was too small to make many fuel cell systems, especially fuel cell vehicles. Thus, in order to attain the popularity of the fuel cell systems in the future, the development of a non Pt catalyst is inevitable. We tried to apply transition metal oxynitrides such as TaON to cathode catalyst. TaON was known as visible light driven photocatalyst. However, the catalytic activities for oxygen reduction were unknown.

Experimental

TaON and TaN were prepared by heating Ta2O5 powder (Rare Metallic, purity: 99.9%) in alumina tube reactor under a flow of ammonia at 1123K for a certain hours. The crystallographic structure of the catalyst was analyzed by the XRD. Microanalysis of nitrogen content was performed using a Leco CHNS-932 elemental analyzer, and the surface analysis was performed by XPS.

After the 0.02g samples were added to 1cm³ distilled water by ultrasonic treatment, the solution of 3mm³ was picked up and dropped on the glass syringe carbon rod (Tokai carbon, diameter: 8mm). Then a diluted NaF solution was dropped on the samples to cover them and to keep them on the carbon surface.

The electrocatalytic activity for ORR and the stability in acid electrolyte (0.1mol dm⁻³ H2SO4) was evaluated by electrochemical measurements. A conventional 3-electrode cell was utilized, and the reversible hydrogen electrode (RHE) was the reference in the same solution. The cyclic voltammetry (scan rate: 100mV s⁻¹) was performed under nitrogen atmosphere to investigate the stability of prepared catalysts, and the slow scan voltammetry (scan rate: 5mV s⁻¹) was performed under oxygen atmosphere to examine the catalytic activity of ORR. A current density was expressed by geometric area. All experiments were performed at 303K (30°C) under the gas pressure of 1 atm.

Results and discussion

Figure 1 shows X-ray diffraction patterns of Ta2O5 and the prepared samples. β-TaON is a yellow to yellowish-green oxynitride that has the same structure as baddeleyite, monoclinic ZrO2. The structure of Ta2N5 consists of TaN5 octahedra and is similar to TiO2 (anovite). The XRD patterns corresponded to those of β-TaON and TaN, respectively. No impurity phase was observed in the XRD patterns. Figure 2 shows the slow scan voltammogram under oxygen atmosphere. Ta2O5, TaN and TaON except TaO0.85N1.05 (nitrogen content: 7wt%) had little catalytic activity for ORR. However, the reduction current of TaO0.85N1.05 (nitrogen content: 7wt%) for ORR was observed below about 0.8V vs. RHE. Because under nitrogen atmosphere such a reduction current was not observed, the reduction current was responsible for oxygen reduction. It was shown that the catalytic activity was very sensitive to bulk composition and/or surface state, because only TaO0.85N1.05 (nitrogen content: 7wt%) sample had some catalytic activity.

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