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Catalytic performance of iron group monometal and nanoalloy catalysts in electrochemical oxidation of ammonia

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1. Introduction

Ammonia (NH_3) is a high-hydrogen-content chemical and has attracted much attention as a carbon-free fuel applicable to fuel cells. Therefore, efficient electrooxidation of NH_3 is a challenge for the practical use of NH_3 fuel. It has been reported that Pt-loading electrodes exhibit the highest activity for NH_3 electrooxidation among electrodes covered with monometal catalysts and Pt-based binary or ternary catalysts show higher activities than those of pure Pt catalysts. From the view point of cost-saving and stable supply of catalyst materials, application of earth-abundant elements to electrocatalyst is a critical issue. However, there have been few reports on performances of the catalysts composed of these elements for NH_3 electrooxidation. Recently, we successfully synthesized an atomically-well-mixed Fe-group ternary nanoalloy (NA) catalyst1), namely, a carbon-supported FeCoNi (FeCoNi/C) NA, by a two-step reduction method. The FeCoNi/C NA catalyst showed the good affinity to an alcohol-group and the higher activity for electrooxidation of alcohol than that of monometallic Fe catalyst due to synergetic effects of constituent metals. Therefore, the FeCoNi/C NA catalyst is expected to show the good activity for electrooxidation of NH_3, that is a polar molecule similar to alcoholic chemicals. In this study, we synthesized a series of Fe-group catalysts including nanometals (NMs) and binary and ternary NAs and investigated their catalytic performances for NH_3 electrooxidation by cyclic voltammetry (CV) measurement.

2. Experimental

The Fe-group NM and NA catalysts were synthesized by the same procedure reported previously1). The catalysts were characterized by XRD, STEM, EDX and EELS measurements. All CV measurements were carried out in aqueous solution of 1 M KOH electrolyte in the presence and absence of 1 M NH_3 under Ar flow. Carbon felts (8 cm^2) modified with the catalysts (10 mg) were used as working electrodes. A coiled Pt wire and Hg/HgO electrode were used as a counter and reference electrode, respectively.

3. Results and discussion

Figure 1 shows the cyclic voltammograms for carbon-supported Pt (Pt/C) and FeCoNi/C electrodes. The anodic peak observed on the FeCoNi/C located at 0.55 V vs RHE, which is 0.3 V lower than that observed on Pt/C. The maximum current density using the FeCoNi/C was almost the same as that using the Pt/C, indicating that the FeCoNi/C catalyst shows the excellent performances for NH_3 electrooxidation. The CV measurements were carried out using the Fe-group NM and binary NA electrodes. Detailed comparison of the experimental results revealed catalytic roles of each Fe-group metal and alloying effects on the activities for NH_3 oxidation.

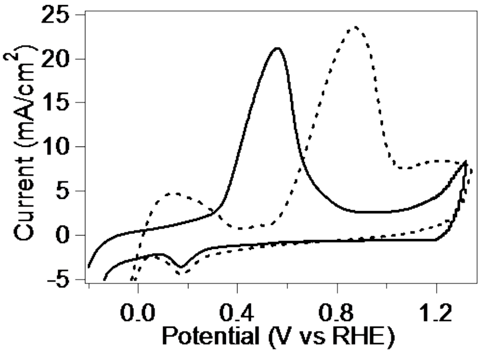

Figure 1 Cyclic voltammograms recorded on the FeCoNi/C (solid line) and Pt/C (broken line) as working electrodes in 1 M KOH aqueous solution in the presence of NH_3.

1) T. Matsumoto, M. Sadakiyo, M. L. Ooi, S. Kitano, T. Yamamoto, S. Matsumura, K. Kato, T. Takeguchi, M. Yamauchi, *Scientific Reports*, **2014**, *4*, 5620.