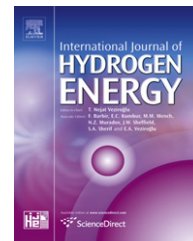


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Carbon nanotubes-supported colloidal Ag–Pd nanoparticles as electrocatalysts toward oxygen reduction reaction in alkaline electrolyte

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ABSTRACT

Ag–Pd nanoparticles with compositional ratios of 1:1 (Ag_1Pd_1), 2:1 (Ag_2Pd_1), and 4:1 (Ag_4Pd_1) and supported on multiwall carbon nanotubes (CNTs) were prepared by the self-regulated reduction of sodium dodecyl sulfate, and then, they were used as catalysts for oxygen reduction reactions (ORRs) in 1 M NaOH solution. Polarization curves showed that, among the prepared nanocatalysts, $\text{Ag}_4\text{Pd}_1/\text{CNT}$ nanocatalysts showed higher activity. During the ORRs, two types of oxygen coverages given by the Temkin isotherm and Langmuir isotherm were observed for low and high overpotentials, respectively. Koutecky–Levich plots showed that the number of electrons involved in the ORRs catalyzed by $\text{Ag}_1\text{Pd}_1/\text{CNT}$, $\text{Ag}_2\text{Pd}_1/\text{CNT}$, and $\text{Ag}_4\text{Pd}_1/\text{CNT}$ were 2.11, 1.88, and 2.25, respectively. These ORRs proceeded through a two-electron pathway. Polarization curve in the electrolyte with methanol revealed that $\text{Ag}_4\text{Pd}_1/\text{CNT}$ has high methanol tolerance during ORRs.

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1. Introduction

Carbon nanotubes (CNTs) are often used as a support for electrocatalysis owing to their high electrochemical stability and conductivity in cyclic energy conversion systems such as fuel cells [1–5]. When metallic nanoparticles supported on CNTs are used as anodic catalysts in fuel cells, they have been shown to exhibit great activity and stability toward the oxidation of methanol [1,2,5–8] and formic acid [9]. Furthermore, recently, CNTs have been directly used as cathodic catalysts in oxygen reduction reactions (ORRs) occurring at the oxygen electrode of fuel cells [10–12]. However, polarization often occurs at the oxygen electrode [13]. In order to reduce the effect of polarization on the electrode, it is necessary to enhance the activity of the electrocatalysts involved in the ORR. Ag nanoparticles are currently attracting much attention as ORR catalysts for application in alkaline fuel cells

[14–16]. In general, the activities of Ag catalysts are slightly lower than those of Pt catalysts in ORRs because of their weak interaction for binding O_2 . However, inexpensive Ag nanoparticles have been shown to have higher stability than pure Pt cathodes during long-term operation. Recently, Ag-based alloy nanoparticles, Ag–Co, have been reported to show potential in ORRs through above three-electron pathway [17].

Recently, based on self-regulated reduction in pure sodium alkyl sulfate micelles, we have sequentially reported a synthesis route for the preparation of Pd nanoparticles supported on various carbon nanotubes (Pd/CNTs) by using sodium n-tetradecyl sulfate (SC_{14}S) as a reducing agent and as an attaching agent in the reflux of $\text{SC}_{14}\text{S}/\text{CNT}$ solution [18]. In this study, Ag–Pd nanoparticles with compositional ratios of 1:1 (Ag_1Pd_1), 2:1 (Ag_2Pd_1), and 4:1 (Ag_4Pd_1) and supported on CNTs were prepared using sodium n-dodecyl sulfate (SDS) in a manner similar to Pd/CNTs. The electrochemical properties

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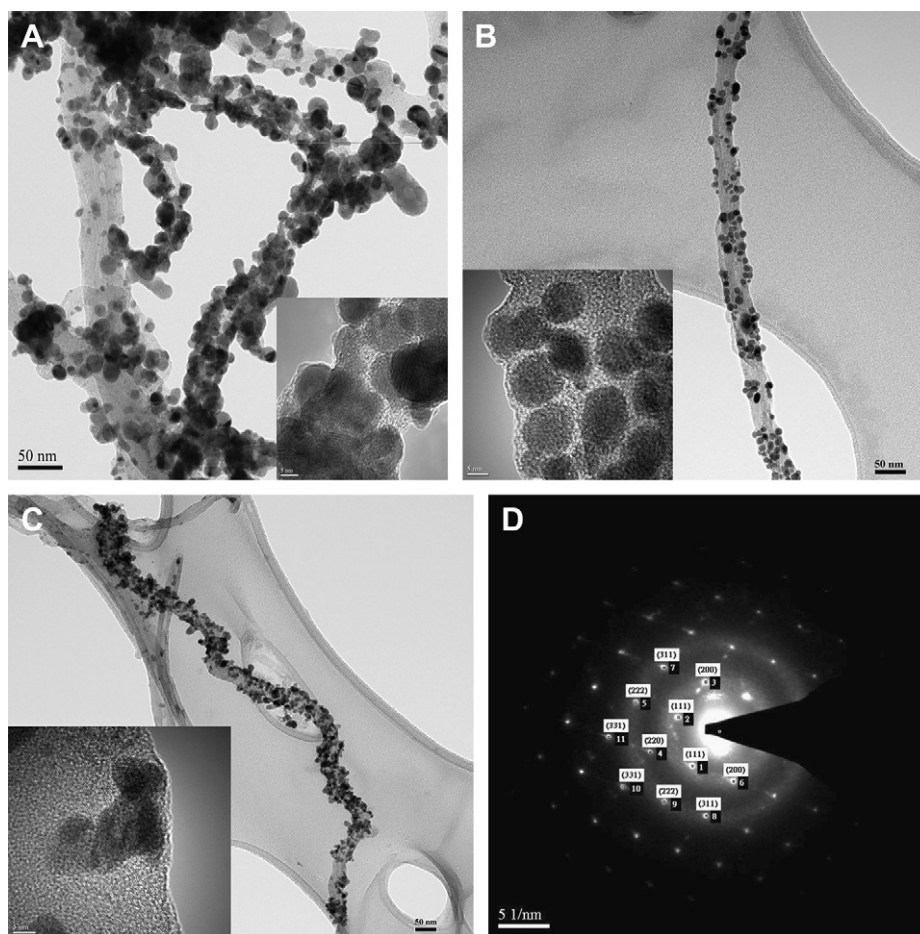


Fig. 1 – TEM images and diffraction pattern of prepared Ag–Pd nanoparticles-supported multiwall carbon nanotubes: (A) $\text{Ag}_1\text{Pd}_1/\text{CNT}$; (B) $\text{Ag}_2\text{Pd}_1/\text{CNT}$; (C) $\text{Ag}_4\text{Pd}_1/\text{CNT}$ (inset: high-resolution TEM images); (D) the diffraction pattern of $\text{Ag}_4\text{Pd}_1/\text{CNT}$.

of these alloy nanocatalysts in ORRs with an alkaline solution were studied. The activity and kinetics of the nanocatalysts were observed by performing electrochemical measurements.

2. Experimental

2.1. Preparation and analysis of materials

In the synthesis pathway, ~ 25 mg of raw catalyst-free multiwall carbon nanotubes was capped with SDS micelles and dispersed in 250 ml of 0.1 M SDS aqueous solution by sonicator agitation for ~ 4 h. Immediately after sonication, the SDS aqueous solution containing the dispersed CNTs was refluxed in an oil bath (~ 130 °C) for ~ 3 h. As discussed in our previous report [19], some of the SDS amphiphiles in the CNTs/SDS solution gradually released 1-dodecanol at the refluxing temperature. Subsequently, ~ 2.23 mmol silver nitrite and ~ 0.56 mmol palladium acetate were added to the CNTs/SDS solution and were slowly reduced by 1-dodecanol to zero-valence atoms in the micellar core; thus, Ag_4Pd_1 nanoparticles were formed. After 20 h, Ag_4Pd_1 nanoparticles supported on the carbon nanotubes ($\text{Ag}_4\text{Pd}_1/\text{CNTs}$) were readily observed as floatation on the refluxing aqueous

solution. When ~ 0.56 mmol and ~ 1.12 mmol silver nitrite were added to the reflux solution, Ag_1Pd_1 and Ag_2Pd_1 nanoparticles supported on carbon nanotubes ($\text{Ag}_1\text{Pd}_1/\text{CNTs}$ and $\text{Ag}_2\text{Pd}_1/\text{CNTs}$, respectively) were observed.

For performing TEM (JEOL, JEM-2000EX) measurements, the hydrophobic nanocomposites (in CHCl_3) were dropped onto a copper grid with a lacey support film and were dried naturally.

2.2. Electrochemical measurement

Before performing electrochemical measurement, the nanocomposites were well-dispersed and then extracted by adding 10 ml of CHCl_3 to the refluxing aqueous solution. CHCl_3 was removed from the Ag–Pd/CNTs solution at a reduced pressure of -10 °C. Then, the alloy catalyst powders were redispersed in 20 ml of ethanol and precipitated by high-speed centrifugation at 4000 rpm. Then, the black catalysts were washed with ethanol several times. 5 mg of the dry catalyst powders was added and further dispersed into 1 ml of the aqueous solution via ultrasonic vibration. 30 μl of the resulting aqueous solution was dropped onto a glassy carbon electrode (GCE) with a surface area of 0.07 cm^2 and was heated at 70 °C to evaporate H_2O . In order to prevent the catalyst from mixing in the electrolyte during the measurement, the GCE was rinsed

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