Employment of Fiber-Shaped Co Modified with Au Nanoparticles as Anode in Direct NaBH₄-H₂O₂ and N₂H₄-H₂O₂ Fuel Cells

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In this study, fiber-shaped cobalt (Coподоб) was electroplated on the copper surface and modified with various amounts of gold nanoparticles (denoted as AuCoобраз/Cu) with the aim to use it as the anode material in direct NaBH₄-H₂O₂ (DBHPFC) and N₂H₄-H₂O₂ (DHHPFC) fuel cells. The galvanic displacement technique has been used for the deposition of Au nanoparticles onto the Coобраз surface. The AuCoобраз/Cu catalysts were prepared with the Au loadings in the range of 10.9–84.4 μgAu cm⁻². Single fuel cell tests were performed by employing the prepared Coобраз/Cu and different AuCoобраз/Cu catalysts as the anode and a Pt sheet as the cathode. It was found that the peak power densities up to 162 mW cm⁻² for DHHPFC and 188 mW cm⁻² for DBHPFC were obtained at a temperature of 25°C using the AuCoобраз/Cu catalyst with the Au loading of 84.4 μgAu cm⁻² as the anode. The highest specific peak power density values of 12018 mW mg⁻¹Au for DHHPFC and 14954 mW mg⁻¹Au for DBHPFC were obtained, when employing the AuCoобраз/Cu catalyst with the lowest Au loading of 10.9 μgAu cm⁻² as the anode material.

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The interest of low-temperature fuel cells (FC), including direct borohydride fuel cells (DBFCs), as well as direct hydrazine fuel cells (DHFCs), have been growing over the past decades. The use of sodium borohydride (NaBH₄) and hydrazine (N₂H₄), those have hydrogen content of 10.6 and 12.5 wt%, respectively, as a fuel in FC is quite beneficial to alternative hydrogen or methanol fuel cells. Operation of DBFC is typically based on borohydride (BH₄⁻) oxidation involving eight electrons at the anode (Eq. 1) and oxygen reduction at the cathode (Eq. 2).

\[ \text{BH}_4^- + 8\text{OH}^- \rightarrow \text{BO}_2^- + 6\text{H}_2\text{O} + 8e^- \quad E^0 = -1.24 \text{ V vs. SHE} \quad [1] \]

\[ 2\text{O}_2 + 4\text{H}_2\text{O} + 8e^- \rightarrow 8\text{OH}^- \quad E^0 = 0.40 \text{ V vs. SHE} \quad [2] \]

The overall cell reaction is as follows:

\[ \text{BH}_4^- + 2\text{O}_2 \rightarrow \text{BO}_2^- + 2\text{H}_2\text{O} \quad E_0 = 1.64 \text{ V} \quad [3] \]

The DHFC is based on the oxidation of hydrazine in an alkaline media, occurring at the anode (Eq. 4) and oxygen reduction at the cathode (Eq. 2).

\[ \text{N}_2\text{H}_4 + 4\text{OH}^- \rightarrow \text{N}_2 + 4\text{H}_2\text{O} + 4e^- \quad E^0 = -1.16 \text{ V vs. SHE} \quad [4] \]

The total cell reaction can be written as Eq. 5:

\[ \text{N}_2\text{H}_4 + 2\text{O}_2 \rightarrow \text{N}_2 + 4\text{H}_2\text{O} \quad E_0 = 2.93 \text{ V} \quad [8] \]

The fact that hydrogen peroxide is a liquid oxidant at normal temperature and pressure, simplifies its transportation, storage, and handling, when compared to that of oxygen. It has the additional advantage of being good to use in a small volume single stack fuel cell. Although using H₂O₂ as oxidant enables to take advantage of the pH difference between anode/cathode and thus gain some “free” Nernst potential difference (leading to higher OCV compared to O₂ as oxidant), the pH gradient raises a lot of practical issue (junction potential, degradation of the membrane, etc.).

Since borohydride and hydrazine are used as fuel, the development of electrocatalysts with reasonable costs and a high electroactivity toward its oxidation is industrially vital. The electro-oxidation of borohydride10–15 and hydrazine16–20 on the surface of different catalysts has been properly studied recently. Herein, we use a fiber-shaped cobalt coating modified with small amounts of Au nanoparticles as the anode in direct borohydride-hydrogen peroxide and hydrazine-hydrogen peroxide fuel cells.

**Experimental**

**Chemicals.**—NaBH₄ (99%), H₂AuCl₄ (99.9%), N₂H₄ (50–60%), C₂H₄N₂O (N-(2-Hydroxyethyl) ethylenediamine, 99%) were purchased from Sigma-Aldrich Supply, CoSO₄ (Alfa Aesar, 98%), H₂O₂ (Eurochemicals, 35%), NaOH (98.8%), H₂SO₄ (96%), HCl (36%) were purchased from Chempute Company. All chemicals were of analytical grade. Deionized (Elix 3 Millipore) water was used to prepare all the solutions. A Nafion N117 membrane was purchased from DuPont (Wilmington, DE).

**Fabrication of catalysts.**—The catalysts were prepared by a two-step process, which involves electrodeposition of cobalt on the copper surface followed by a spontaneous gold displacement from the 1 mM H₂AuCl₄ + 0.1 M HCl solution (denoted as gold(III)-containing solution). Prior to the electroplating of Co coating, the surface of the Cu foil (1 × 1 cm) was pretreated with SiC emery paper (grade 2500) and MgO powder, etched in 10% H₂SO₄ and rinsed with deionized water. A rectangular glass cell that has a Cu foil cathode parallel to two stainless steel sheets anodes was used. A Co coating that has a fiber structure was electroplated on a Cu foil from an electrolyte containing 40 g 1⁻¹ CoSO₄, 100 g 1⁻¹ NaOH and 60 g 1⁻¹ N-(2-Hydroxymethyl) ethylenediamine using a galvanostatic control at a cathode current density of 40 mA cm⁻² for 20 min at a temperature of 20 ± 2°C.
Cofiber/Cu electrodes were immersed into the gold(III)-containing solution at a temperature of 25°C for 0.5, 1 and 5 min, respectively.

**Fuel cell test.**—The performance of direct NaBH4-H2O2 and N2H4-H2O2 fuel cells was evaluated using a Zennium electrochemical workstation (ZAHNER-Elektrik GmbH & Co. KG) at a temperature of 25°C. The schematic diagram of the assembled direct NaBH4-H2O2 or N2H4-H2O2 fuel cells was shown in Fig. 1.

The prepared Co/Cu and AuCo/Cu catalysts with a geometric area of 2 cm² were employed as the anode and a Pt sheet as the cathode. Each compartment of the cell contained 100 ml of the corresponding aqueous electrolyte. The anolyte was composed of an alkaline mixture of 1 M NaBH4 or 1 M N2H4 in 4 M NaOH. The catholyte was contained of 5 M H2O2 in 1.5 M HCl. The anolyte and catholyte solutions were prepared immediately before the measurements. A Nafion N117 membrane was used to separate the anodic and cathodic compartments of the single direct NaBH4-H2O2 or N2H4-H2O2 fuel cells. The load was applied in steps of 50 mV. Each step lasted 30 second (one point per second) and the cell voltage was continuously applied from one value to next without disconnecting the cell. Power density values were calculated from the applied cell voltage and steady-state current. The presented current and peak power densities were normalized with respect to the geometric area of catalysts.

**Results and Discussion**

In the present study, the performance of direct alkaline NaBH4-H2O2 and N2H4-H2O2 single fuel cells has been investigated by using the fiber-shaped Co modified with Au nanoparticles as the anode material. The simple and cost-effective electrochemical and chemical methods were used for fabrication of the catalysts. At first, the Co coating with a fiber structure and the thickness of approximately 3 μm was electrodeposited on the Cu surface. As seen from the data of SEM analysis in Fig. 2a, the Co coating, that has the fibers in the order of tens of nanometers in thickness and hundreds of nanometers in length, was electrodeposited on the Cu surface. Then, the prepared Co(Cu)/Cu electrodes were immersed into the gold(III)-containing solution for various time periods. This simple procedure is known as galvanic displacement, during which the deposition of a noble metal occurs by the oxidation of a precursor metal adlayer deposited on the substrate at the open-circuit potential.5,10 Immersion of Co(Cu)/Cu into the gold(III)-containing solution for 0.5, 1 and 5 min results in the formation of Au nanoparticles on the Co(Cu)/Cu surface in size of 10 up to 50 nm (Figs. 2b–2d). The Au and Co loadings in the prepared catalysts were determined using ICP-OES. The summarized data are given in Table I. The Au loadings in the prepared AuCo(Cu) catalysts were in the range of 10.9 up to 84.4 μgAu cm⁻² (Table I).

The stability of the Au nanoparticles modified fiber-shaped Co catalysts deposited on the Cu surface was examined in DBHPFC and DHHPFC single fuel cells. An alkaline direct NaBH4-H2O2 and N2H4-H2O2 fuel cells were constructed with the Co(Cu)/Cu and different AuCo(Cu) catalysts as the anode and Pt sheet as the cathode (Fig. 1). The solutions of 1 M NaBH4 or 1 M N2H4 in 4 M NaOH and 5 M H2O2 in 1.5 M HCl were used as the anolyte and catholyte, respectively. A Nafion N117 membrane was used as a separator. Fuel cell measurements were performed at a temperature of 25°C. It should be noted that during the discharge process, small bubbles of hydrogen and oxygen were observed on the Co(Cu)/Cu and AuCo(Cu) catalysts due to the homogeneous decomposition of the anode and cathode. The obtained fuel cell polarization curves and the corresponding power density curves against the current density by employing the prepared Co(Cu)/Cu and AuCo(Cu) catalysts as the anodes for DBHPFC and DHHPFC are presented in Fig. 3. The main obtained parameters are summarized in Table II.

The direct N2H4-H2O2 and NaBH4-H2O2 fuel cells exhibited an open circuit voltage of ca. 1.7 and 1.9 V, respectively. It was found that in both types of fuel cells the power density values are higher in the case of the investigated AuCo(Cu) catalysts as compared to those of Co(Cu)/Cu (Fig. 3). Highest peak power densities up to 162 mW cm⁻² for DHHPFC and 188 mW cm⁻² for DBHPFC were obtained at a temperature of 25°C using the AuCo(Cu)/Cu catalyst with the Au loading of 84.4 μgAu cm⁻² as the anode. Peak power densities up to 162 mW cm⁻² for DHHPFC were attained at a current density of 270 mA cm⁻² and a cell voltage of 0.60 V. The peak power densities up to 188 mW cm⁻² for DBHPFC were attained at a current density of 235 mA cm⁻² and a cell voltage of 0.80 V. Peak power density values for DHHPFC, recorded on the AuCo(Cu)/Cu anode catalyst with the Au loading of 84.4 μgAu cm⁻² are ca. 1.1, 1.2 and 1.5 times higher than those on the catalysts with the Au loadings of 22.4 and 10.9 μgAu cm⁻² and Co(Cu)/Cu catalyst, respectively. In the case of DHHPFC, peak power density values, recorded on the AuCo(Cu)/Cu catalyst with the Au loading of 84.4 μgAu cm⁻² are ca. 1.1, 1.2 and 1.3 times higher than those on the catalysts with the Au loadings of 22.4 and 10.9 μgAu cm⁻² and Co(Cu)/Cu catalyst, respectively (Fig. 3, Table II).

To compare the power density of the prepared AuCo(Cu) catalysts, the obtained power density values were normalized by the Au

![Figure 1. Schematic of the assembled direct NaBH4-H2O2 or N2H4-H2O2 fuel cells. Anode catalyst - Co(Cu)/Cu or AuCo(Cu)/Cu, cathode catalyst - Pt anolyte - 1 M NaBH4 or 1 M N2H4 in 4 M NaOH, catholyte - 5 M H2O2 in 1.5 M HCl.](image)

![Figure 2. SEM micrographs of the top side views of as-prepared Co(Cu)/Cu (a) and different AuCo(Cu)/Cu catalysts: (b) 10.9, (c) 24.4 and (d) 84.4 μgAu cm⁻².](image)

| Catalysts | Au deposition time, min | Au loading, μg cm⁻² | Co loading, μg cm⁻² |
|-----------|------------------------|---------------------|---------------------|
| Co(Cu)/Cu | 0.5                    | 10.9                | 2353.0              |
| AuCo(Cu)/Cu | 1                   | 22.4                | 1615.5              |
|            | 5                     | 84.4                | 1002.5              |
loadings for each catalyst. The summarized data are given in Fig. 4 and Table II. As seen, the highest specific power density (mW mg⁻¹Au) was obtained using the catalysts with the lowest used Au loading, e.g. 10.9 μgAu cm⁻² in both types of FC (Table II). Highest specific peak power density values of 12018 mW mg⁻¹Au for N₂H₄-H₂O₂ and 14954 mW mg⁻¹Au for NaBH₄-H₂O₂ were obtained, when employing the AuCo₃/Cu catalyst with the Au loading of 10.9 μgAu cm⁻² as the anode material. The obtained specific peak power density values for DHHPFC using the AuCo₃/Cu catalyst with the Au loading of 10.9 μgAu cm⁻² are ca. 1.9 and 6.3 times higher as compared to that at the AuCo₃/Cu catalysts with the Au loadings of 22.4 and 84.4 μgAu cm⁻², respectively. In the case of DBHPFC, the specific peak power density is ca. 2.0 and 6.7 times higher at the AuCo₃/Cu catalyst, which has the lowest Au loading, as compared to that at the AuCo₃/Cu catalysts, prepared with higher Au loadings.

To evaluate the performance of DHHPFC and DBHPFC by employing the Co₃/Cu and AuCo₃/Cu as the anodes, the peak power density values were normalized by the Co and total catalyst (Au + Co).

| Type of fuel Cell | Catalysts | Au loading (μg cm⁻²) | E_peak (V) | j_peak (mA cm⁻²) | P (mW cm⁻²) | Specific P (mW mg⁻¹Au) | Specific P (mW mg⁻¹cat) |
|------------------|-----------|---------------------|-----------|------------------|-------------|------------------------|-------------------------|
| DHHPFC           | AuCo₃/Cu  | -                   | 0.60      | 183              | 110         | -                      | 46.7                    |
|                  | AuCo₃/Cu  | 10.9                | 0.60      | 218              | 131         | 12018                  | 80.5                    |
|                  | AuCo₃/Cu  | 22.4                | 0.55      | 259              | 142         | 6339                   | 90.3                    |
|                  | AuCo₃/Cu  | 84.4                | 0.60      | 270              | 162         | 1919                   | 149.0                   |
|                  | AuCo₃/Cu  | 10.9                | 0.85      | 191              | 142         | 14954                  | 100.2                   |
|                  | AuCo₃/Cu  | 22.4                | 0.80      | 211              | 169         | 7545                   | 107.5                   |
|                  | AuCo₃/Cu  | 84.4                | 0.80      | 235              | 188         | 2227                   | 173.0                   |

Table II. Electrochemical parameters of the DHHPFC and DBHPFC employing the Co₃/Cu and AuCo₃/Cu catalysts as the anode.

![Figure 3](image1.png)  
**Figure 3.** Cell polarization and power density curves for the DBHPFC (a) and DHHPFC (b) using the Co₃/Cu and different AuCo₃/Cu anode catalysts with the anolyte consisted of 1 M NaBH₄ (a) or 1 M N₂H₄ (b) in 4 M NaOH and 5 M H₂O₂ + 1.5 M HCl catholyte at a temperature of 25°C.

![Figure 4](image2.png)  
**Figure 4.** Specific power density curves for the DBHPFC (a) and DHHPFC (b) for each AuCo₃/Cu anode catalyst. Anolyte consisted of 1 M NaBH₄ (a) or 1 M N₂H₄ (b) in 4 M NaOH and catholyte consisted of 5 M H₂O₂ in 1.5 M HCl at a temperature of 25°C.
mass, respectively. The summarized data are given in Table II. It should be noted that the Co@Au/C catalyst seems to be an attractive material on its own, whereas the decoration of it by Au nanoparticles significantly increases performance of the both DHHPFC and DBHPFC fuel cells (Table II). The AuCo@Cu/C anode catalysts with the Au loadings in the range of 10.9–84.4 μgAu cm⁻² exhibited ca. 1.7–3.2 times higher specific power density value (mW mg⁻¹ Au) as compared to those for the employed Co@Cu/C anode in the DHHPFC tests. In the case of DHHPFC, ca. 1.7–2.9 times higher specific power density values were obtained using the investigated AuCo@Cu/C anode catalysts as compared to that for pure Co@Cu/C anode.

Data given in Table III presents the comparison of the power performance data of DHHPFC and DBHPFC in recent years.22,27–33 As we can see, the obtained peak power density values for DHHPFC that operated using the Co@Cu/C and AuCo@Cu/C (84.4 μgAu cm⁻²) catalysts as the anodes and at a temperature of 25 °C, are significantly greater compared to those obtained on the previously reported Au-based catalysts.22,27,28 The DHHPFC with the AuCo@Cu/C (10.9 μgAu cm⁻²) anode catalyst shows the highest specific power density (mW mg⁻¹ Au) as compared to that for Au-based catalysts.29–33 Therefore, the highest specific power density based on total mass of catalyst shows the AuCo@Cu/C anode catalyst with the Au loading of 84.4 μgAu cm⁻² as compared to that on the previously reported Au-based catalysts.22,23

Therefore, a fiber-shaped Co coating and that modified with low amounts of Au nanoparticles would be promising anode catalysts for the application of direct N₂H₄-H₂O₂ and NaBH₄-H₂O₂ fuel cells.

### Conclusions

The simple and cost-effective electrochemical method was used for the deposition of the fiber-shaped Co coating onto the Cu surface. Au nanoparticles were then deposited onto the fiber-shaped Co coating by the galvanic displacement technique. It was found that the Au loadings in the prepared AuCo@Cu/C catalysts were in the range of 10.9–84.4 μg Au cm⁻². Pure Co@Cu/C and AuCo@Cu/C catalysts were tested as the anode material in the direct N₂H₄-H₂O₂ and NaBH₄-H₂O₂ fuel cells. The direct N₂H₄-H₂O₂ and NaBH₄-H₂O₂ fuel cells exhibited an open circuit voltages of ca. 1.7 and 1.9 V respectively. The deposition of low amount of Au nanoparticles on the fiber-shaped Co coating resulted in an enhanced peak power densities for the direct N₂H₄-H₂O₂ and NaBH₄-H₂O₂ fuel cells as compared to those of pure Co@Cu/C. Peak power densities up to 162 mW cm⁻² for N₂H₄-H₂O₂ and 188 mW cm⁻² for NaBH₄-H₂O₂ were obtained at a temperature of 25 °C using the AuCo@Cu/C anode catalyst with the Au loading of 84.4 μg Au cm⁻². The highest specific peak power density values of 12018 mW mg⁻¹ Au for N₂H₄-H₂O₂ and 14954 mW mg⁻¹ Au for NaBH₄-H₂O₂ were obtained, when employing the AuCo@Cu/C anode catalyst prepared with the lowest Au loading of 10.9 μg Au cm⁻².
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