

Employment of Au/Co and Au/CoB as materials for anodes in borohydride–hydrogen peroxide fuel cells

A. Balčiūnaitė^{1*},

Z. Sukackienė¹,

L. Tamašauskaitė-Tamašiūnaitė¹,

D. M. F. Santos²,

E. Norkus¹

¹Department of Catalysis,
Center for Physical Sciences and Technology,
Saulėtekio Ave. 3,
LT-10257 Vilnius, Lithuania

²CEFEMA, Instituto Superior Técnico,
Universidade de Lisboa,
1049-001 Lisbon, Portugal

Au nanoparticles modified Co and CoB coatings deposited on the Cu surface are studied as anode materials for direct NaBH₄–H₂O₂ fuel cells. The catalysts are prepared via electroless deposition of Co or CoB layers on the Cu surface followed by galvanic displacement of hierarchical Co by Au. The performance of direct borohydride–hydrogen peroxide fuel cells (DBHPFCs) using Au/Co/Cu or Au/CoB/Cu as the anode and Pt as the cathode is studied at different temperatures. A 1 M NaBH₄ + 4 M NaOH solution is used as the anolyte and 5 M H₂O₂ + 1.5 M HCl is used as the catholyte, with a Nafion N117 membrane separating the two compartments. The DBHPFC using Au/Co/Cu and Au/CoB/Cu as the anode catalyst exhibits high electrochemical performance compared to that using Co/Cu and CoB/Cu as the anode catalyst. The highest specific peak power densities of 33.9 kW g_{Au}⁻¹ at 25 °C, at the current density of 150 mA cm⁻² and cell voltage of 0.70 V and 58.1 kW g_{Au}⁻¹ at 55 °C, at 240 mA cm⁻² and 0.75 V were attained using the Au/CoB/Cu anode with the Au loading of 3.1 μg_{Au} cm⁻².

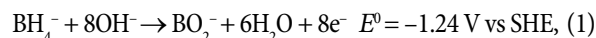
Keywords: gold, cobalt, cobalt boride, anodes, direct borohydride–peroxide fuel cell

INTRODUCTION

In recent years, resources of fossil fuels (such as coal, oil, natural gas) are decreasing, and only a small part of the electricity is produced using renewable energy sources. New and environmentally friendly energy sources exploration and development is very important in today's society. Fuel cells are alternative energy sources that directly convert chemical energy into electrical energy [1–3]. They are actively investigated as cleaner power generators than fossil fuel-based technologies. These studies point out fuel cells as promising candidates for minimizing the usage of fossil fuels.

One type of fuel cell is the direct borohydride fuel cell (DBFC), which uses borohydride (NaBH₄ or KBH₄) aqueous solution as the fuel, and is considered to be a potential energy installation [4–8]. These fuel cells have been drawing increasing attention because of their high energy density, the low toxicity of borohydride, their good power performance, and the feasibility of using non-noble metals as both the anode and cathode catalysts [9–13]. The main

reactions occurring in the alkaline direct borohydride are the oxidation of BH₄⁻ ions at the anode (Eq. 1) and the reduction of oxygen at the cathode (Eq. 2), with the overall DBFC reaction being given by Eq. 3:

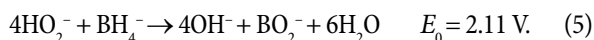


The direct oxidation of the BH₄⁻ ion in an alkaline medium involves the transfer of eight electrons at the equilibrium potential of -1.24 V vs SHE, which, in combination with reduction of oxygen, gives a theoretical equilibrium cell potential of 1.64 V (Eqs. 1–3). The product of this reaction is metaborate (BO₂⁻), an inert and non-toxic product that can be recycled back to BH₄⁻.

The direct liquid feed borohydride–hydrogen peroxide fuel cell (DBHPFC) is based on the BH₄⁻ oxidation at the anode and on the hydrogen peroxide (H₂O₂) reduction at the cathode. The use of H₂O₂ instead of O₂ as an oxidant can yield higher cell potential values [14–17]. These fuel

* Corresponding author. E-mail: balciunaite.aldona@gmail.com

cells can be used as prototype batteries for undersea water vehicles [16, 17]. The direct H_2O_2 reduction in alkaline solution at the cathode of the DBHPFC is given by Eq. 4 [16], and therefore the overall cell reaction can be written as Eq. 5 and is calculated by subtraction of E_K (Eq. 4) – E_A (Eq. 1):



As shown, E^0 of 0.87 V vs SHE for the direct reduction of H_2O_2 (Eq. 4) is higher than that for the O_2 reduction (0.40 V vs SHE, Eq. 2). The theoretical cell voltage and specific energy of the DBHPFC is larger compared to that of the DBFC. The cell voltage of the DBHPFC can range from 2.11 to 3.01 V (depending on the pH) and the specific energy is 17 Wh g^{-1} , whereas the cell voltage and specific energy of DBFC are lower, being 1.64 V and 9.25 Wh g^{-1} , respectively. It is considered that it depends on a lower activation barrier and faster kinetics of the two electron H_2O_2 reduction compared to the four electron O_2 reduction [18]. The use of H_2O_2 as a liquid oxidant in the fuel cell makes its storage, transport, handling and controllable feeding to a fuel cell easier than in the case of O_2 gas.

It is well known that Au-alloy catalysts have high electrocatalytic activity for the direct oxidation of BH_4^- and that the noble metal catalysts make the DBFC relatively expensive. Developing Au-alloys with non-noble metals to decrease the amount of the noble metal in the anode electrocatalyst is an effective way to reduce the cost of DBFCs. Many researchers have been therefore interested in exploring lower-cost substitutes. For example, some transition metals (Co, Ni, Cu) [19–21] have been successfully applied as an alternative to gold in the anode of DBFCs. More recently, much effort has been engaged in developing binary or alloy catalysts [22–24]. It has been determined that bimetallic catalysts usually show a higher activity and stability than the monometallic ones [19, 25, 26].

However, to the author's knowledge, the employment of Co alloys as anode catalysts for the DBHPFC has not been sufficiently discussed. In our previous work [19, 26], it has been shown that the gold-cobalt-copper and gold-cobalt-boron-copper (denoted as Au/Co/Cu and Au/CoB/Cu) catalysts prepared via simple chemical methods exhibit high electrocatalytic activity towards the oxidation of borohydride [19, 26]. Therefore, the aim of this study was to investigate the possibility to use the Au/Co/Cu and Au/CoB/Cu catalysts as anode material in the direct liquid feed borohydride–hydrogen peroxide fuel cell at different temperatures.

EXPERIMENTAL

Chemicals

HAuCl_4 (Sigma-Aldrich, 99.9%), $\text{KAu}(\text{CN})_2$ (Sigma-Aldrich, 98%), $\text{CoSO}_4 \cdot 7\text{H}_2\text{O}$ (Chempur, 99.5%), $\text{C}_4\text{H}_8\text{ONH} \cdot \text{BH}_3$ (borane morpholine complex, Sigma-Aldrich, 97%),

$\text{NH}_2\text{CH}_2\text{COOH}$ glycine, Chempur, 99%), $(\text{NH}_4)_2\text{C}_6\text{H}_6\text{O}_7$ (ammonium citrate, Chempur, 99%), NaBH_4 (Sigma-Aldrich, 96%), NaOH (AnalaR NORMAPUR, 99%), H_2O_2 (Merck, 35%), HCl (Carlo Erba, 37%), and H_2SO_4 (Chempur, 96%) were used as received and all solutions were prepared with deionized (Elix 3 Millipore) water. All chemicals were of analytical grade. A Nafion N117 membrane was purchased from DuPont (Wilmington, DE).

Fabrication of catalysts

The Co/Cu, CoB/Cu, Au/Co/Cu and Au/CoB/Cu catalysts were prepared in the same manner as described in detail by Sukackienė et al. [19]. Briefly, the Co or CoB layers were electrolessly deposited on the Cu surface with a geometric area of 2 cm^2 using borane morpholine complex as a reducing agent. The duration of electroless deposition of Co and CoB coatings on the Cu surface was 30 and 100 min, respectively. The Co plating bath consisted of 0.05 M CoSO_4 , 0.05 M $\text{C}_4\text{H}_8\text{ONH} \cdot \text{BH}_3$ and 0.2 M $\text{NH}_2\text{CH}_2\text{COOH}$. To obtain CoB coatings, a Co plating bath with the same composition, but without glycine was used. The bath operated at pH 7 and at a temperature of 30 °C. The thickness of the pure Co and CoB coatings obtained was ca. 1 μm . Au nanoparticles were deposited on the prepared Co/Cu and CoB/Cu electrodes by immersion of Co/Cu and CoB/Cu electrodes into the 1 mM HAuCl_4 solution (pH = 1.8) (denoted as an acidic Au-containing solution) and 3.5 mM $\text{KAu}(\text{CN})_2$ + 0.4 M $(\text{NH}_4)_2\text{C}_6\text{H}_6\text{O}_7$ complex (pH = 5) (denoted as a weak acidic Au-containing solution) at a temperature of 30 °C for 0.5, 1 and 5 min. The prepared catalysts were used for direct borohydride–hydrogen peroxide fuel cell test measurements without any further treatment.

Fuel cell test experiments

Single cell tests were carried out in a lab-scale DBHPFC by employing the prepared Co/Cu, CoB/Cu, Au/Co/Cu or Au/CoB/Cu electrodes (with a geometric area of 2 cm^2) as the anode and a Pt sheet as the cathode. Each compartment of the cell contained 100 mL of static electrolyte solution, with the anolyte containing 1 M NaBH_4 + 4 M NaOH and the catholyte containing 5 M H_2O_2 + 1.5 M HCl . A Nafion N117 membrane was used to separate the anodic and cathodic compartments of the single direct NaBH_4 – H_2O_2 fuel cell. The membrane active area was ca. 30 cm^2 . Cell measurements were conducted using a Zennium electrochemical workstation (ZAHNER-Elektrik GmbH & Co. KG). The performance of the fuel cell was evaluated by recording the cell polarization curves and obtaining the corresponding power density curves at temperatures in the range of 25–55 °C.

RESULTS AND DISCUSSION

In the present study, the Au/Co/Cu or Au/CoB/Cu catalysts were examined as anode materials in a direct alkaline NaBH_4 – H_2O_2 single fuel cell. The catalysts were prepared by means of the electroless deposition of Co or CoB coatings

followed by the spontaneous deposition of Au nanoparticles via galvanic displacement from an acidic or a weak acidic Au-containing solutions. The Au nanoparticles are homogeneously dispersed on the Co/Cu and CoB/Cu surfaces, as was shown in [19] and [26]. It was found that the Au loadings in the prepared Au/Co/Cu or Au/CoB/Cu catalysts were in the 3–44 $\mu\text{g cm}^{-2}$ range (Table 1). The DBHPFC was con-

structed using the prepared Co/Cu, CoB/Cu, Au/Co/Cu or Au/CoB/Cu catalysts as the anode and the Pt sheet as the cathode. Fuel cell measurements were carried out at temperatures of 25, 35, 45 and 55 °C. During the cell discharge process, small bubbles of hydrogen and oxygen were observed at the electrodes surface due to the chemical decomposition of BH_4^- and H_2O_2 at the anode and at the cathode, respectively. Figure 1

Table 1. DBHPFC performance with Au/Co/Cu and Au/CoB/Cu anodes

No.	Catalyst	Au loading, $\mu\text{g cm}^{-2}$	T, °C	E at peak power density, V	j at peak power density, mA cm^{-2}	Peak power density, mW cm^{-2}	Specific peak power density, $\text{W g}_{\text{Au}}^{-1}$
1.	Au/Co/Cu*	9.3	25	0.850	205.5	174.9	18800
			55	0.750	259.3	194.6	20900
2.	Au/Co/Cu*	11.6	25	0.700	264.3	185.2	16000
			55	0.850	261.2	222.2	19200
3.	Au/Co/Cu*	36.1	25	0.750	265.9	199.6	5500
			55	0.750	380.2	285.4	7900
4.	Au/CoB/Cu*	8.0	25	0.850	201.8	171.7	21461
			55	0.750	268.7	201.6	25206
5.	Au/CoB/Cu*	14.3	25	0.750	240.6	180.5	12625
			55	0.800	271.2	217.1	15179
6.	Au/CoB/Cu*	44.2	25	0.750	249.3	187.1	4233
			55	0.750	311.8	234.0	5294
7.	Au/Co/Cu**	5.8	25	0.750	203.1	152.4	26300
			55	0.800	273.1	218.6	37700
8.	Au/Co/Cu**	7.1	25	0.750	270.0	202.6	28500
			55	0.800	299.3	239.6	33700
9.	Au/Co/Cu**	15.3	25	0.850	259.6	220.8	14400
			55	0.850	313.0	266.3	17400
10.	Au/CoB/Cu**	3.1	25	0.700	149.9	105.0	33900
			55	0.750	239.9	180.0	58100
11.	Au/CoB/Cu**	5.2	25	0.850	171.8	146.1	28100
			55	0.750	258.0	193.6	37200
12.	Au/CoB/Cu**	14.0	25	0.850	240.5	204.5	14600
			55	0.800	322.4	258.0	18400

* Au/Co/Cu and Au/CoB/Cu were prepared by immersion of Co/Cu and CoB/Cu in an acidic (pH = 1.8) Au-containing solution for 0.5 (1, 4), 1 (2, 5) and 5 (3, 6) min

** Au/Co/Cu and Au/CoB/Cu were prepared by immersion of Co/Cu and CoB/Cu in a weak acidic (pH = 5) Au-containing solution for 0.5 (7, 10), 1 (8, 11) and 5 (9, 12) min

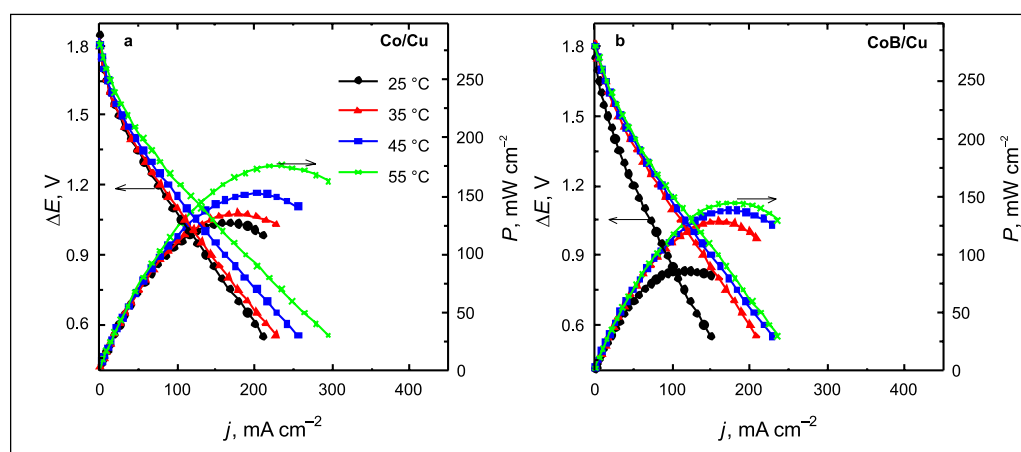


Fig. 1. Cell polarization and corresponding power density curves of DBHPFCs using Co/Cu (a) and CoB/Cu (b) anode catalysts fuelled by anolyte containing 1 M NaBH_4 + 4 M NaOH and 5 M H_2O_2 + 1.5 M HCl catholyte, operated at different temperatures (25–55 °C)

presents the fuel cell polarization curves and the corresponding power densities against the current density by employing the Co/Cu (a) and CoB/Cu (b). As seen, the CoB/Cu and Co/Cu generate a maximum power density of ca. 86–145 and 127–176 mW cm^{-2} , respectively, at 25–55 °C temperatures. Notably, peak power density values increase with the increase in the DBHPFC temperature and are greater when using pure Co as anode material as compared with those of CoB.

The polarization curves and the corresponding power densities of DBHPFC with the Au/Co/Cu and Au/CoB/Cu catalysts

prepared by immersion of Co/Cu or CoB/Cu in an acidic (a–c) and in a weak acidic (d–f) Au-containing solutions for 0.5 (a, d), 1 (b, e) and 5 (c, f) min are presented in Figs. 2 and 3. The summarized data are given in Table 1. As seen from the data, peak power density increases with increase in the temperature of DBHPFC. The maximum power densities of DBHPFC obtained at 25 °C ranged from ca. 105 to 221 mW cm^{-2} and from 180 to 285 mW cm^{-2} at 55 °C, using the investigated Au/Co/Cu and Au/CoB/Cu catalysts as anodes. It should be noted that the performance of the DBHPFC with the Au/Co/Cu and

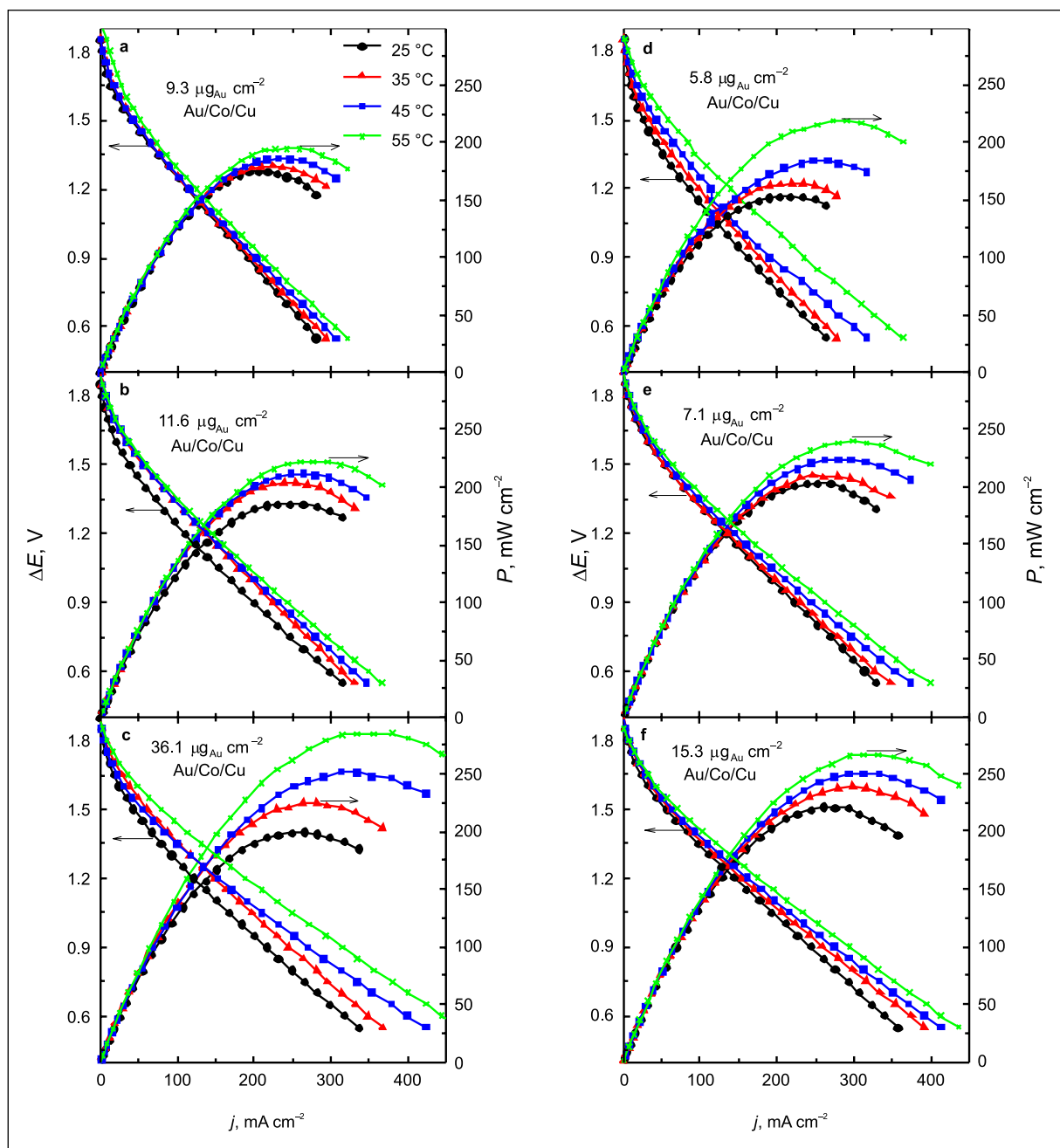


Fig. 2. Cell polarization curves and corresponding power density plots for the DBHPFC using Au/Co/Cu anode catalysts with anolyte containing 1 M NaBH_4 + 4 M NaOH and 5 M H_2O_2 + 1.5 M HCl catholyte. Au/Co/Cu were prepared by immersion of Co/Cu into 1 mM HAuCl_4 (pH 1.8) (a–c) and 3.5 mM KAu(CN)_2 + 0.4 M $(\text{NH}_4)_2\text{C}_6\text{H}_6\text{O}_7$ complex (pH 5) (d–f) at 30 °C for 0.5 (a, d), 1 (b, e) and 5 (c, f) min

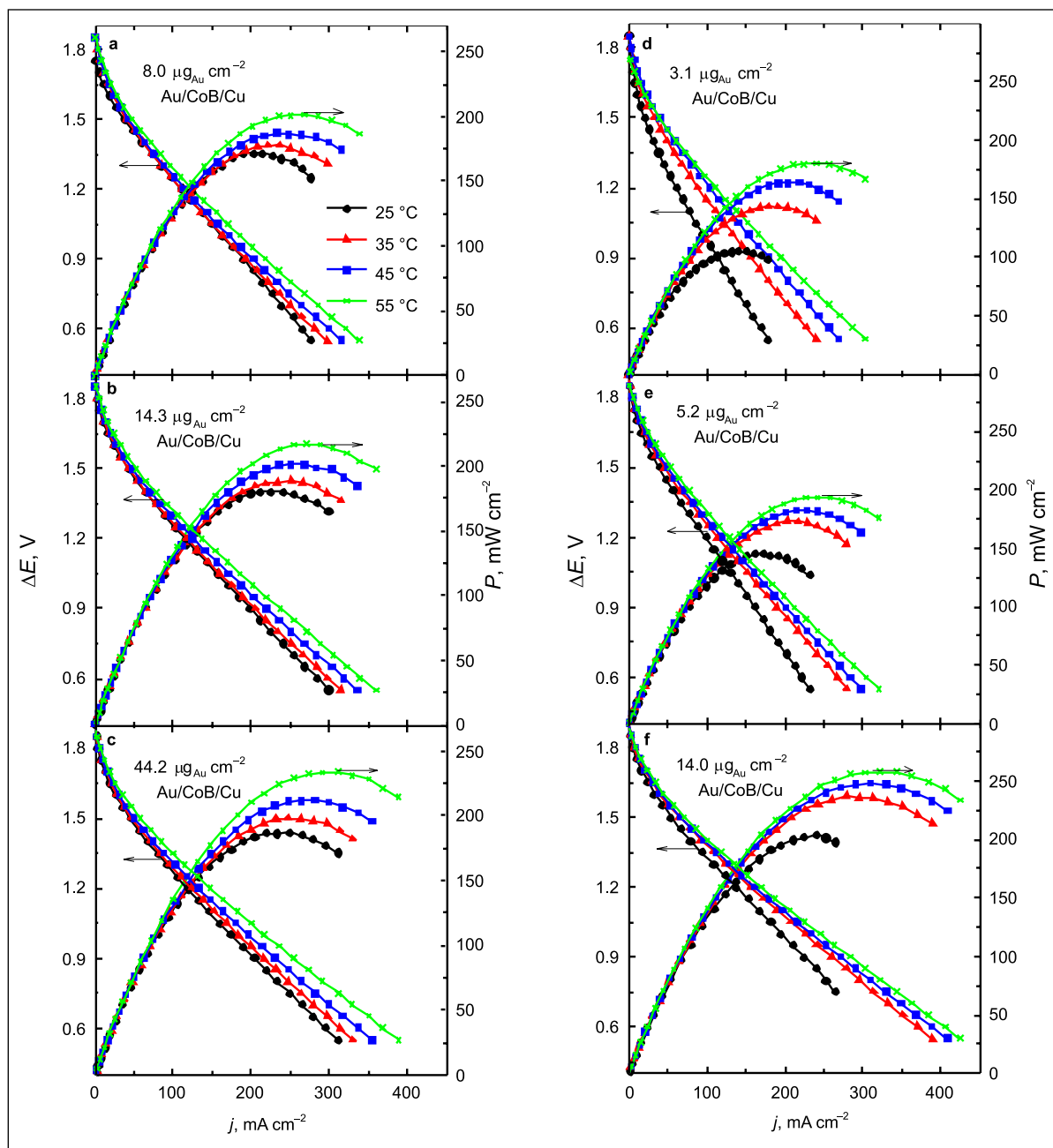


Fig. 3. Cell polarization curves and corresponding power density plots for the DBHPFC using Au/CoB/Cu anode catalysts with anolyte containing 1 M NaBH_4 + 4 M NaOH and 5 M H_2O_2 + 1.5 M HCl catholyte. Au/CoB/Cu were prepared by immersion of CoB/Cu into 1 mM HAuCl_4 (pH 1.8) (a–c) and 3.5 mM $\text{KAu}(\text{CN})_2$ + 0.4 M $(\text{NH}_4)_2\text{C}_6\text{H}_6\text{O}_7$ complex (pH 5) (d–f) at 30 °C for 0.5 (a, d), 1 (b, e) and 5 (c, f) min

Au/CoB/Cu anodes with low Au loadings is higher than that of the DBHPFC with Co/Cu and CoB/Cu anodes.

To evaluate the activity of investigated Au/Co/Cu and Au/CoB/Cu anodes, the power density values of DBHPFC were normalized in reference to the Au loadings for each catalyst. Figure 4 presents the specific power densities of DBHPFC constructed with the Au/Co/Cu (a, b) and Au/CoB/Cu (c, d) anodes prepared by immersion of Co/Cu or CoB/Cu in an acidic (a, c) and in a weak acidic (b, d) Au-containing solutions for 0.5, 1 and 5 min. It should be noted that higher specific power densities of DBHPFC were attained with Au/Co/Cu and

Au/CoB/Cu anodes prepared by immersion of Co/Cu and CoB/Cu electrodes into a weak acidic Au-containing solution for various time periods (Fig. 4b, d).

The maximum power densities of the DBHPFC, of $33.9 \text{ kW g}_{\text{Au}}^{-1}$ at 25 °C and of $58.1 \text{ kW g}_{\text{Au}}^{-1}$ at 55 °C, were obtained on the Au/CoB/Cu anode with the Au loading of $3.1 \mu\text{g}_{\text{Au}} \text{ cm}^{-2}$, at a current density of 240 mA cm^{-2} and cell voltage of 0.75 V (Fig. 4d).

Table 2 compares the DBHPFC performance data obtained using Co/Cu, CoB/Cu, Au/Co/Cu and Au/CoB/Cu anode catalysts with other previously reported DBHPFCs. It is

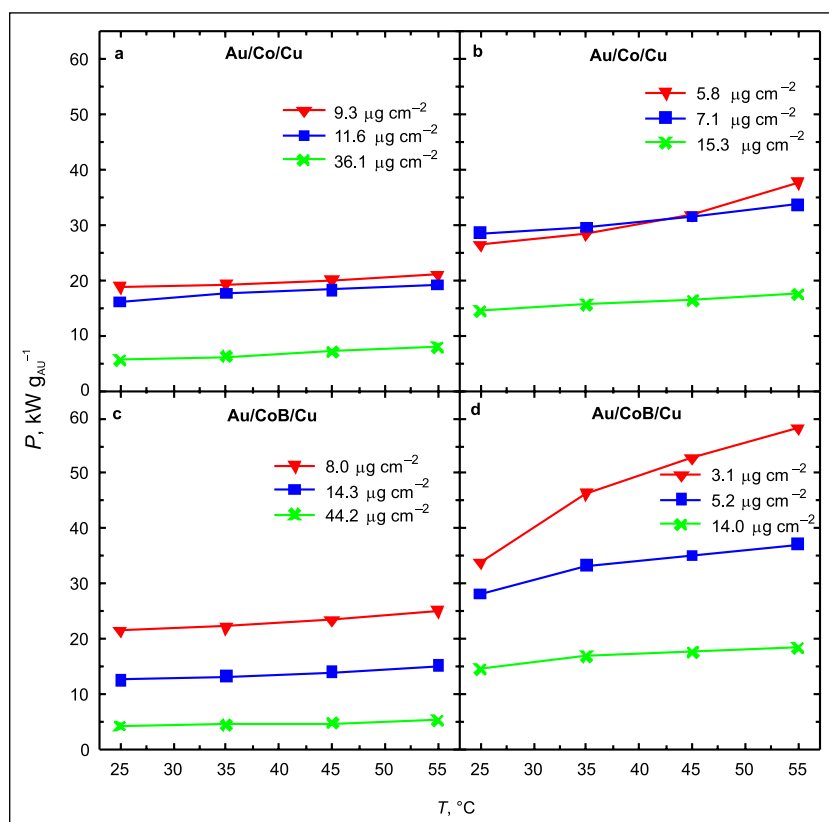


Fig. 4. Maximum specific power density values for DBHPFC using Au/Co/Cu (a, b) and Au/CoB/Cu (c, d) anode catalysts having different Au loadings with anolyte containing 1 M NaBH₄ + 4 M NaOH and 5 M H₂O₂ + 1.5 M HCl catholyte at different temperatures. Au/Co/Cu and Au/CoB/Cu were prepared by immersion of Co/Cu and CoB/Cu in an acidic (a, c) and a weak acidic (b, d) Au-containing solutions at 30 °C for 0.5 (▼▼, red online), 1 (■, blue) and 5 (××, green) min

Table 2. The comparison of the DBHPFC performance with different anodes

Anode (catalyst loading)	T, °C	Anolyte	P, mW cm ⁻²	P, W g ⁻¹ _{Au}	Ref.
Co/Cu	25	1 M NaBH ₄ + 4 M NaOH	126.6	–	This work
CoB/Cu	25	1 M NaBH ₄ + 4 M NaOH	86.0	–	This work
Au/Co/Cu* (0.0093 mg _{Au} cm ⁻²)	25	1 M NaBH ₄ + 4 M NaOH	174.9	18800	This work
Au/Co/Cu** (0.0058 mg _{Au} cm ⁻²)	25	1 M NaBH ₄ + 4 M NaOH	152.4	26300	This work
Au/CoB/Cu* (0.0080 mg _{Au} cm ⁻²)	25	1 M NaBH ₄ + 4 M NaOH	171.7	21461	This work
Au/CoB/Cu** (0.0031 mg _{Au} cm ⁻²)	25	1 M NaBH ₄ + 4 M NaOH	105.0	33900	This work
Au@CoB (0.7 mg cm ⁻²)	25	0.8 M KBH ₄ + 6 M KOH	85.0	121	[23]
Au ₄₅ Co ₅₅ /C (4.5 mg cm ⁻²)	25	1 M NaBH ₄ + 3 M NaOH	66.5	73.9	[11]
Au/C (4.0 mg cm ⁻² , 22.8 wt.% Au)	20	1 M NaBH ₄ + 3 M NaOH	25.8	28.3	[27]
Au/C (0.5 mg _{Au} cm ⁻²)	25	2 M NaBH ₄ + 6 M NaOH	~26.0	52.0	[28]
Au/C (0.5 mg _{Au} cm ⁻²)	20	25% NaBH ₄ + 6 M NaOH	34.4	68.8	[29]
AuPd (5.0 mg cm ⁻² , 20 wt.% metal)	60	2 M NaBH ₄ + 2 M NaOH	33.0	6.60	[12]

* Au/Co/Cu and Au/CoB/Cu were prepared by immersion of Co/Cu and CoB/Cu in an acidic Au-containing solution (pH 1.8) for 0.5 min.

** Au/Co/Cu and Au/CoB/Cu were prepared by immersion of Co/Cu and CoB/Cu in a weak acidic Au-containing solution (pH 5) for 0.5 min.

seen that significantly higher power densities were obtained in the DBHPFC with Au/Co/Cu and Au/CoB/Cu anodes as compared to those obtained on previously reported Au-met-

al [11, 12, 23] or Au/C [27–29] anode catalysts (Table 2). Notably, these results were obtained by the Au/Co/Cu and Au/CoB/Cu anodes with lower Au loadings.

CONCLUSIONS

Co and CoB coatings, deposited on the Cu surface and modified with low amounts of Au in the range of 3–44 $\mu\text{g}_{\text{Au}} \text{cm}^{-2}$ have been studied as anode materials in direct $\text{NaBH}_4\text{-H}_2\text{O}_2$ fuel cells. The Au/Co/Cu and Au/CoB/Cu electrocatalysts were prepared via simple chemical methods. The direct borohydride hydrogen peroxide fuel cell using Au/Co/Cu and Au/CoB/Cu as the anode catalyst exhibits high electrochemical performance compared with that of Co/Cu and CoB/Cu as the anode catalyst. The highest specific peak power densities of 33.9 $\text{kW g}_{\text{Au}}^{-1}$ at 25 °C, at the current density of 150 mA cm^{-2} and the cell voltage of 0.70 V, and 58.1 $\text{kW g}_{\text{Au}}^{-1}$ at 55 °C, at 240 mA cm^{-2} and 0.75 V were attained using the Au/CoB/Cu anode with the Au loading of 3.1 $\mu\text{g}_{\text{Au}} \text{cm}^{-2}$.

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Au/Co IR Au/CoB PANAUDOJIMAS ANODO MEDŽIAGOMIS NATRIO BOROHIDRIDO-VANDENILIO PEROKSIDO KURO ELEMENTUOSE

Santrauka

Ant Cu paviršiaus nusodinti Au/Co ir Au/CoB katalizatoriai tirti kaip anodo medžiagos tiesioginiame $\text{NaBH}_4\text{-H}_2\text{O}_2$ kuro elemente. Naudojant Au/Co/Cu ir Au/CoB/Cu anodo katalizatoriais $\text{NaBH}_4\text{-H}_2\text{O}_2$ kuro elemente, kai nusodinto Au įkrova yra 3–44 $\mu\text{g}_{\text{Au}} \text{cm}^{-2}$, gautos pastebimai didesnės galios tankio vertės, lyginant jas su Co/Cu ir CoB/Cu anodo katalizatoriais. Didžiausi specifiniai galios tankiai – 33,9 $\text{kW g}_{\text{Au}}^{-1}$, esant 25 °C temperatūrai, 150 mA cm^{-2} srovės tankiui ir 0,70 V celės įtampai, ir 58,1 $\text{kW g}_{\text{Au}}^{-1}$, esant 55 °C temperatūrai, 240 mA cm^{-2} srovės tankiui ir 0,75 V celės įtampai, buvo gauti panaudojant Au/CoB/Cu katalizatorių su nusodinto Au įkrova 3,1 $\mu\text{g}_{\text{Au}} \text{cm}^{-2}$.