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Application of platinum–cobalt–boron as the anode material for sodium borohydride–hydrogen peroxide fuel cells

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The electroless deposition and galvanic displacement methods were used for the fabrication of cobalt–boron (CoB) catalysts modified with small amounts of platinum crystallites in the range of 9.8 to 14.4 μg_{Pt} cm^{-2}. The prepared catalysts were studied as the anode materials for direct borohydride–hydrogen peroxide ($NaBH_4/H_2O_2$) fuel cells at temperatures of 25–55°C. Polarization curves have been recorded with the aim to evaluate the fuel cell performance using the prepared CoB and that modified with Pt crystallites as the anode catalysts.

For all catalysts (pure CoB and PtCoB) investigated, the peak power density values increase consecutively with the increment in temperature from 25°C up to 55°C. The values from 86–146 mV cm^{-2} and 146–234 mV cm^{-2} were determined for pure CoB and PtCoB catalysts, respectively. The highest specific peak power density of 21.5 kWg_{Pt}^{-1} was achieved at 55°C temperature when the PtCoB catalyst with the Pt loading of 9.8 $\mu g_{Pt} cm^{-2}$ was employed as the anode catalyst in the $NaBH_4/H_2O_2$ single fuel cell.

Keywords: Pt, CoB, morpholine borane, fuel cell, $NaBH_4/H_2O_2$

INTRODUCTION

Search of alternative energy sources is getting more and more active when the traditional energy resources are decreasing. Fuel cells are environment-friendly energy sources, in which the chemical energy is turned into the electrical one [1–3]. They are intensively investigated as cleaner power generators compared to fossil fuel-based technologies. The borohydride fuel cell, which uses an alkaline fuel cell system fed with an aqueous solution of (usually sodium) borohydride ($NaBH_4$) and oxygen as an oxidant (DBFC, $NaBH_4/O_2$), has recently proven to be of a great interest, as it displays a high theoretical specific energy density (9.3 Wh g^{-1}) [4, 5]. Operating of DBFC is based on the ongoing reactions at the anode and cathode, e.g. oxidation of sodium borohydride and reduction of oxygen, respectively. At the anode of the fuel cell, sodium borohydride is oxidized according to Eq. 1:

$$BH_4^- + 8OH^- \rightarrow BO_2^- + 6H_2O + 8e^-, \qquad (1)$$
$$E^0_a = -1.24 \text{ V vs SHE.}$$

At the cathode of the same fuel cell, oxygen is reduced according to Eq. 2:

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$$2O_2 + 4H_2O + 8e^- \rightarrow 8OH^-,$$

$$E^0_c = +0.40 \text{ V vs SHE.} \quad (2)$$

The overall cell reaction in DBFC is

$$BH_4^- + 2O_2 \rightarrow BO_2^- + 2H_2O,\ E^0_{cell} = +1.64 \text{ V.} \quad (3)$$

Furthermore, the $NaBH_4/O_2$ fuel cell exhibits a higher theoretical voltage of 1.64 V (Eq. 3), compared with that of fuel cell systems, such as CH_3OH/O_2 (1.19 V), H_2/O_2 (1.23 V), $HCOOH/O_2$ (1.45 V), and N_2H_4/O_2 (1.56 V) [6]. The use of hydrogen peroxide (H_2O_2) in the both alkaline and acidic media as an oxidant in the DBFC system (named as $NaBH_4/H_2O_2$) increases the theoretical cell voltage and specific energy density of this fuel cell as compared with those of the $NaBH_4/O_2$ fuel cell [6]. Since the electrochemical reduction of H_2O_2 proceeds without formation of poisonous products, the use of H_2O_2 as an oxidant in the fuel cells is attractive for further research. The reduction of H_2O_2 in an alkaline medium at the cathode of the $NaBH_4/O_2$ fuel cell is given in Eq. 4 [7], and, therefore, the overall cell reaction can be written as Eq. 5:

$$4HO_2^- + 4H_2O + 8e^- \rightarrow 12OH^-,$$

$$E^0 = 0.87 \text{ V vs SHE;} \quad (4)$$

$$4HO_2^- + BH_4^- \rightarrow 4OH^- + BO_2^- + 2H_2O,$$

$$E_0 = 2.11 \text{ V.} \quad (5)$$

In acidic media, the reduction of H_2O_2 at the cathode of fuel cell occurs according to Eq. 6:

$$4H_2O_2 + 4H^+ + 8e^- \rightarrow 8H_2O,$$

$$E^0_c = +1.77 \text{ V vs SHE.} \quad (6)$$

The overall cell reaction in this situation provides a theoretical cell potential of 3.01 V according to Eq. 7:

$$BH_4^- + 4H_2O_2 \rightarrow BO_2^- + 6H_2O,$$

$$E^0_{cell} = +3.01 \text{ V.} \quad (7)$$

The theoretical cell voltage of the $NaBH_4/H_2O_2$ fuel cell when H_2O_2 is used in both the alkaline and acidic media as an oxidant is 2.11 and 3.01 V, respectively, which is almost 0.5 and 1.4 V higher than that of the $NaBH_4/O_2$ fuel cell (1.64 V) [7].

Electrocatalysts have an important role in the performance of $NaBH_4/O_2$ and $NaBH_4/H_2O_2$ fuel cells [8–30]. The efficiency of the catalyst depends on different parameters, e.g. on the nature of the substrate, the composition of the catalysts and the size of the nanoparticles formed. The main attention is paid to the search of new nanostructured substances, which will be able to increase the performance of fuel cells. In the past years the researchers have explored different metals as anode catalysts in the fuel cells. It is well known that the use of Pt [10, 13, 14, 16, 17, 19–24] and Au [18, 25–29] alloys catalysts enhances the technological performance of the $NaBH_4/H_2O_2$ fuel cell, however, the noble metal catalysts make it relatively expensive. Therefore, many researchers have been interested in exploring lower-cost substitutes. It was shown that the Pt or Au composites with transition metals, e.g. Co, Ni, Cu and Fe, have demonstrated higher activity and stability for the oxidation of sodium borohydride than the pure Pt or Au catalysts [8, 13, 14, 20–29]. In our previous study [19] it was shown that the CoB catalysts modified with small amounts of Pt crystallites in the range of ca. 10–14 $\mu g_{Pt}\ cm^{-2}$ exhibit a high electrocatalytic activity for the electrooxidation of sodium borohydride. The catalysts were prepared using the electroless metal plating and galvanic displacement methods. In the present study, we investigated the performance of the sodium borohydride–hydrogen peroxide ($NaBH_4/H_2O_2$) fuel cell at different temperatures by employing the CoB catalysts modified with small amounts of Pt crystallites as the anode material.

EXPERIMENTAL

Fabrication and characterization of catalysts

The catalysts of CoB and that modified with small amounts of Pt crystallites (denoted as PtCoB/Cu) were the same as described in our previous study [19]. Briefly, the CoB layers were deposited on the Cu sheets (denoted as CoB/Cu) with a geometric area of 2 cm^2 using morpholine borane ($C_4H_8ONH{\cdot}BH_3$) as a reducing agent. The electroless cobalt-plating bath consisted of 0.05 M $CoSO_4$ and 0.05 M $C_4H_8ONH{\cdot}BH_3$. The bath operated at pH 7 and at 30°C temperature. The deposition time of the CoB layers on the Cu sheets was 100 min. Then, the galvanic displacement method was used for the deposition of Pt crystallites on the CoB surface [19]. Dipping of the prepared CoB/Cu electrodes into the 1 mM H_2PtCl_6 solution for 10, 30 and 60 s results in the deposition of Pt crystallites of ~10–25, 20–35, and 20–45 nm, respectively, on the surface of CoB/Cu. It was found that the PtCoB/Cu catalysts were prepared with Pt loadings of 9.8, 10.6 and 14.4 μg_{Pt} cm^{-2}. The prepared catalysts were used as the anodes for direct $NaBH_4/H_2O_2$ fuel cell test measurements without any further treatment.

Fuel cell test experiments

Single fuel cell tests were carried out in the lab-scale $NaBH_4/H_2O_2$ by employing the prepared CoB/Cu or PtCoB/Cu electrodes as the anodes and a Pt sheet as the cathode. Each compartment of the cell contained 100 mL of a static electrolyte solution, with the anolyte containing 1 M $NaBH_4$ + 4 M NaOH and the catholyte containing 1.5 M HCl + 5 M H_2O_2. A Nafion N117 membrane was used to separate the anodic and cathodic compartments of the single direct $NaBH_4/H_2O_2$ fuel cell [11]. 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 25–55°C temperatures.

RESULTS AND DISCUSSION

In the present study, the performance of a direct alkaline $NaBH_4/H_2O_2$ single fuel cell has been investigated by using the catalysts of CoB and CoB modified with small amounts of Pt crystallites as the anode materials. The low-cost electroless metal deposition and galvanic displacement methods were used for the preparation of the CoB/Cu and PtCoB/Cu catalysts. Figure 1 presents the fuel cell polarization curves and the corresponding power density curves against the current density by employing the CoB/Cu as the anode catalyst at 25–55°C temperatures. The fuel cell displayed an open circuit voltage of ca. 1.8 V. As seen, peak power density values increase with the increase in temperature from 25 up to 55°C. The obtained values ranged from 86 to 145 mW cm^{-2} for the CoB/Cu catalyst used as the anode in the $NaBH_4/H_2O_2$ single fuel cell (Fig. 1).

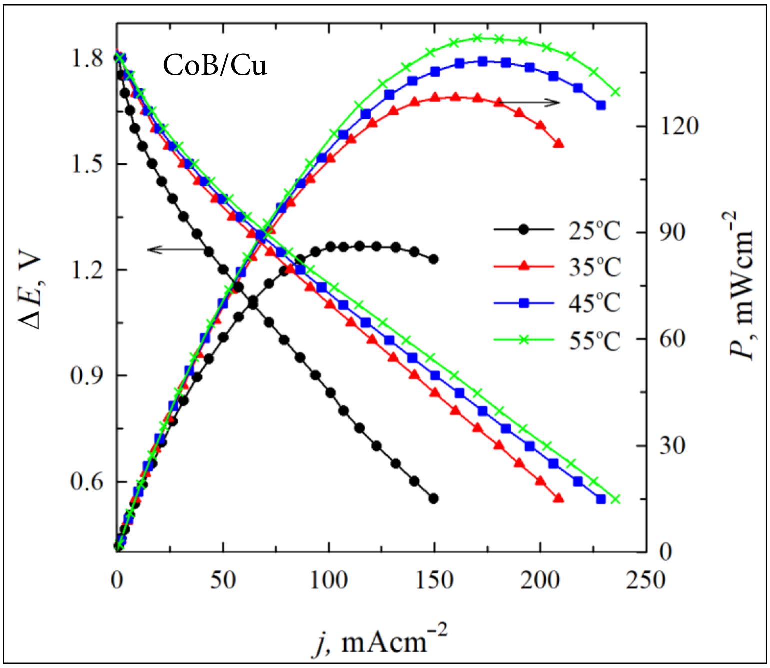

Fig. 1. Cell polarization and power density curves for the $NaBH_4/H_2O_2$ single fuel cell using the CoB/Cu catalysts as the anodes with the anolyte consisting of the 1 M $NaBH_4$ + 4 M NaOH and 5 M H_2O_2 + 1.5 M HCl catholyte and operation temperatures ranging from 25 to 55°C

The fuel cell polarization curves and the corresponding power density curves against the current density by employing different PtCoB/Cu catalysts as the anode catalysts at 25–55°C temperatures in the range of 25–55°C are shown in Fig. 2. The summarized data are given in Table 1. The fuel cell displayed an open circuit voltage of ca. 1.9 V. The highest peak power densities of 211, 222 and 234 mW cm^{-2} are achieved at 55°C temperature when employing the PtCoB/Cu anode catalysts with Pt loadings of 9.8, 10.6 and 14.4 μg_{Pt} cm^{-2}, respectively. The measured peak power densities of the $NaBH_4/H_2O_2$ fuel cell with different PtCoB/Cu anodes are increased with the increase in the temperature from 25 to 55°C. The values obtained ranged from 146 to 234 mW cm^{-2} for the PtCoB/Cu catalysts with the Pt loading ranging from 9.8 to 14.4 μg_{Pt} cm^{-2}. It has been determined that power density values are significantly higher in the case of the investigated PtCoB/Cu catalysts as compared to those of pure CoB/Cu (Figs. 1, 2).

Peak power densities at 25°C temperature for PtCoB/Cu with the Pt loadings of 9.8, 10.6 and 14.4 μg_{Pt} cm^{-2} were ca. 1.7, 1.9 and 2.0, respectively, times higher as compared with those of pure CoB/Cu. Ca. 1.4, 2.6 and 2.7 times higher peak power densities of the $NaBH_4/H_2O_2$ fuel cell at 55°C temperature were obtained on the PtCoB/Cu with the Pt loadings of 9.8, 10.6 and 14.4 μg_{Pt} cm^{-2}, respectively, compared with those for CoB/Cu.

For evaluation of the activity of the prepared PtCoB/Cu catalysts used as the anodes in the $NaBH_4/H_2O_2$ fuel cell, the power density values were normalized by the Pt loadings for each catalyst. The obtained values represent the specific power densities, i.e. power density per gram of Pt in the PtCoB/Cu catalysts. The summarized data are given in Table 1 and Fig. 3. It can be observed that the $NaBH_4/H_2O_2$ fuel cell, in which the PtCoB/Cu catalysts with different Pt loadings were employed as the anodes, produces from 12.3 to 14.9 kW g_{Pt}^{-1} specific power densities at room temperature (25°C), whereas from 16.3 to 21.5 kW g_{Pt}^{-1} at 55°C temperature (Fig. 3).

Comparison of the operation parameters of the $NaBH_4/H_2O_2$ fuel cell with the employed anode CoB/Cu and PtCoB/Cu catalysts prepared in this study and those presented in literature, such as PtM-based [10, 21, 23, 24] or AuM-based [18, 25] electrodes, is summarized in Table 2. It is clearly seen that the power density output and specific power density of the $NaBH_4/H_2O_2$ fuel cell using the investigated CoB/Cu catalysts modified with small amounts of Pt crystallites as the anodes are significantly higher than those of PtM (M = Ni,

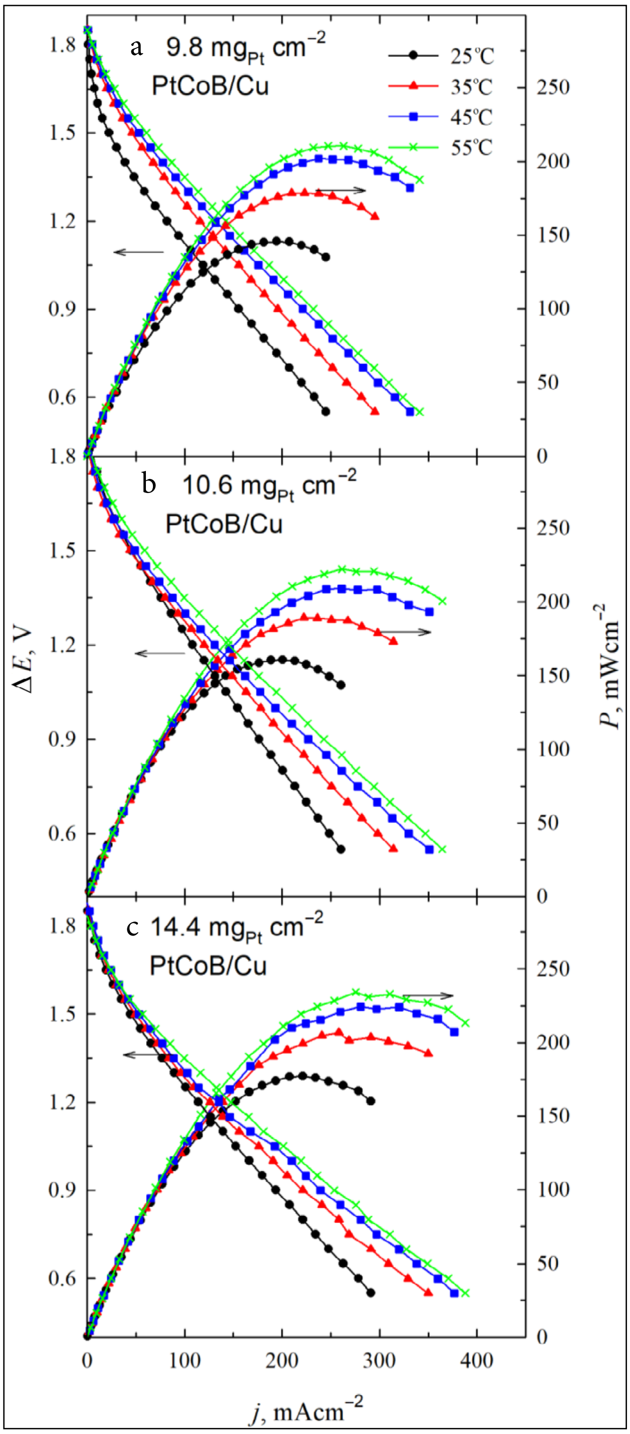

Fig. 2. Cell polarization and power density curves for the $NaBH_4/H_2O_2$ single fuel cell using the PtCoB/Cu catalysts with the Pt loading of 9.8 (a), 10.6 (b) and 14.4 (c) μg_{Pt} cm^{-2} as the anodes with the anolyte consisting of the 1 M $NaBH_4$ + 4 M NaOH and 5 M H_2O_2 + 1.5 M HCl catholyte and operation temperatures ranging from 25 to 55°C

Table 1. **Fuel cell parameters of the $NaBH_4/H_2O_2$ single fuel cell employing the CoB/Cu and PtCoB/Cu anode catalysts**

T, °C	Catalysts	Pt loading, μg cm^{-2}	Peak power density, mW cm^{-2}	Current density at peak power density, mA cm^{-2}	Cell voltage at peak power density, V	Specific peak power density, kW g_{Pt}^{-1}
25	CoB/Cu	–	86.0	114.7	0.75	–
35			128.0	160.0	0.80	–
45			138.3	172.8	0.80	–
55			144.8	170.3	0.85	–
25	PtCoB/Cu	9.8	145.8	194.3	0.75	14.9
35			178.5	209.9	0.85	18.2
45			201.9	237.4	0.85	20.6
55			210.5	263.0	0.80	21.5
25		10.6	160.5	200.6	0.80	15.1
35			189.2	222.4	0.85	17.8
45			209.0	261.2	0.80	19.7
55			222.4	261.5	0.85	21.0
25		14.4	177.0	221.2	0.80	12.3
35			206.7	258.3	0.80	14.4
45			224.4	280.5	0.80	15.6
55			234.3	275.5	0.85	16.3

Table 2. **Operation parameters of the $NaBH_4/H_2O_2$ fuel cell using different anode catalysts**

Anode	Cathode	*P*, mW cm^{-2}	Loading, mg cm^{-2}	Specific *P*, W g^{-1}	Anolyte	Catholyte	*T*, °C	Ref.
CoB/Cu	Pt	86.0	–	–	1 M $NaBH_4$ + 4 M NaOH	5 M H_2O_2 + 1.5 M HCl	25	This work
PtCoB/Cu		145.8	0.0098 mg_{Pt} cm^{-2}	14900				
		160.5	0.0104 mg_{Pt} cm^{-2}	15100				
		177.0	0.0144 mg_{Pt} cm^{-2}	12300				
NiPt/C	Pt/C 0.5 mg cm^{-2}	60.10	1	60	2 M $NaBH_4$ + 2 M NaOH	2 M H_2O_2 + 0.5 M H_2SO_4	25	[10]
Pt_2Cu/NPC	Au/XC-72	89.0	0.024 mg_{Pt} cm^{-2}	3708	1 M $NaBH_4$ + 3 M NaOH	2 M H_2O_2 + 0.5 M H_2SO_4	25	[24]
$Pt_{67}Co_{33}$/C	Pt/C 0.9 mg cm^{-2}	79.7	0.9	114	1 M $NaBH_4$ + 3 M NaOH	2 M H_2O_2 + 0.5 M H_2SO_4	25	[21]
PtNi/G	Pt/G 0.5 mg cm^{-2}	60.4	1	60	1 M $NaBH_4$ + 2 M NaOH	2 M H_2O_2 + 0.5 M H_2SO_4	60	[23]
$Au_{45}Co_{55}$/C	Au/C	66.5	4.5	73.9	1 M $NaBH_4$ + 3 M NaOH	2 M H_2O_2 + 0.5 M H_2SO_4	25	[25]
Au@Co-B	$LaNi_{0.9}Ru_{0.1}O_3$	85.0	0.7	121	0.8 M KBH_4 + 6 M KOH (membraneless, air cathode)		25	[18]

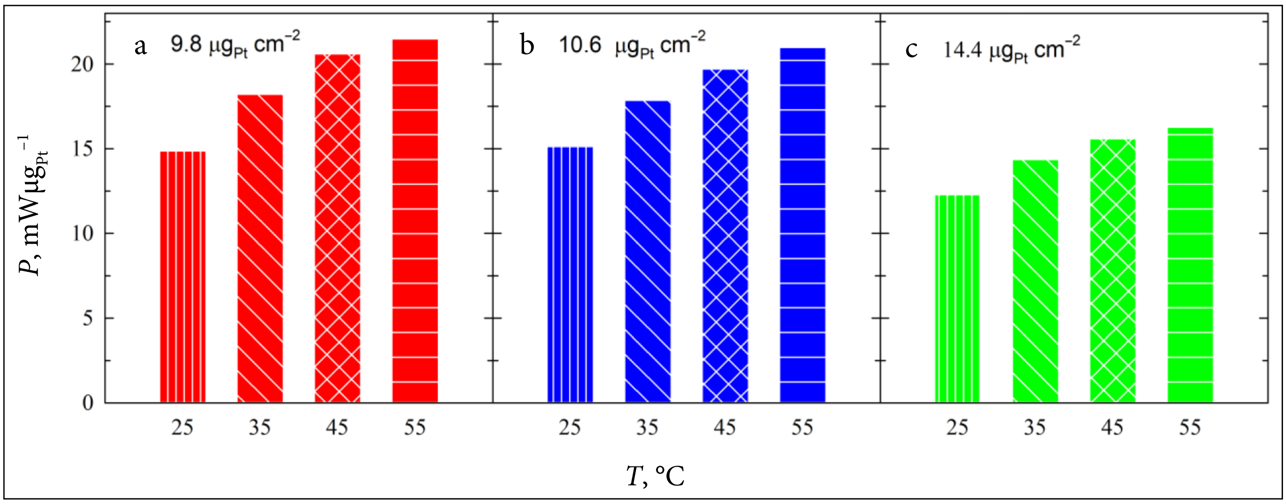

Fig. 3. The dependence of specific peak power densities of the $NaBH_4/H_2O_2$ fuel cell on temperature by using the PtCoB/Cu catalysts with the Pt loadings of 9.8 (a), 10.6 (b) and 14.4 (c) µg$_{Pt}$ cm^{-2}

Co, Cu) [10, 21, 23, 24] or AuCo [18, 25] used as the anode catalysts (Table 2).

The decoration of the CoB/Cu electrodes with small amounts of Pt crystallites in the range of 9–14 µg$_{Pt}$ cm^{-2} significantly enhances the performance of the $NaBH_4/H_2O_2$ fuel cell and seems to be promising anode materials for it.

CONCLUSIONS

The CoB and PtCoB catalysts formed by means of electroless cobalt deposition and galvanic displacement by Pt were tested as the anodes for the direct borohydride–hydrogen peroxide fuel cell. The assembled $NaBH_4/H_2O_2$ fuel cells using the CoB and PtCoB catalysts with the Pt loading in the range of ca. 10–14 µg$_{Pt}$ cm as the anodes exhibit an open circuit voltage of ca. 1.8–1.9 V. For all catalysts investigated (pure CoB and PtCoB), the peak power density values increase consecutively with the increment in temperature from 25°C up to 55°C. The values from 86–146 mV cm^{-2} and 146–234 mV cm^{-2} were determined for the pure CoB and PtCoB catalysts, respectively. The decoration of the CoB/Cu electrodes with small amounts of Pt significantly enhances the performance of the $NaBH_4/H_2O_2$ fuel cell. The highest peak power density up to 177 mW cm^{-2} at room temperature has been obtained with the CoB/Cu catalyst, which has the Pt loading of 14.4 µg$_{Pt}$ cm^{-2}. Furthermore, the greatest specific peak power density of 21.5 kW g$_{Pt}$$^{-1}$ has been obtained on the PtCoB/Cu anode catalyst with the Pt loading of 9.8 µg$_{Pt}$ cm^{-2} at 55°C temperature.

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PLATINOS-KOBALTO-BORO TAIKYMAS KAIP ANODO MEDŽIAGOS NATRIO BOROHIDRIDO-VANDENILIO PEROKSIDO KURO ELEMENTUOSE

Santrauka

Platinos-kobalto-boro (PtCoB) ir kobalto-boro (CoB) katalizatoriai buvo nusodinti ant vario paviršiaus taikant cheminio metalų nusodinimo ir galvaninio pakeitimo metodus. Suformuoti PtCoB/Cu ir CoB/Cu katalizatoriai buvo anodais integruojami tiesioginiame natrio borohidrido-vandenilio peroksido ($NaBH_4/H_2O_2$) kuro elemente. Pt elektrodas buvo naudojamas katodu. Anolitu pasirinktas 1 M $NaBH_4$ + 4 M NaOH tirpalas, o katolitu – 5 M H_2O_2 + 1,5 M HCl. Kuro elemento parametrų matavimai buvo atlikti esant 25, 35, 45 ir 55 °C temperatūroms. Atviros grandinės sąlygomis celės įtampa yra apie 1,8–1,9 V. Naudojant PtCoB/Cu anodo katalizatorius $NaBH_4/H_2O_2$ kuro elemente, kai nusodintos Pt įkrova yra 9,8–14,4 $\mu g_{Pt}\ cm^{-2}$, buvo gautos didesnės galios tankio vertės, lyginant jas su CoB/Cu anodo katalizatoriumi. Didžiausias galios tankis 177 mW cm^{-2} esant 25 °C temperatūrai, 221 mA cm^{-2} srovės tankiui ir 0,80 V celės įtampai buvo gautas panaudojant PtCoB/Cu katalizatorių su nusodintos Pt įkrova 14,4 $\mu g_{Pt}\ cm^{-2}$. Didžiausias specifinis galios tankis 21,5 kW g_{Pt}^{-1} gautas naudojant PtCoB/Cu katalizatorių su nusodintos Pt įkrova 9,8 $\mu g_{Pt}\ cm^{-2}$ esant 55 °C temperatūrai.