

An alkaline direct borohydride fuel cell with hydrogen peroxide as oxidant

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Abstract

A novel alkaline direct borohydride fuel cell (ADBFC) using varying concentrations of hydrogen peroxide as oxidant and sodium borohydride with sodium hydroxide, each of differing concentration, as fuel is reported. A peak power density of ca. 150 mW cm^{-2} at a cell voltage of 540 mV can be achieved from the optimized ADBFC operating at 70°C .

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1. Introduction

Proton exchange membrane fuel cells (PEFCs) are subject to problems of carbon monoxide poisoning of anode [1–3] when using a reformer, and hydrogen storage when using a direct fuel. Therefore, alternative hydrogen-carrying liquid fuels, such as methanol, have attracted attention for fuelling PEFCs directly with methanol [4–6]. Such cells are referred to as direct methanol fuel cells (DMFCs). This use of methanol highly simplifies the engineering problems at the front end of the fuel cell and hence reduces system complexity and lowers cost [7]. Nevertheless, DMFCs have limitations of low open-circuit potential, low electrochemical activity, and methanol cross-over [8].

Attempts are being made to overcome the above limitations by using other hydrogen-carrying liquid fuels such as borohydrides [9–16], e.g., sodium borohydride it at has a

capacity of 5.67 Ah g^{-1} and a hydrogen content of about 11 wt.%. Amendola et al. [12,13] were the first to report an OH^- -ion conducting anion exchange membrane-based borohydride–air fuel cell, which had a power density close to 60 mW cm^{-2} at 70°C . It was found that the cell suffered from borohydride cross-over as BH_4^- -ions could pass through the anion-exchange membrane. In additions, it is mandatory to scrub carbon dioxide from the air inlet of such a fuel cell to avoid carbonate fouling. In order to mitigate borohydride cross-over, Suda [15] and Li et al. [16,17] adopted a fuel-cell structure with a Nafion membrane as the electrolyte in order to separate the fuel from the cathode. But even in the borohydride–air cell reported by Suda [15], it would be compulsory to scrub carbon dioxide, not only to avoid carbonate fouling but also to prevent accumulation of alkali in the cathode pores so as to facilitate oxidant flux [15].

This communication describes an alkaline direct borohydride fuel cell (ADBFC) that uses hydrogen peroxide as oxidant to prevent carbonate fouling of the cathode. The use of hydrogen peroxide as oxidant in fuel cells also extends their operation to locations with limited air convection, e.g., underwater applications.

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2. Experimental

2.1. Preparation of AB₅ and AB₂ alloys

Various AB₅ and AB₂ alloys were prepared by arc-melting stoichiometric amounts of the constituent metals in a water-cooled copper crucible under argon atmosphere [18–23]. The alloy ingots were mechanically pulverized to fine powders. The various AB₅ alloys employed in this study were M_mNi_{4.5}Al_{0.5}, M_mNi_{3.2}Al_{0.2}Mn_{0.6}Co_{1.0}, M_mNi_{3.55}Al_{0.3}Mn_{0.4}Co_{0.75}, and M_mNi_{3.2}Al_{0.2}Mn_{0.6}B_{0.03}Co_{1.0}, where M_m (Misch metal) comprises La-30 wt.%, Ce-50 wt.%, Nd-15 wt.%, Pr-5 wt.%. The AB₂ alloy had the composition Zr_{0.9}Ti_{0.1}V_{0.2}Mn_{0.6}Cr_{0.05}Co_{0.05}Ni_{1.2}.

2.2. Physical characterization of alloy catalysts

Various AB₅ and AB₂ alloys were characterized by recording their powder X-ray diffraction (XRD) patterns on a Siemens D-5005 X-ray diffractometer using Cu K α radiation. The alloys were also subjected to energy dispersive analysis by X-rays (EDAX) employing a Jeol JSM-840A scanning electron microscope to determine the composition of their constituent elements.

2.3. Preparation of carbon-supported platinum (Pt/C) catalyst

The sulfito-complex route [24–27] was adopted to prepare 60 wt.% platinumized carbon. The required amount of Vulcan-XC 72R carbon was suspended in distilled water

and agitated in an ultrasonic water bath to form the slurry. The required amount of Na₆Pt(SO₃)₄ was dissolved in 1 M H₂SO₄ and diluted with distilled water. The solution was added dropwise to the carbon slurry with constant stirring at 80 °C. This was followed by the addition of 8.9 M H₂O₂ with the temperature maintained at 80 °C, which resulted in vigorous gas evolution. The solution was further stirred for 1 h. Subsequently, platinumized carbon was obtained by adding 1 wt.% formic acid solution, which was filtered, washed copiously with hot distilled water, and dried in an air oven at 80 °C for 2 h.

2.4. Electrochemical characterization of borohydride fuel cells

For the electrochemical characterization of the ADBFCs, membrane electrode assemblies (MEAs) were obtained by sandwiching a pre-treated Nafion[®]-117 polymer electrolyte membrane between the anode and cathode. To prepare the anode catalyst layer, a slurry of AB₅ or AB₂ alloy obtained by ultra-sonicating the required amount of alloy with 5 wt.% Vulcan XC-72R carbon and 7 wt.% of Nafion[®] in iso-propyl alcohol was pasted on carbon paper (Toray TGP-H-090) of 0.28 mm thickness. The loading of alloy catalyst was 5 mg cm⁻² and was kept identical for all the MEAs. The cathode comprises a backing layer, a gas-diffusion layer, and a reaction layer. A teflonized (20 wt.% Teflon) carbon paper (Toray TGP-H-090) of 0.28 mm thickness was employed as the backing layer for the cathode. To prepare the gas-diffusion layer, Vulcan-XC 72R carbon was suspended in water and agitated in an ultrasonic water bath. To this, 13 wt.% Teflon

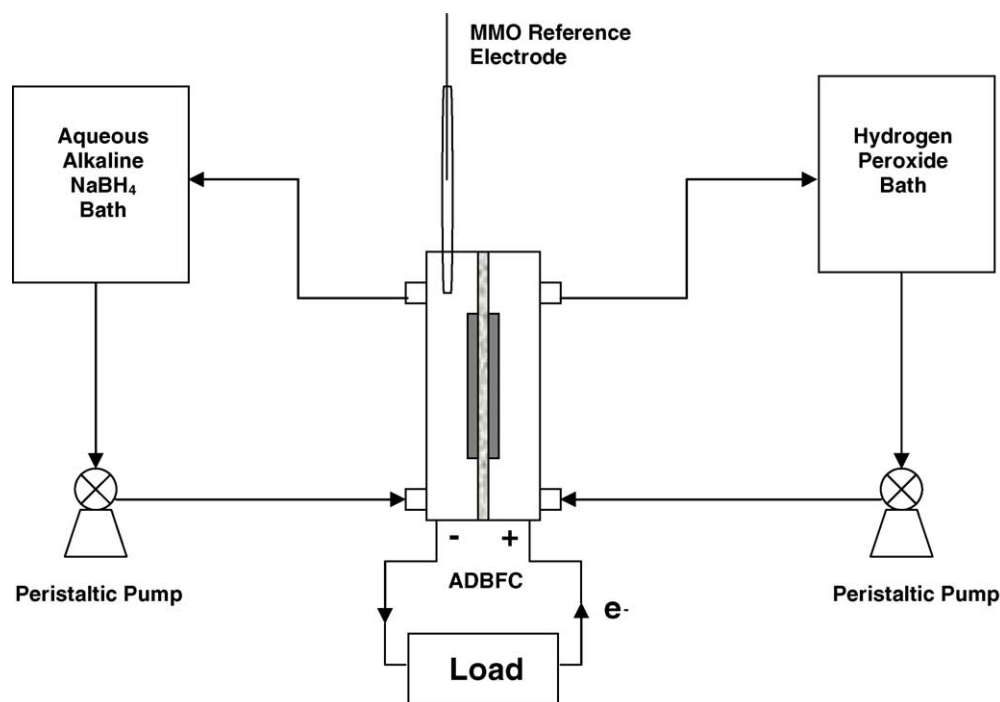


Fig. 1. Schematic representation of ADBFC operating with hydrogen peroxide as oxidant.

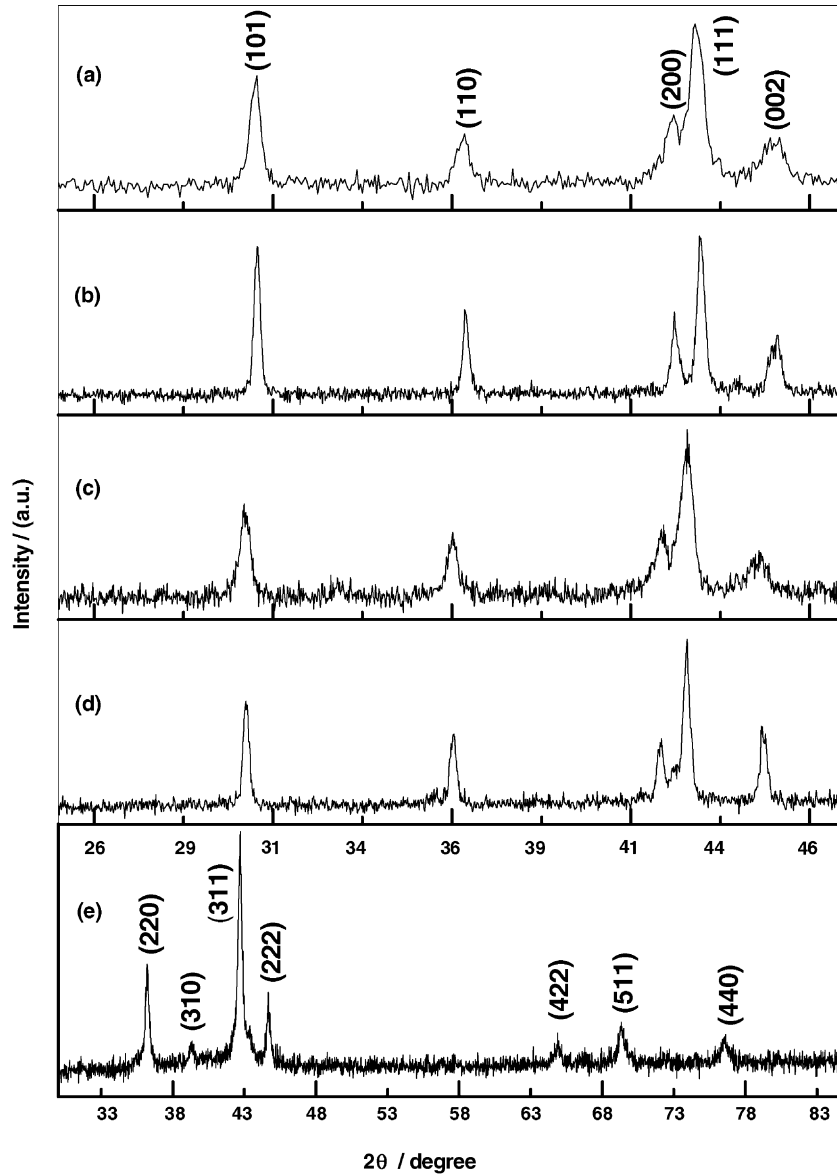


Fig. 2. X-ray powder diffraction patterns for M_m -based AB_5 -group alloys: (a) $M_mNi_{3.55}Al_{0.3}Mn_{0.4}Co_{0.75}$; (b) $M_mNi_{4.5}Al_{0.5}$; (c) $M_mNi_{3.2}Al_{0.2}Mn_{0.6}B_{0.03}Co_{1.0}$; (d) $M_mNi_{3.2}Al_{0.2}Mn_{0.6}Co_{1.0}$; (e) $Zr_{0.9}Ti_{0.1}V_{0.2}Mn_{0.6}Cr_{0.05}Co_{0.05}Ni_{1.2}$ AB_2 -group.

(Fluon GP-2) suspension was added with continuous agitation. The required amount of cyclohexane was then added to it dropwise. The resultant slurry was spread on to a teflonized carbon paper and dried in an air oven at $80^\circ C$ for 2 h. To prepare the reaction layer, the required amount of the catalyst (60 wt.% Pt/C) was suspended in iso-propyl alcohol. The

mixture was agitated in an ultrasonic water bath, and 7 wt.% of Nafion[®] solution was added to it with continuing agitation for 1 h. The catalyst ink thus obtained was coated on to the gas-diffusion layer of the electrode. The cathode contained 60 wt.% Pt/C catalyst with a platinum loading of 1 mg cm^{-2} , which was kept identical in all the MEAs. A Nafion loading

Table 1
Structural details and lattice parameters of various AB_5 -group and AB_2 -group alloys

S. no.	Composition	Structure type	Lattice parameters
1	$M_mNi_{3.55}Al_{0.3}Mn_{0.4}Co_{0.75}$ (AB_5 -group)	CaCu ₅ -type, space group: $P6/mmm$	$a = 4.99 \text{ \AA}$; $c = 4.00 \text{ \AA}$
2	$M_mNi_{4.5}Al_{0.5}$ (AB_5 -group)	CaCu ₅ -type, space group: $P6/mmm$	$a = 4.96 \text{ \AA}$; $c = 4.02 \text{ \AA}$
3	$M_mNi_{3.2}Al_{0.2}Mn_{0.6}B_{0.03}Co_{1.0}$ (AB_5 -group)	CaCu ₅ -type, space group: $P6/mmm$	$a = 4.99 \text{ \AA}$; $c = 4.04 \text{ \AA}$
4	$M_mNi_{3.2}Al_{0.2}Mn_{0.6}Co_{1.0}$ (AB_5 -group)	CaCu ₅ -type, space group: $P6/mmm$	$a = 4.99 \text{ \AA}$; $c = 4.06 \text{ \AA}$
5	$Zr_{0.9}Ti_{0.1}V_{0.2}Mn_{0.6}Cr_{0.05}Co_{0.05}Ni_{1.2}$ (AB_2 -group)	C-15 cubic laves phase	$a = 7.04 \text{ \AA}$

Table 2
Compositions of various AB₅- and AB₂-group alloy samples

S. no.	Nominal composition	Composition as determined by EDAX
1	MmNi _{3.55} Al _{0.3} Mn _{0.4} Co _{0.75}	Mm _{0.76} Ni _{3.71} Al _{0.34} Mn _{0.32} Co _{0.71}
2	MmNi _{4.5} Al _{0.5}	Mm _{0.89} Ni _{4.58} Al _{0.48}
3	MmNi _{3.2} Al _{0.2} Mn _{0.6} B _{0.03} Co _{1.0}	Mm _{1.2} Ni _{3.03} Al _{0.24} Mn _{0.59} B ₀ Co _{0.93}
4	MmNi _{3.2} Al _{0.2} Mn _{0.6} Co _{1.0}	Mm _{1.06} Ni _{3.31} Al _{0.27} Mn _{0.61} Co _{0.79}
5	Zr _{0.9} Ti _{0.1} V _{0.2} Mn _{0.6} Cr _{0.05} Co _{0.05} Ni _{1.2}	Zr _{0.85} Ti _{0.03} V _{0.17} Mn _{0.59} Cr _{0.04} Co _{0.05} Ni _{1.28}

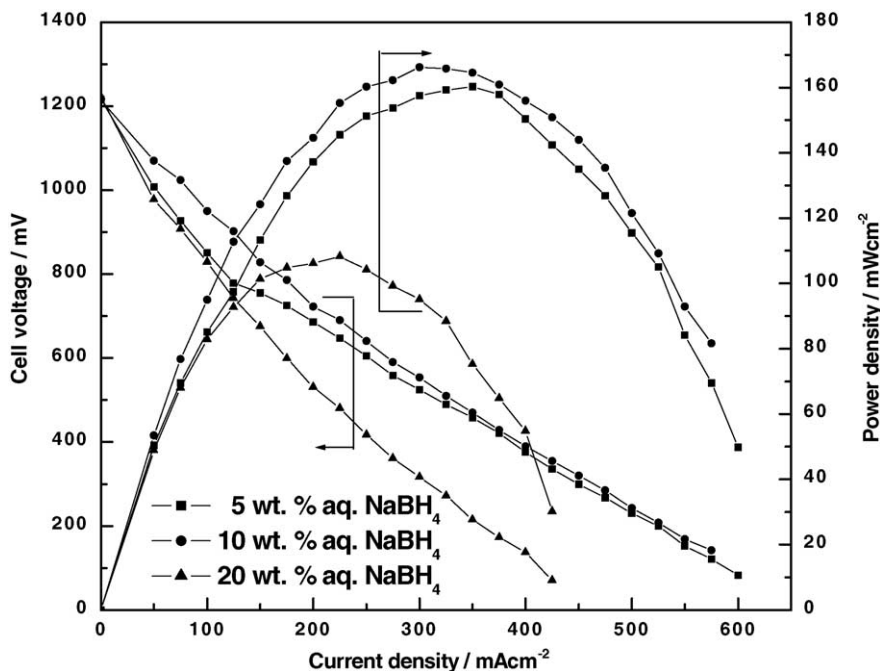


Fig. 3. Cell polarization data for ADBFC-1 operating at 70 °C with varying concentrations of aqueous NaBH₄ in 20 wt.% aqueous NaOH at anode and 8.9 M H₂O₂ at cathode.

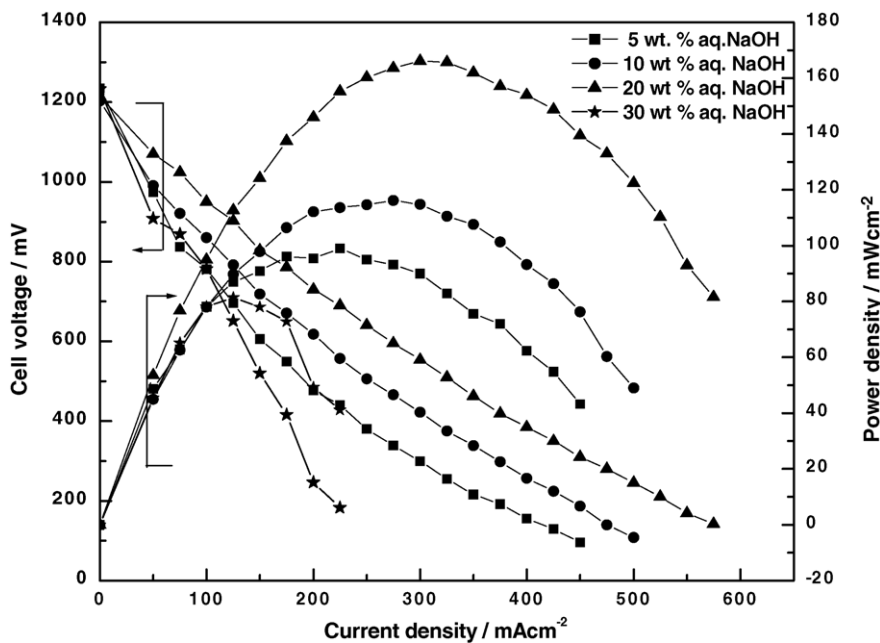


Fig. 4. Cell polarization data for ADBFC-1 operating at 70 °C with 10 wt.% aq. NaBH₄ in varying concentrations of aqueous NaOH at anode and 8.9 M H₂O₂ at cathode.

of 0.25 mg cm^{-2} was applied to the surface of each electrode. The MEA was obtained by hot-pressing the cathode and anode on either side of a pre-treated Nafion[®]-117 membrane at 60 kg cm^{-2} and $125 \text{ }^\circ\text{C}$ for 3 min.

Liquid-feed ADBFCs were assembled with various MEAs. The anode and cathode of the MEA were contacted on their rear with gas/fluid flow-field plates that were machined from high-density graphite blocks in which channels were machined to achieve minimum mass-polarization in the ADBFCs. The ridges between the channels make electrical contact with the back of the electrode and conduct the current to the external circuit. The channels supply alkaline sodium borohydride solution to the anode and hydrogen peroxide to the cathode. Electrical heaters were placed behind each of the graphite blocks to heat the cell to the desired temperature. An alkaline solution of sodium borohydride was pumped to the anode chamber through a peristaltic pump. Hydrogen peroxide solution of varying concentration was introduced into the cathode chamber through another peristaltic pump. The graphite blocks were also provided with electrical contacts and tiny holes to accommodate thermocouples. An ADBFC is shown schematically in Fig. 1. After installing single cells in the test station, performance evaluation studies were initi-

ated. To humidify and modify the Nafion membrane into the Na^+ -form, 20 wt.% NaOH aqueous solution was circulated at $80 \text{ }^\circ\text{C}$ for 3 h prior to the operation of the ADBFC.

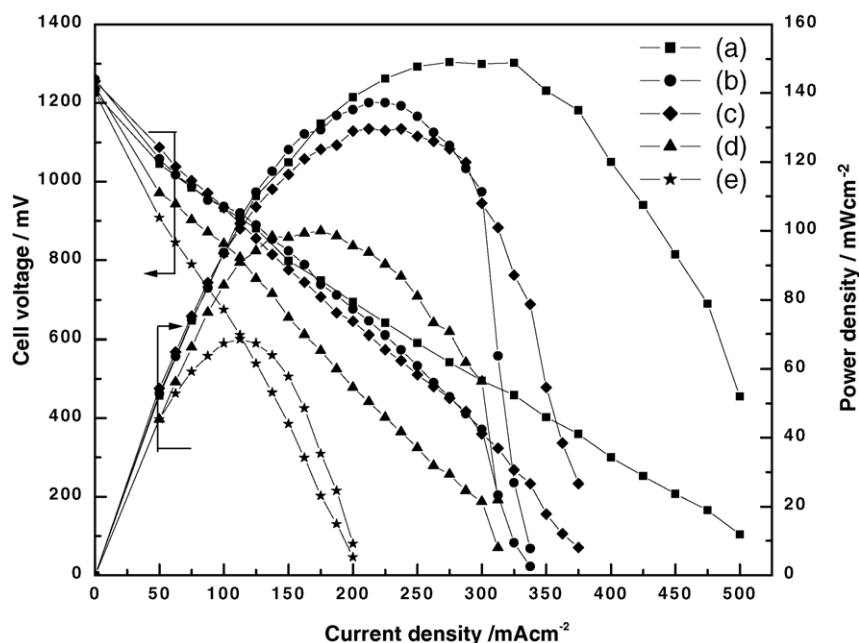
Galvanostatic polarization data for various ADBFCs in the temperature range between 30 and $70 \text{ }^\circ\text{C}$ were recorded by circulating varying concentrations of alkaline aqueous sodium borohydride solution (3 ml min^{-1}), and varying concentrations of hydrogen peroxide solution (5.5 ml min^{-1}) in the anode and cathode chambers, respectively. Anode and cathode polarization data at various temperatures for the best-performing ADBFC were also obtained by employing a Hg|HgO, OH^- (MMO) reference electrode.

In order to determine the Faradaic efficiency of oxidation, an alkaline solution containing 1.26×10^{-3} moles of sodium borohydride was injected into the anode chamber of the fuel cell. Subsequently, the cell was operated for 60 min at a load current density of 50 mA cm^{-2} at $70 \text{ }^\circ\text{C}$. The amount of hydrogen liberated during this experiment was obtained by collecting it at the anode outlet by downward displacement of water in a graduated tube. The amount of sodium borohydride left in the analyte was determined by collecting hydrogen gas evolved upon acidification to ascertain the mass balance of the reaction [28].

Table 3

Performance data at $70 \text{ }^\circ\text{C}$ for various ADBFCs

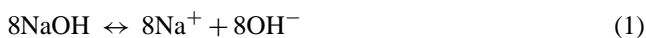
Cell no.	Anode catalyst loading	Cathode catalyst loading	Peak power density/ mW cm^{-2}
ADBFC-1	$\text{M}_m\text{Ni}_{3.55}\text{Al}_{0.3}\text{Mn}_{0.4}\text{Co}_{0.75}$ (5 mg cm^{-2})	60 wt.% Pt/C (1 mg cm^{-2} of Pt)	150
ADBFC-2	$\text{M}_m\text{Ni}_{4.5}\text{Al}_{0.5}$ (5 mg cm^{-2})	60 wt.% Pt/C (1 mg cm^{-2} of Pt)	130
ADBFC-3	$\text{M}_m\text{Ni}_{3.2}\text{Al}_{0.2}\text{Mn}_{0.6}\text{B}_{0.03}\text{Co}_{1.0}$ (5 mg cm^{-2})	60 wt.% Pt/C (1 mg cm^{-2} of Pt)	125
ADBFC-4	$\text{M}_m\text{Ni}_{3.2}\text{Al}_{0.2}\text{Mn}_{0.6}\text{Co}_{1.0}$ (5 mg cm^{-2})	60 wt.% Pt/C (1 mg cm^{-2} of Pt)	100
ADBFC-5	$\text{Zr}_{0.9}\text{Ti}_{0.1}\text{V}_{0.2}\text{Mn}_{0.6}\text{Cr}_{0.05}\text{Co}_{0.05}\text{Ni}_{1.2}$ (5 mg cm^{-2})	60 wt.% Pt/C (1 mg cm^{-2} of Pt)	70

Anode feed: 10 wt.% NaBH_4 in 20 wt.% aqueous NaOH; cathode feed: 8.9 M H_2O_2 .Fig. 5. Galvanostatic polarization data at $70 \text{ }^\circ\text{C}$ for: (a) ADBFC-1; (b) ADBFC-2; (c) ADBFC-3; (d) ADBFC-4; (e) ADBFC-5.

3. Results and discussion

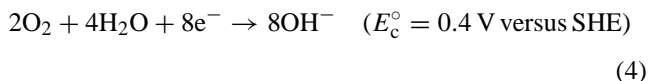
Powder X-ray diffraction patterns of various AB₅ and AB₂-group alloys employed in this study are shown in Fig. 2(a)–(e) [19–23]. The XRD patterns for AB₅-group alloys (Fig. 2(a)–(d)) were indexed in a hexagonal space group: *P6/mmm*. The structural details and lattice parameters for these alloys are given in Table 1. The powder XRD pattern (Fig. 2(e)) for the AB₂-group alloy of composition Zr_{0.9}Ti_{0.1}V_{0.2}Mn_{0.6}Cr_{0.05}Co_{0.05}Ni_{1.2} suggests that it crystallizes in C-15 cubic Laves phase with a lattice parameter, $a = 7.04 \text{ \AA}$. The average particle size of various alloy samples was found to be about 60 μm . The nominal and estimated compositions of the various alloys, as determined by energy dispersive analysis by X-rays (EDAX), are given in Table 2.

In the ADBFC, sodium borohydride is oxidized at the anode according to the following reactions:

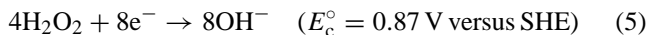


$$(E_a^\circ = -1.24 \text{ V versus SHE}) \quad (2)$$

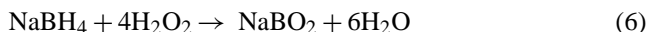
At the cathode, two reactions can take place: (i) decomposition of H₂O₂ to O₂ followed by electro-reduction of the latter [29] according to the reactions:



and (ii) direct electro-reduction of H₂O₂, i.e.,



Accordingly, the net cell reaction in such an ADBFC is:



The cell potentials for this ADBFC will be 1.64 and 2.11 V for cathode reactions (4) and (5), respectively.

The polarization data for ADBFCs obtained galvanostatically at various temperatures are shown in Figs. 3–7. In order to find an optimum concentration of the NaBH₄ fuel in the ADBFC, 8.9 M H₂O₂ solution was fed with a

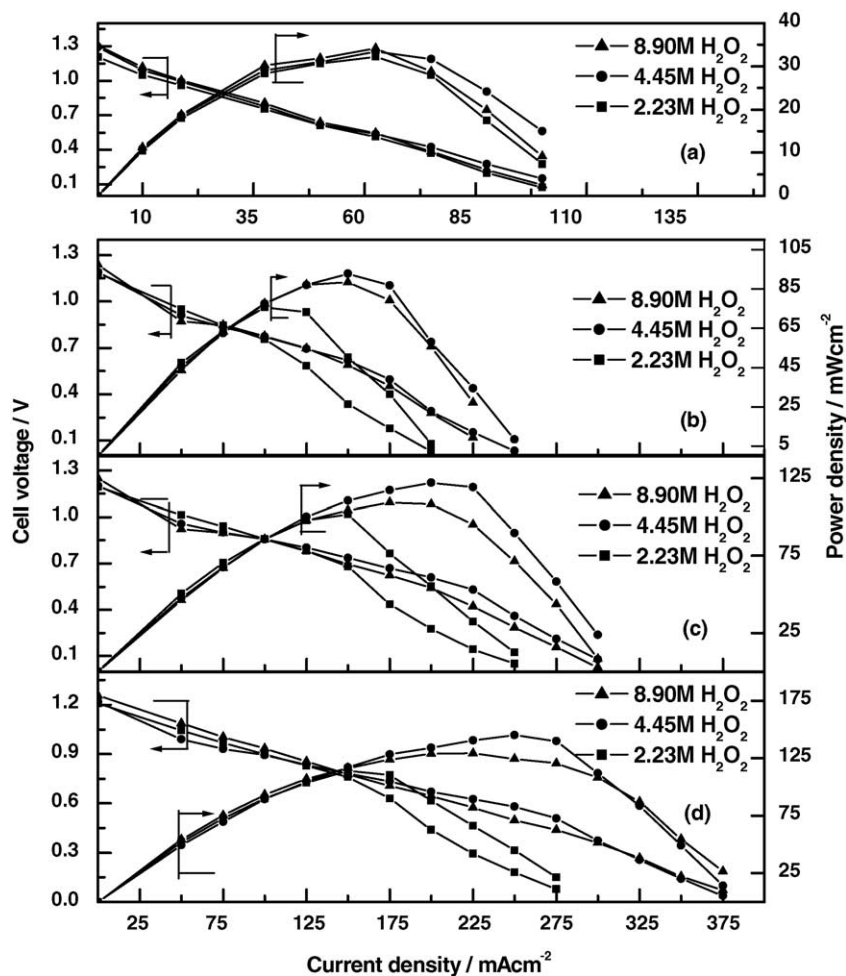


Fig. 6. Galvanostatic polarization data for ADBFC-1 with alkaline fuel of optimized concentration fed at anode and varying concentrations of oxidant fed at cathode at: (a) 30 °C; (b) 50 °C; (c) 60 °C; and (d) 70 °C.

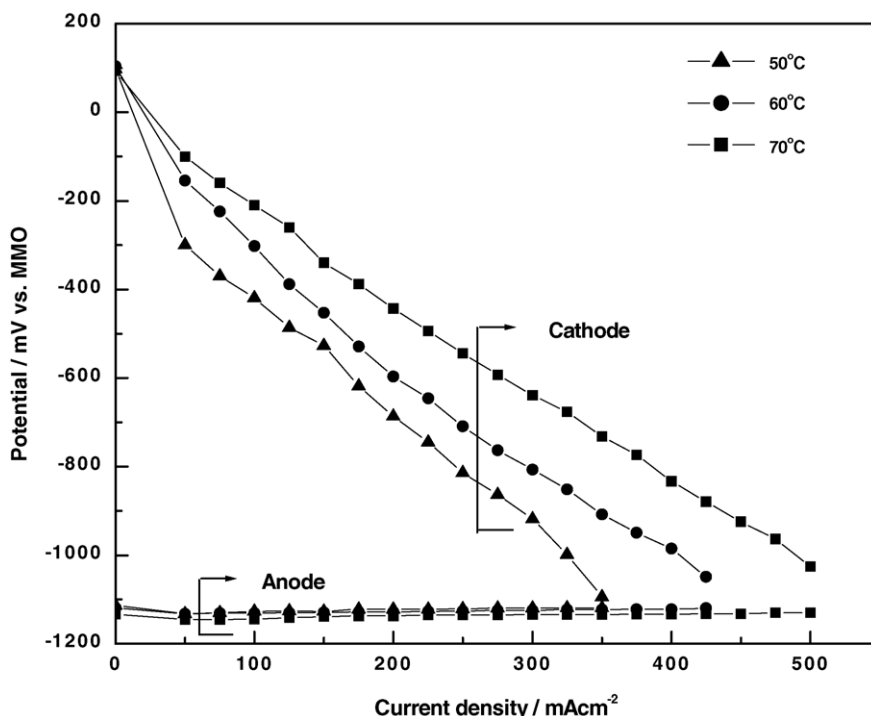


Fig. 7. Anode and cathode polarization data for ADBFC-1 at 50, 60 and 70 °C.

peristaltic pump at a feed rate of 5 ml min^{-1} to the cathode, while varying concentrations of NaBH_4 in 20 wt.% aq. NaOH were fed at the anode with another peristaltic pump at a feed rate of 3 ml min^{-1} ; the data are shown in Fig. 3. In accordance with Li et al. [16], a 10 wt.% NaBH_4 solution is found optimum for the ADBFC operation. In a similar manner, the optimum concentration of aqueous NaOH in the fuel was determined at 70 °C, and the results are given in Fig. 4. It is found that a 20 wt.% aq. NaOH is optimum for the ADBFC operation. The cell performance data at 70 °C for ADBFCs operating with 10 wt.% NaBH_4 in 20 wt.% aq. NaOH as fuel and 8.9 M H_2O_2 solution as oxidant, while employing various AB₅- and AB₂-group alloys as the anode catalyst, are presented in Fig. 5 and the data are summarized in Table 3. The best performance is found for ADBFC-1.

In order to find the optimum concentration of the oxidant in ADBFC, NaBH_4 in aqueous NaOH of optimum concentration was used at the anode while varying concentrations of H_2O_2 solution were fed to the cathode of ADBFC-1. The pertinent cell performance data at temperatures between 30 and 70 °C are shown in Fig. 6(a)–(d). ADBFC-1 delivers a maximum power density of about 35 mW cm^{-2} at 30 °C (Fig. 6(a)), and a maximum power density of ca. 150 mW cm^{-2} at 70 °C (Fig. 6(d)). Differences in the cell performance of ADBFCs operating with various concentrations of H_2O_2 are negligible at low current densities, but become noticeable at high current densities. A 4.45 M H_2O_2 solution is optimum for ADBFC operation in the temperature

range between 30 and 70 °C. It has been reported [16] that an ADBFC operating with oxygen feed at its cathode can have a peak power density as high as 160 mW cm^{-2} [16]. Accordingly, electrical performance data shown in Fig. 6 demonstrate that hydrogen peroxide is a viable substitute for gaseous oxygen in the operation of ADBFCs. The Faradaic efficiency of oxidation in the ADBFC at 70 °C is estimated to be 83%, and the rate of hydrogen evolved at the anode was measured to be $2 \times 10^{-7} \text{ mole s}^{-1}$.

Anode and cathode polarization data as shown in the temperature range between 50 and 70 °C were also obtained for ADBFC-1, Fig. 7. The data indicated that the ADBFC is cathode-limited. The enhancement in power density with increase in temperature could be due to the increased conductivity of the membrane electrolyte in addition to the increased reaction kinetics. It has been demonstrated by Li et al. [16] that using higher concentrations of NaBH_4 could deteriorate the cathode performance of an ADBFC due to the permeability of borohydride from anode to the cathode compartment in the cell. It is noteworthy, however, that the cross-over concentrations of borohydride in ADBFCs are much lower than methanol cross-over concentrations reported for DMFCs [30,31].

4. Conclusions

The study demonstrates that it is possible to assemble and operate an alkaline direct borohydride fuel cell with

hydrogen peroxide as oxidant with a maximum power density of about 150 mW cm^{-2} while operating at 70°C . The cell performance depends highly on the concentration of hydrogen peroxide since the power density may change by as high a value as 25 mW cm^{-2} with peroxide concentration. It is found that $4.45 \text{ M H}_2\text{O}_2$ is optimum for the ADBFCs reported here.

Acknowledgments

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