



High performance polymer chemical hydrogel-based electrode binder materials for direct borohydride fuel cells

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ABSTRACT

Novel, cost-effective, high-performance, and environment-friendly electrode binders, comprising polyvinyl alcohol chemical hydrogel (PCH) and chitosan chemical hydrogel (CCH), are reported for direct borohydride fuel cells (DBFCs). PCH and CCH binders-based electrodes have been fabricated using a novel, simple, cost-effective, time-effective, and environmentally benign technique. Morphologies and electrochemical performance in DBFCs of the chemical hydrogel binder-based electrodes have been compared with those of Nafion® binder-based electrodes. Relationships between the performance of binders in DBFCs with structural features of the polymers and the polymer-based chemical hydrogels are discussed. The CCH binder exhibited better performance than a Nafion® binder whereas the PCH binder exhibited comparable performance to Nafion® in DBFCs operating at elevated cell temperatures. The better performance of CCH binder at higher operating cell temperatures has been ascribed to the hydrophilic nature and water retention characteristics of chitosan. DBFCs employing CCH binder-based electrodes and a Nafion®-117 membrane as an electrolyte exhibited a maximum peak power density of about 589 mW cm⁻² at 70 °C.

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1. Introduction

A fuel cell that utilizes a borohydride compound, usually sodium borohydride (NaBH₄) in aqueous alkaline medium, directly as a fuel is referred to as direct borohydride fuel cell (DBFC). DBFCs possess some attractive features, such as high open circuit voltage (OCV) and high electrochemical performance at ambient conditions of temperature as well as pressure. Because of these desirable properties, DBFCs have become an attractive field of research [1–6]. An important constituent of an electrochemical energy conversion or storage device is the electrode binder. Polymers are generally employed as electrode binders in various types of fuel cells. Polymeric binders not only keep the electrode materials bound to the electrode substrates/current collectors but also help in achieving high fuel cell performance by establishing three-point contact among reactant (fuel/oxidant), electro-catalyst and polymer electrolyte membrane (PEM). Perfluorosulfonic acid (Nafion®) and poly(tetrafluoroethylene) (PTFE) are widely employed as electrode binders in fuel cells. On mass basis, the cost of Nafion® is about 500 times that of PTFE [7]. The high cost of Nafion® is one of the biggest hurdles in the successful commercialization of fuel cells. Considerable effort has been directed towards the development

of cost-effective PEMs as alternatives to Nafion® membranes. In contrast, research directed towards developing cost-effective and high performance electrode binder alternatives is limited. Nafion® ionomer has been employed as electrode binder in fuel cells that employ polymer membranes other than Nafion® as a membrane [8]. Jung et al. [9] studied DMFCs employing a membrane electrode assembly (MEA) comprising sulfonated poly(ether ether ketone)-based electrode binder as well as PEM. It has been observed that a DMFC employing MEA comprising sulfonated poly(arylene ether sulfone) (sPAES)-based electrode binder as well as PEM showed better performance stability than a DMFC employing MEA comprising Nafion® binder and a sPAES-based PEM [10]. The enhanced performance of the DMFC with MEA comprising electrode binder as well as PEM made of the same polymer has been ascribed to greater compatibility and better adhesion between PEM and electrodes in the MEA. Krishnan et al. [8] reported studies on polymer electrolyte fuel cells (PEFCs) that use sulfonated poly(ether sulfone)-based catalyst binder in conjunction with PEM made of the same polymers. Lee et al. [11] synthesized a hydrocarbon binder (SPI-BT) in sulfonated polyimide (SPI) triethyl ammonium salt-form and employed it in DMFCs. Muldoon et al. reported [12] synthesis and use as electrode binder of non-fluorinated sulfonated polyphosphazene in hydrogen/air fuel cells and observed that its performance was comparable to Nafion® binder. Wilson reported [13] polyvinyl alcohol (PVA) as electrode binder for fuel cells. However, PVA cannot be used as electrode binder in fuel cells that use aqueous

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solutions as fuel and/or oxidant because of its high solubility in water.

Hydrogels are 3-dimensional polymeric networks that absorb and retain water in amounts that greatly exceed their dry weight. Network formation and insolubility in aqueous media are due to the presence of chemical cross-links or physical entanglements. Chemical hydrogels are formed by chemical reaction between a polymer and a cross-linking reagent [14]. Polymer hydrogels can be used as ion-conduction media and hence as solid electrolytes in electrochemical devices. Glutaraldehyde-cross-linked PVA/PAA blends and gelatin hydrogels have been reported as solid electrolytes in electrochemical supercapacitors [15,16]. It was observed that PVA/PAA blend hydrogel electrolytes were stable in acidic, alkaline, and neutral media whereas gelatin hydrogel electrolyte was stable only in neutral media. Choudhury et al. reviewed various hydrogel polymer electrolytes with regard to their application in electrochemical supercapacitors [17].

PVA is a cheap, non-toxic, and chemically stable polymer [18]. The –OH group of PVA reacts with –CHO group of glutaraldehyde in the presence of a protic acid catalyst to form PCH at ambient conditions of temperature and pressure [19]. Chemical hydrogels, such as PCH, are insoluble in water and have high structural integrity as well as good thermal stability. A blend of poly(acrylic acid) (PAA) with PCH has been developed by Philipp et al. [20] for use as ion-exchange polymer. The same PVA/PAA hydrogel has been evaluated as a coating on graphite electrodes of electrochemical sensors [21]. Chitosan (CS) is a natural polymer that is obtained by alkaline deacetylation of chitin. Chitin is the second most abundant bio-polymer in nature after cellulose [22]. CS is weakly alkaline in nature and is soluble in weak acids such as acetic acid, which converts the glucosamine unit (R–NH₂) of CS into its protonated form (R–NH₃⁺). Because of the presence of cationic moiety (–NH₃⁺) on its polymer backbone, CS dissolves in aqueous media and behaves as a polycation. Being inexpensive, biodegradable, nontoxic, and hydrophilic in nature, CS is used as an additive in foods, as a hydrating agent in cosmetics, and as a pharmaceutical agent in biomedicines [23]. CS dissolved in dilute aqueous solution of acetic acid reacts with aqueous glutaraldehyde to form CCH by Schiff base mechanism [24]. Rohindra et al. [25] studied the variation of equilibrium swelling ratio of CCH with exposure time, temperature, and pH of aqueous medium. CS has been employed as PEM in fuel cells [26–28]. There are reports in literature on the use of PVA and CS in various modified forms as PEMs for fuel cells. Recently, CCH and PCH have been reported as electrode binders in DBFCs [19,29]. It may be noted that chemical hydrogel binders can be used not only in DBFCs but also in other electrochemical energy devices [30].

In the present paper, we report our studies on morphological features as well as electrochemical performance of PCH and CCH vis-à-vis Nafion[®] as electrode binders in DBFCs. The DBFCs have been assembled with Misch-metal-based AB₅ alloy as anode, carbon-supported palladium (Pd/C) as cathode and Nafion[®]-117 membrane electrolyte (NME) as separator. All the DBFCs reported here have been studied using an aqueous alkaline solution of NaBH₄ as fuel and an aqueous acidified solution of H₂O₂ as oxidant in active mode using peristaltic pumps and at various cell temperatures.

2. Experimental details

2.1. Preparation of PCH, CCH, and Nafion[®] binders-based electrodes

To prepare anode catalyst inks, a pre-determined amount of La_{10.5}Ce_{4.3}Pt_{0.5}Nd_{1.4}Ni_{60.0}Co_{12.7}Mn_{5.9}Al_{4.7}, an AB₅ alloy powder (Ovonic Battery Company), was mixed thoroughly with 10 wt.% Vulcan XC 72 carbon powder in a glass vial. Water was added to

this mixture to create a slurry, which was agitated in an ultrasonic water bath (Bransonic[®] ultrasonic cleaner) for about 2 h. Then one of three different polymeric binders was added. The first binder, PCH, was added as an aqueous solution containing PVA (0.05 g mL⁻¹) and glutaraldehyde (25%) [19]. The second was a CCH binder solution containing CS (0.25 g mL⁻¹) dissolved in 1% (v/v) aqueous acetic acid solution. The third binder was a Nafion[®] binder solution (5 wt.% solution, Ion Power, Inc.). Binder solutions were added drop-wise to the alloy powder-carbon powder slurry under ultrasonic agitation, which was continued for another 2 h after the binder addition. The resulting anode composition was approximately 30 mg cm⁻² AB₅ alloy, and 5 wt.% PCH or 0.5 wt.% CCH or 5 wt.% Nafion[®]. The amount of AB₅ alloy, PCH, CCH, and Nafion[®] binder in anode composition were kept constant in all the MEAs studied.

The cathode catalyst ink was prepared following a similar procedure, in which required quantity of 10 wt.% carbon-supported palladium (Pd/C) (Aldrich) was mixed with appropriate volume of water in a glass vial and the suspension was ultrasonically agitated for ~ 2 h. Subsequently, required volume of a solution of PCH binder comprising an optimized aqueous solution mixture of PVA (0.05 g mL⁻¹) and glutaraldehyde (25%) or CCH binder solution comprising an aqueous solution of CS (0.25 g mL⁻¹) dissolved in 1% (v/v) aqueous acetic acid solution or Nafion[®] binder solution (5 wt.% solution, Ion Power, Inc.) was added drop wise to the suspension of Pd/C in water with ultrasonic agitation continued for another 2 h. The loadings of Pd metal and PCH, CCH, as well as Nafion[®] binders in cathode were about 1 mg cm⁻², 20 wt.%, 2 wt.%, and 20 wt.%, respectively. The loadings of Pd metal and PCH, CCH, as well as Nafion[®] binders in cathode were kept identical in all the MEAs studied.

The resulting anode or cathode inks were applied to pre-weighed carbon cloth substrates (Zorflex[®] Activated Carbon Cloth, FM 10, Chemviron Carbon/Calgon Carbon Corporation) and were dried under forced air-convection at room temperature. Finally, the PCH bound catalyst-on-cloth was dipped in 10 mL of 90% (v/v) aqueous solution of glacial acetic acid for 5 h. The dried CCH bound catalyst-on-cloth was dipped in 10 mL of 6.25% (v/v) solution of glutaraldehyde for 5 h to cause the cross-linking reaction between PVA or CS and glutaraldehyde to occur. After the treatment, the catalyst-coated carbon cloth was washed thoroughly with DI water to remove excess impurities.

2.2. Scanning electron microscopy studies of electrodes

Scanning electron micrographs of PCH, CCH, and Nafion[®] binder-based anodes as well as cathodes were recorded using a JEOL JSM-IC 848 scanning electron microscope (SEM). SEM studies on anode and cathode samples were carried out at magnifications of 500 and 2500 times. A lower magnification was sufficient to observe morphological features of anode samples because of the relatively large particle size of AB₅ anode catalyst. A higher magnification was necessary to observe morphological features of cathode samples because of the relatively small particle size of Pd/C cathode catalyst.

2.3. Electrochemical characterization of DBFCs

For the electrochemical characterization of PCH, CCH, and Nafion[®] binder-based DBFCs, membrane electrode assemblies (MEAs) were fabricated by sandwiching the anode and cathode on either side of a pre-treated Nafion[®] membrane. MEAs comprising PCH, CCH, as well as Nafion[®] binder-based electrodes and Nafion[®]-117 membrane electrolyte (NME) were employed to assemble various liquid-fed DBFCs. The fuel cell hardware employed in this study was procured from Fuel Cell Technologies, Inc. The active

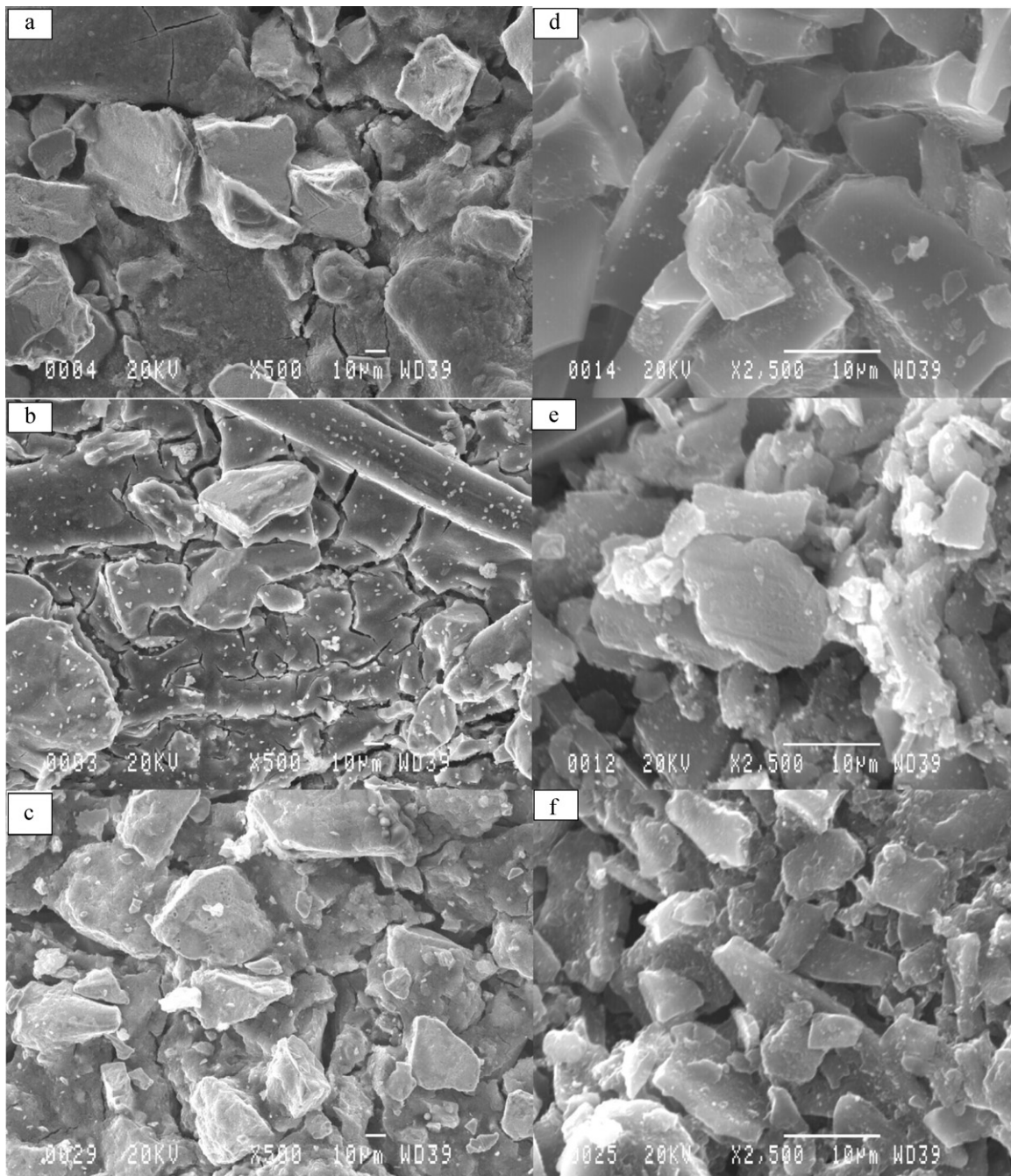


Fig. 1. Scanning electron micrographs of (a) PCH binder-based anode, (b) CCH binder-based anode, (c) Nafion® binder-based anode, (d) PCH binder-based cathode, (e) CCH binder-based cathode, and (f) Nafion® binder-based cathode.

area of each of anode and cathode was 5 cm^2 . The MEAs were placed between anode and cathode flow-field graphite plates and tightened together by applying a torque of 12.2 N m . All the DBFC results reported in this paper were recorded in active mode using peristaltic pumps for both fuel and oxidant solutions. The fuel comprised an aqueous solution of 1.7 M NaBH_4 in 7.0 M NaOH and the oxidant comprised an aqueous solution of $2.5\text{ M H}_2\text{O}_2$ in $1.5\text{ M H}_2\text{SO}_4$. The flow rates for fuel and oxidant solutions were maintained constant at 5 and 10 mL min^{-1} for all the electrochemical studies. After installing the DBFCs in the test station, performance evaluation studies were initiated. Galvanostatic-polarization data for various DBFCs were recorded using a computer-controlled fuel cell test system (Scribner Associates, Inc., Model: 890e).

3. Results and discussion

The PVA polymeric backbone contains polar hydroxyl group ($-\text{OH}$) on alternate carbon atoms. Since $-\text{OH}$ is a weakly ionizing group, its hydrophilicity and water-retention capability is limited. CS contains three different polar functional groups, namely, $-\text{OH}$, primary amine ($-\text{NH}_2$), and ether ($\text{C}-\text{O}-\text{C}$) groups, and possesses six-member ring structures along its polymer backbone. Because of the presence of multiple polar functional groups and six-member ring structures in the backbone, CS forms hydrogen bonds with water and traps water in its ring structures. In other words, CS is a strongly hydrophilic polymer. The backbone of Nafion® contains $\text{C}-\text{F}$ bonds that are polar and hydrophobic [31]. However, Nafion® possesses weakly polar $\text{C}-\text{O}-\text{C}$ linkages and highly dissociable as

well as hydrophilic sulfonic acid ($-\text{SO}_3\text{H}$) group in its structure. Because of the presence of the contrasting properties, Nafion[®] is understood to possess hydrophilic and hydrophobic zones separated by an intermediate region [32]. Since the water-attracting behavior of Nafion[®] is restricted to its hydrophilic region only, its water retaining capability and hence hydrophilic nature is not as high as that of CS. CS possesses higher hydrophilic characteristics as compared to both PVA and Nafion[®] because of its molecular structure and chemistry.

In PCH, polar functional groups present are mainly $-\text{C}-\text{O}-\text{C}-\text{O}-$, $-\text{C}-\text{O}-\text{C}$ and possibly some unreacted $-\text{OH}$. The $-\text{C}-\text{O}-\text{C}-\text{O}-$ and $-\text{C}-\text{O}-\text{C}$ groups in PCH form ring structures [33]. In PCH, water is bonded not only to electronegative oxygen atoms by hydrogen bonding but is also trapped inside $-\text{C}-\text{O}-\text{C}-\text{O}-$ and $-\text{C}-\text{O}-\text{C}$ ring groups. Due to the cross-linking reaction between CS and formaldehyde, an extra CS chain links with the first CS chain. Because of the increased CS chain bonding, the hydrophilicity of CCH increases [22,34]. In turn, the increased hydrophilicity of CCH results in greater water retention, increased ionic conductivity and higher mobility of fuel/oxidant within CCH binder-based electrodes. This leads to higher electrochemical performance in DBFCs with CCH binder-based electrodes at elevated temperatures.

Fig. 1 shows scanning electron micrographs of the (a) PCH binder-based anode, (b) CCH binder-based anode, (c) Nafion[®] binder-based anode, (d) PCH binder-based cathode, (e) CCH binder-based cathode, and (f) Nafion[®] binder-cathode. All the anode and cathode materials employed in this morphological study are identical in all respects with those employed in DBFCs reported here. Since these electrodes exhibited high electrochemical performance in DBFCs, the proportions of catalyst particles and polymer-based binders in these electrode samples are reasonably optimum. In other words, the contents of polymer-based binders in the electrode matrixes are high enough not only to keep the electrode materials intact and bound to the electrode substrate but also to facilitate efficient flux of fuel, oxidant as well as ions to the catalyst surface. Also, the contents of polymer-based binders in the electrode matrixes are low enough to allow efficient flux of electrons among the catalyst particles in the electrode matrix as well as from catalyst particles to the electrode substrate and vice versa. In Fig. 1, the polymer-based binders appear as fluffy material whereas the catalyst material appears as solid particles. Fig. 1 also shows that AB_5 anode catalyst particles and Pd/C cathode catalyst particles are coated with polymer-based binders. The degree of coating of catalyst particles by polymer-based binders appears to be maximum in case of CCH followed by PCH and Nafion[®] binders. This observation is in agreement with the fact that the loadings of CCH binder in both optimized anode and cathode are ten times lower than those of PCH as well as Nafion[®] binders.

The ambient temperature ionic conductivity of PCH [15], CCH [35], and Nafion[®] [35] electrode binders are 2.5×10^{-2} , 1×10^{-3} , $2.6 \times 10^{-2} \text{ S cm}^{-1}$, respectively. The MEAs employed in this study were fabricated by hand pressing the pre-treated Nafion[®]-117 membrane between anode and cathode. The same technique of MEA fabrication was adopted by Liu et al. [36], who opined that it is useful to leave some space between the anode and the membrane so that the fuel being in liquid state would be able to reach the anode easily and also the release of hydrogen evolved by decomposition/hydrolysis of BH_4^- fuel would be facilitated. Liu et al. [37] observed that in a DBFC performance stability test, the cell using an anode tightly pressed onto the membrane exhibited a quick decrease of cell voltage as compared to the cell wherein the anode was loosely pressed onto the membrane. Kim et al. [38] remarked that since the fuel and Na^+ ions can easily transfer through the liquid medium, a close contact between the anode and the membrane is not necessary. To further facilitate the removal of hydrogen bubbles trapped in the gap between the membrane and the anode,

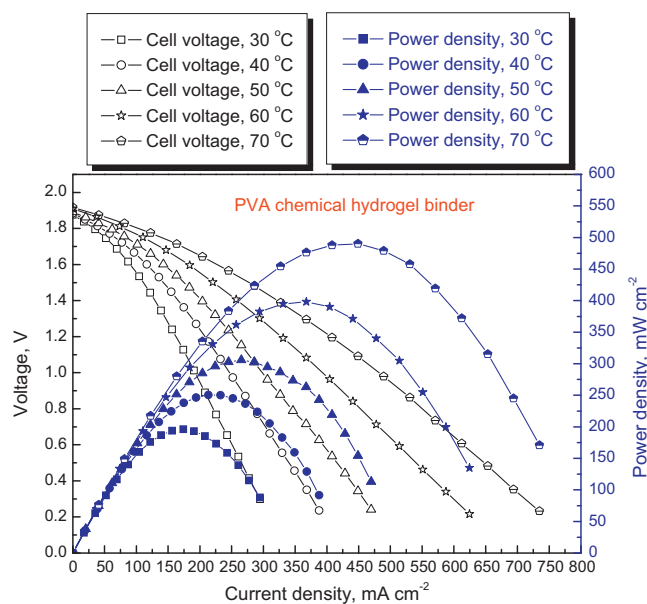


Fig. 2. Plots for cell voltage and power density versus current density for DBFCs with PVA chemical hydrogel binder-based electrodes and NME as electrolyte-cum-separator at different operating cell temperatures.

they adopted a corrugated anode design in which there was a gap of 2 mm between the anode and the membrane. This anode design increased the DBFC performance by 27% as compared to a flat anode. The same considerations apply to cathode side of the DBFCs studied in this work. This is because the oxidant, hydrogen peroxide, is in liquid state and it also decomposes at catalyst surface liberating gaseous oxygen. The electrochemical performance data for DBFCs employing PCH, CCH, and Nafion[®] binders-based electrodes and NME as electrolyte-cum-separator at different operating cell temperatures are shown in Figs. 2, 3, and 4, respectively. The electrochemical performance data related to open circuit voltage, peak power density, and current density corresponding to peak power

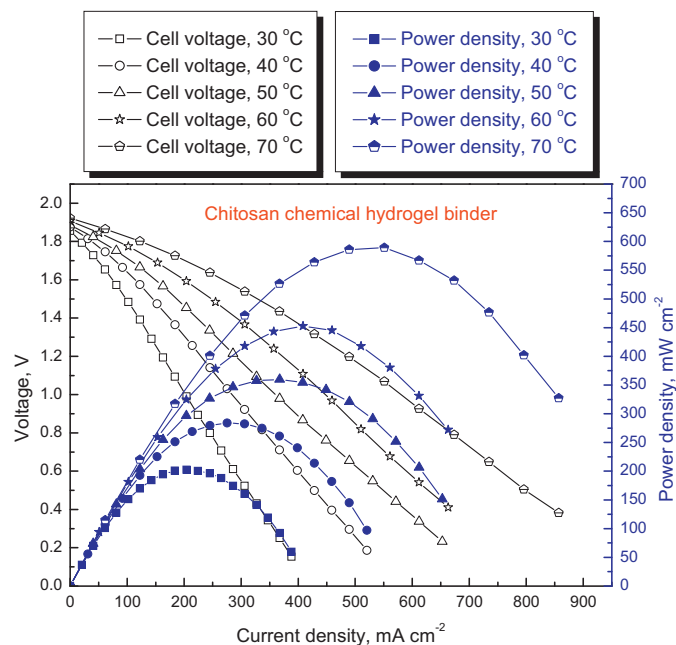


Fig. 3. Graphs for cell voltage and power density versus current density for DBFCs with chitosan chemical hydrogel binder-based electrodes and NME as electrolyte-cum-separator at different operating cell temperatures.

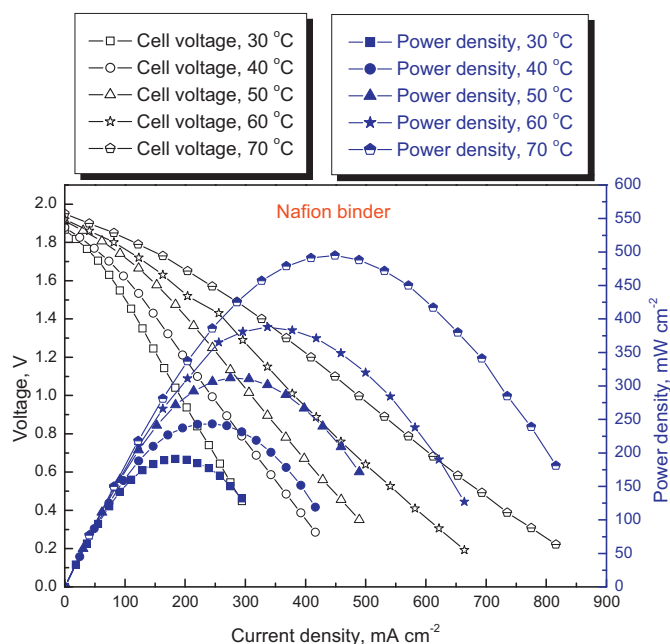


Fig. 4. Plots for cell voltage and power density versus current density for DBFCs with Nafion[®] binder-based electrodes and NME as electrolyte-cum-separator at different operating cell temperatures.

for DBFCs employing PCH, CCH, and Nafion[®] binder-based electrodes and NME as electrolyte-cum-separator at different operating cell temperatures are shown in Fig. 5(a), (b), and (c), respectively. It is evident from Fig. 5(a) that OCV values of DBFCs with all the three electrode binders are about 1.9 V. Moreover, the OCV values of DBFCs with all the three electrode binders increase with increase in cell temperature. The increase of OCV values with increase in cell temperature could be due to the improvement in electrode kinetics of DBFCs. It is evident from Fig. 5(b) that as the cell temperature is increased from 30 to 70 °C, the peak power density increased from 196 to 490 mW cm⁻² for the DBFC employing PCH as electrode binder; from 202 to 589 mW cm⁻² for the CCH binder; and from 191 to 494 mW cm⁻² for the Nafion[®] electrode binder. Fig. 5(c) shows that as the cell temperature is increased from 30

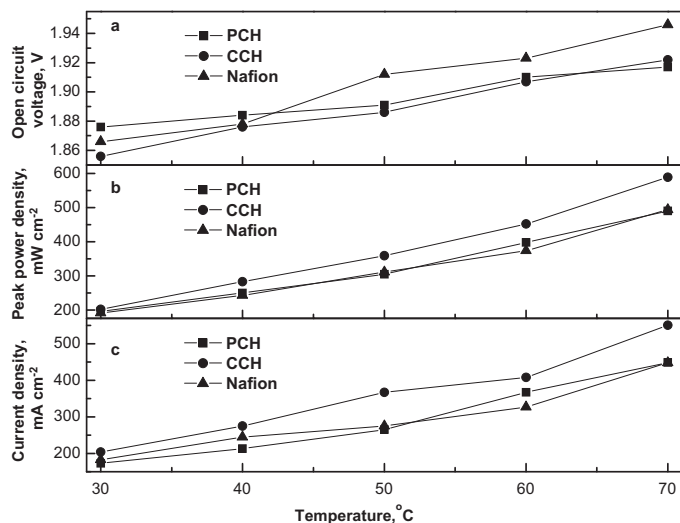


Fig. 5. Graphs of (a) open circuit voltage, (b) peak power density, and (c) current density corresponding to peak power for DBFCs employing PCH, CCH, and Nafion[®] binder-based electrodes and NME as electrolyte-cum-separator at different operating cell temperatures.

to 70 °C, the peak power current density increases from 173 to 449 mA cm⁻² for PCH electrode binder, from 204 to 551 mA cm⁻² for the CCH electrode binder and from 183 to 448 mA cm⁻² for the Nafion[®] electrode binder. From Fig. 5(b) as well as (c), it is evident that the peak power density and current density corresponding to peak power of DBFCs with all the three electrode binders increase with increase in cell temperature. The increase of peak power density and current density corresponding to peak power of DBFCs could be due to the increase in both electrode reaction rates as well as membrane ionic conductivity in the DBFCs. It is noteworthy from Fig. 5(b) as well as (c) that the electrochemical performance data for DBFCs employing PCH and Nafion[®] electrode binders are comparable at all temperatures. In contrast, electrochemical performance data for DBFC employing CCH as electrode binder are better than for DBFCs employing PCH as well as Nafion[®] electrode binders at all temperatures. It is interesting to note that the improvement in electrochemical performance for DBFC employing CCH as the electrode binder becomes more prominent with an increase in cell temperature. The common feature between PCH and Nafion[®] electrode binders is that both are synthetic polymer-based and relatively high hydrophobic in nature. In contrast, the CCH electrode binder is made of a natural polymer that is comparatively high hydrophilic in nature [39–43]. Because of its hydrophilicity, CS is used as a hydrating agent in cosmetics [23]. The greater hydrophilicity suggests an increased ability to retain water. Osifo et al. [44] have observed that chitosan hydrogel membranes take up more water than Nafion[®] membranes in the temperature range of 20–60 °C. Mukoma et al. [45] have observed that chitosan hydrogel membranes absorbed about 60% water compared to about 30% for Nafion[®]-117 membrane. The authors ascribed this observation to enhanced hydrophilic nature of chitosan hydrogel membrane as compared to Nafion[®]-117 membrane. The increased water retention capability of CCH implies that ionic conductivity and mobility of fuel and oxidant within CCH binder-based electrode matrix will be higher than those of the PCH as well as Nafion[®] binders-based electrode matrixes at elevated temperatures. The higher ionic conductivity and mobility of fuel and oxidant within the CCH binder-based electrode matrix leads to an improvement in electrochemical performance of the CCH electrode binder-based DBFC relative to DBFCs employing PCH and Nafion[®] binders-based electrodes at elevated temperatures. This is evident from Fig. 5(b) and (c) that show an improvement in power performance of CCH electrode binder-based DBFC as compared to PCH and Nafion[®] electrode binders-based DBFCs with gradual increase in cell temperature. This result is significant in view of the fact that Nafion[®]-based electrode binders and PEMs suffer from dehydration and hence, loss in fuel cell performance at sufficiently high operating temperatures. Different approaches are adopted to enhance hydrophilic nature of Nafion[®] membrane for use in high operating-temperature PEFCs [46]. Being a highly hydrophilic material [23,39–43], CCH may prove to be a cost-effective and high performance alternative electrode binder or PEM in high operating-temperature PEFCs.

As a result of their hydrophilic nature, polymer hydrogel binders are expected to confer advantages over pristine polymer binders like Nafion[®]. Of particular significance is the potential of the polymer hydrogel binders to establish and maintain three-point contact among reactant (ion/fuel/oxidant), electro-catalyst and PEM. The incumbent binder material, Nafion[®] consists of a combination of hydrophobic polymer base, hydrophilic ionic clusters and an intermediate region [32] that allows effective ion transfer to the catalyst surface when used as electrode binder. PTFE is a hydrophobic electrode binder that is useful in mitigating flooding of cathode while allowing effective oxygen transfer to the cathode catalyst surface. However, PTFE restricts transfer of ions to the catalyst surface due to its hydrophobic nature. The large volume of water

absorbed in the polymer matrix of a polymer hydrogel helps in attaining high mobility of ions, fuel and oxidant within the hydrogel-bonded electrode matrix. In contrast, water absorption and retention capabilities of pristine polymers such as Nafion[®] and PTFE are comparatively small, thereby limiting the transfer efficiency of ion, fuel, and oxidant to the electro-catalyst surface. The loading of a polymer-based binder in the electrode of a fuel cell plays an important role in delivering high electrochemical performance. The effect of Nafion[®] binder content in the anodes of air-breathing DBFCs on their power performance has been studied by Kim et al. [47]. Optimum loadings of PCH binder in anode and cathode of DBFCs was found to be about 5 and 20 wt.%, respectively. A lower loading of PCH binder in the anode was sufficient because the anode comprised mostly of AB₅ metallic powder that has low surface area and only 10 wt.% of Vulcan XC 72 carbon powder that has high surface area. A higher content of PCH binder in the cathode was needed because the cathode comprised of only 10 wt.% Pd metal that has low surface area and 90 wt.% Vulcan XC 72 carbon powder that has high surface area. In other words, the cathode material was fluffier than the anode material and hence needed more content of PCH binder for optimum performance in the DBFCs. It may be noted that for the same electrode materials, the content of PCH binder needed was about ten times higher than that of CCH binder [29]. This difference could be due to the difference in the structural as well as functional characteristics of PVA that is a synthetic polymer and CS that is a natural polymer.

When Nafion[®] or PTFE is employed as an electrode binder, the MEA is generally prepared by hot-compaction technique in which the mixture of electrode material and polymer binder is heated to a temperature that is in the range of glass transition temperature of the binding polymer. At the glass transition temperature, the polymer melts/softens and while solidifying during cooling under pressure, it encompasses the electrode material with the electrode substrate and PEM. Unlike Nafion[®] or PTFE that acts as a binder due to a physical phenomenon such as heating/cooling, the binding actions of PCH and CCH are due to cross-linking induced by reagents such as glutaraldehyde in the presence of a protic acid catalyst under ambient conditions of temperature and pressure. Binding actions of PCH and CCH for the electrode mass are thus accompanied with breaking of some existing covalent bonds and formation of some new covalent bonds.

Polymer-based electrode binders such as Nafion[®] are expensive whereas polymer hydrogel-based electrode binders are not and can be prepared in-house using simple processing methods. Catalyst inks with polymeric binders such as Nafion[®] are generally prepared in organic solvents, such as 2-propanol, because of the high hydrophobic nature of long carbon chain of polymers. The use of organic solvents not only adds to cost but also causes health and environmental hazards. Catalyst inks with hydrogel binders such as PCH and CCH are generally prepared in water, thereby enhancing cost-effectiveness and workplace safety.

4. Conclusions

In this study, it has been observed that a CCH binder performed better than a Nafion[®] binder whereas a PCH binder performed as good as a Nafion[®] binder in DBFCs at elevated cell temperatures. The better electrochemical performance of the CCH binder as compared to that of PCH as well as Nafion[®] binders in DBFCs at elevated cell temperatures is due to the better water retention capability of CCH. Because of the hydrophilic nature and high water retention capability, chitosan-based electrode binder as well as membrane electrolyte are suitable candidates for application in high operating temperature fuel cells. Peak power densities of 196, 202, and 191 mW cm⁻² at corresponding current density values of 173, 204,

and 183 mA cm⁻² have been observed for DBFCs with PCH, CCH, and Nafion[®]-based binders respectively at 30 °C. Peak power densities of 490, 589, and 494 mW cm⁻² at corresponding current density values of 449, 551, and 448 mA cm⁻² have been observed for DBFCs employing PCH, CCH, and Nafion[®]-based binders respectively at 70 °C.

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