

Research Article

Enhanced Performance of Membraneless Sodium Percarbonate Fuel Cells

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This paper presents the continuous flow operation of membraneless sodium percarbonate fuel cell (MLSPCFC) using acid/alkaline bipolar electrolyte. In the acid/alkaline bipolar electrolyte, percarbonate works both as an oxidant as well as reductant. Sodium percarbonate affords hydrogen peroxide in aqueous medium. The cell converts the energy released by H_2O_2 decomposition with H^+ and OH^- ions into electricity and produces water and oxygen. At room temperature, the laminar flow based microfluidic membraneless fuel cell can reach a maximum power density of 28 mW/cm^2 with the molar ratio of $[\text{Percarbonate}]/[\text{NaOH}] = 1$ as fuel and $[\text{Percarbonate}]/[H_2SO_4] = 2$ as oxidant. The paper reports for the first time the use of sodium percarbonate as the oxidant and reductant. The developed fuel cell emits no CO_2 and features no proton exchange membrane, inexpensive catalysts, and simple planar structure, which enables high design flexibility and easy integration of the microscale fuel cell into actual microfluidic systems and portable power applications.

1. Introduction

The advancement of portable power electronics and the continual integration of functionality into a single all encompassing device has created an increased demand on energy supply. These portable applications include not only common appliances such as cell phones, laptop computers, and personal organizers, but also more specialized devices such as clinical and diagnostic tests, microanalytical systems for field tests, and global positioning systems [1]. It is a more and more accepted fact that fuel cells are one of the most fascinating and interesting aspects of today's technology. The interest in fuel cells has been intensified due to several requirements, such as environmental safety, simple fuel storage, good cost-efficiency, and, of course, sufficient electrical performance. No fuel cells have satisfied all these requirements, although various types of fuel cells have been competitively developed. Therefore, it is a pressing requirement and very important to search for another fuel that can overcome these inherent problems [2]. To date, microscale systems research has

focused mostly on miniaturization of functional components, leaving miniaturization of long lifetime power sources as one of the major challenges.

A microfabrication method—fabrication inside capillaries using multistream laminar flow—provided the idea for a novel type of fuel cell, which eliminates several of the technical issues related to the use of polymer electrolyte membrane fuel cells (PEMs), such as fuel crossover [3], membrane degradation, a long startup time, ohmic losses, size, fabrication and water management limited durability of catalysts [4]. A membrane, as used in membrane-electrode assemblies (MEAs), brings about a resistance to the transport of ions through the electrolytes. Furthermore, MEAs are expensive components at present. Therefore, the cell structure without the membrane has two merits, decrease of electrical resistivity in the cell and inexpensive material cost.

In this paper, the specific power source of a fuel cell is studied that utilizes a unique property of fluid flow at the microscale, multistream laminar flow (Figure 1).

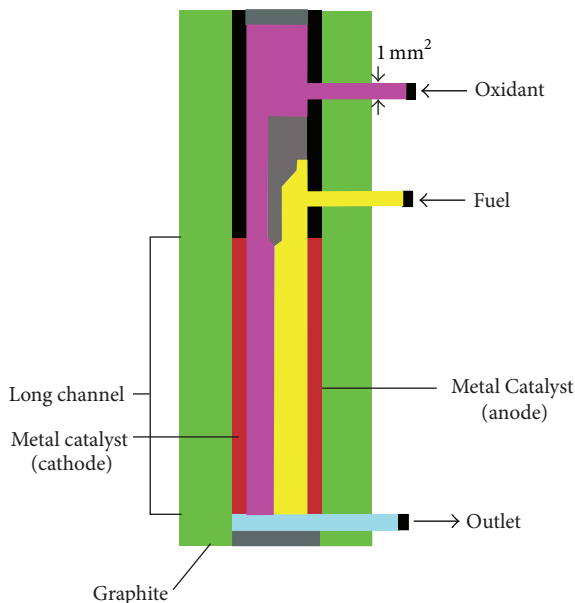


FIGURE 1: The side view of the E-type planar membraneless micro-fuel cell.

The dimensions and operating conditions of the microfluidic fuel cells discussed here are such that fluid flow is pressure driven and characterized by a Reynolds number (Re) less than 10. Two different aqueous streams, one containing fuel and the other with oxidant, are introduced into the same channel at $Re \leq 2100$ [5–7]. The aqueous streams will proceed to flow laminarily in parallel over the anode and cathode electrodes placed on opposing side walls within the microfluidic channel.

Since the diffusion process at the liquid/liquid interface determines the performance of the membraneless fuel cell, the two key parameters are the Reynolds number $Re = UH/\nu$ and the Peclet number $Pe = UH/D_{H^+}$ where U is the mean flow velocity in the reaction channel, and H is the channel height; ν is the kinematic viscosity of the fuel/oxidant liquids. For simplification, we assume for an operation temperature of 25°C a kinematic viscosity of water $\nu = 0.871 \times 10^{-6} \text{ m}^2 \text{ s}^{-1}$ for both fuel and oxidant liquids and a diffusion coefficient of proton $D_{H^+} = 9.31 \times 10^{-9} \text{ m}^2 \text{ s}^{-1}$. With the known volumetric flow rate and channel cross-section of 1 mm^2 , the Reynolds numbers and the Peclet numbers can be estimated with the previous properties.

Fuel and oxidant react at the electrodes while the two liquid streams and their common liquid-liquid interface provide the required ionic conductance to complete the fuel cell chemistries. All the aforementioned membrane-related issues can be avoided in the MLSPCFCs studied here. The flexibility and the performance implications of operating MLSPCFCs under “dual-media,” that is, one electrode is acidic and the other one is alkaline condition, will be the focus of this study.

In this communication, first time we introduce sodium percarbonate ($2\text{Na}_2\text{CO}_3 \cdot 3\text{H}_2\text{O}_2$) and demonstrate the performance of a membraneless sodium percarbonate fuel cell

(MLSPCFC). Sodium percarbonate is a true peroxy salt and is a convenient source of hydrogen peroxide [8, 9]



The sodium percarbonate fuel cell is unique from previous fuel cells using H_2O_2 , and the MLSPCFC uses percarbonate not only as an oxidant but also as a reductant. As for the use of sodium percarbonate, the direct methanol hydrogen peroxide fuel cell [10] and the aluminum-hydrogen peroxide semifuel cell [11] have been reported. In these reports, H_2O_2 was used only as the oxidant, and methanol or aluminum was used as the reductant. In contrast, our MLSPCFC uses sodium percarbonate, both as the oxidant and as the reductant.

Many experimental studies had been performed in developing membraneless microfuel cells. Recently, Da Mota et al. reported a different membraneless, microfuel cell concept at room-temperature that also lacks a PEM [12]. A variety of fuels were used in these investigations such as formic acid, methanol, and hydrogen solutions. However, since they all used oxygen solution as the oxidant, the performance of these microfuel cells was found to be restricted severely by the low transport efficiency of oxygen in the cathode stream. An experimental study using hydrogen peroxide has been proposed for a Y-shaped microchannel fuel cell [13].

Compared to hydrogen, methanol, and formic acid, hydrogen peroxide has a higher overall theoretical open circuit potential and maximum efficiency as indicated in Table 1. Furthermore, the hydrogen peroxide is used not only as a reductant but also as an oxidant. The decomposition of hydrogen peroxide in anode and cathode streams can generate electric power which is comparable to a typical air-breathing DMFC at room temperature and produce no carbon dioxide emission and thus will be explored as the fuel with the membraneless fuel cell in this paper.

The MLSPCFC has some advantages, such as sodium percarbonate being a cheap, nontoxic, large scale industrial chemical used primarily in detergents and as a mild oxidant. The cell is more environmentally friendly than the DMFC because there is no CO_2 emission and the sodium percarbonate fuel can be handled more simply than hydrogen, as it is well-known fact that sodium percarbonate solution is a widespread safe disinfectant. On the performance side, the MLSPCFC generates electric power comparable to a typical air-breathing DMFC when operating in a microchemical channel at room temperature. In addition, the MLSPCFC requires no membrane-electrode assemblies (MEAs). Thus, the cost for the materials is low and the structure of the cell is simple. In this study, a new branch of simplified architectures that is unique from those that have been reported in the literature has been developed by eliminating and integrating the key components of a conventional MEA.

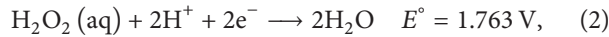
2. Results and Discussion

2.1. Performance of the Membraneless Sodium Percarbonate Fuel Cell (MLSPCFC). The performance of MLSPCFC using a fuel stream of 0.75 M percarbonate + 0.75 M NaOH and an oxidant stream of 0.75 M percarbonate + 0.375 M H_2SO_4 was investigated. In aqueous medium percarbonate affords H_2O_2 .

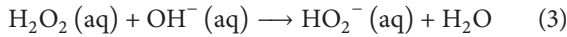
TABLE 1: Summary of thermodynamic data for different fuels.

Fuel	Reaction	n (e)	$-\Delta H^\circ$ (kJ/mol)	$-\Delta G^\circ$ (kJ/mol)	E_{rev}° (V)	Max efficiency (%)
Hydrogen	$H_2 + 1/2O_2 \rightarrow H_2O$ (l)	2	286	237.3	1.23	82.97
Methanol	$CH_3OH + 1/2O_2 \rightarrow CO_2 + 2H_2O$ (l)	6	726.6	702.5	1.21	96.68
Formic acid	$HCOOH + 1/2O_2 \rightarrow CO_2 + H_2O$ (l)	2	270.3	285.5	1.48	105.6
Hydrogen peroxide	$H_2O_2 + HO_2^- + 2H^+ + OH^- \rightarrow O_2 + 3H_2O$ (l)	2	138	176	1.83	127.54

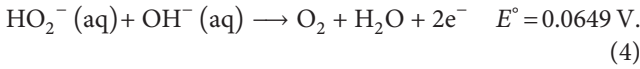
H_2O_2 is usually known as an oxidant. In an acid electrolyte, the electron-gaining reaction of the oxidant proceeds as follows [14]:



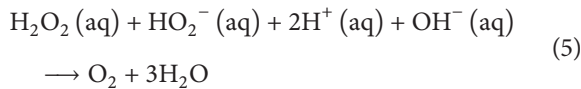
where (aq) means the molecule is dissolved in water and E° is the standard reduction potential. On the other hand, H_2O_2 can act as a reductant, depending on reaction conditions such as pH and content of strong oxidants. In an alkaline electrolyte, the electron-losing reaction of H_2O_2 proceeds in two steps, the first being



which is the acid-base reaction between the weak acid, H_2O_2 , and the alkaline electrolyte. The second is



Here, we use the acid and alkaline electrolytes contacting one another (acid/alkaline bipolar electrolyte). This is the principal novel point for the MLSPCFC, because reactions (2) and (4) can proceed to generate electricity in the respective electrolytes. Then, the overall electrochemical reaction of the MLSPCFC is



and involves the disproportionation reaction of H_2O_2 together with the combination of H^+ and OH^- ions. The calculated electromotive force (emf) of (5) is 1.828 V (=1.763 + 0.0649 V). The inherent value of the electromotive force of the MLSPCFC is higher than that of the HFC (1.23 V) and the PEMFC or DMFC (1.21 V). However, the open circuit potential (OCP) of the MLSPCFC becomes much lower than the EMF. Thus, the OCP does not significantly contribute to the observed phenomena.

The acid/alkaline bipolar electrolyte illustrated in Figure 2 consists of sulfuric acid, as well as sodium hydroxide, solutions. For the reaction of (5) to proceed continuously, SO_4^{2-} ion neutralizes Na^+ ion electrically at the acid/alkaline bipolar electrolyte interface. Consequently, the MLSPCFC produces water and oxygen but no CO_2 . In this regard, the MLSPCFC is more environmental friendly than the DMFC.

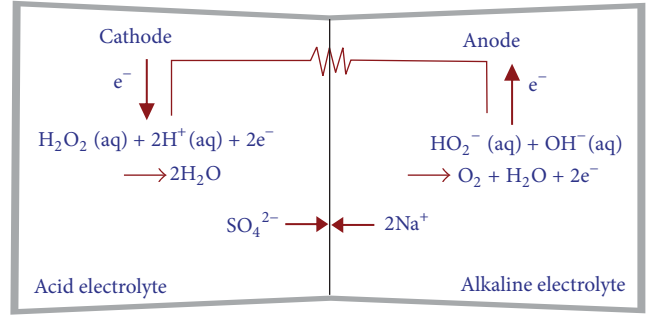


FIGURE 2: Operating principle of membraneless sodium percarbonate fuel cell (MLSPCFC) using acid/alkaline bipolar electrolyte.

The measurements were carried out at room temperature with external resistances and PC controlled digital multimeters, keeping the molar ratio of [Percarbonate]/[H_2SO_4] = 2 as oxidant and [Percarbonate]/[NaOH] = 1 as fuel. The percarbonate concentration was varied from 0.075 to 0.75 mol/L, a range which is lower than the concentration of normal percarbonate disinfectant. These acid and alkaline solutions with the same percarbonate concentration flowed over the cathode and the anode, respectively. The open circuit voltage, V_{oc} , increases slightly with increasing percarbonate concentration. This is attributed to the activity of the reactant as expressed in the Nernst equation [2].

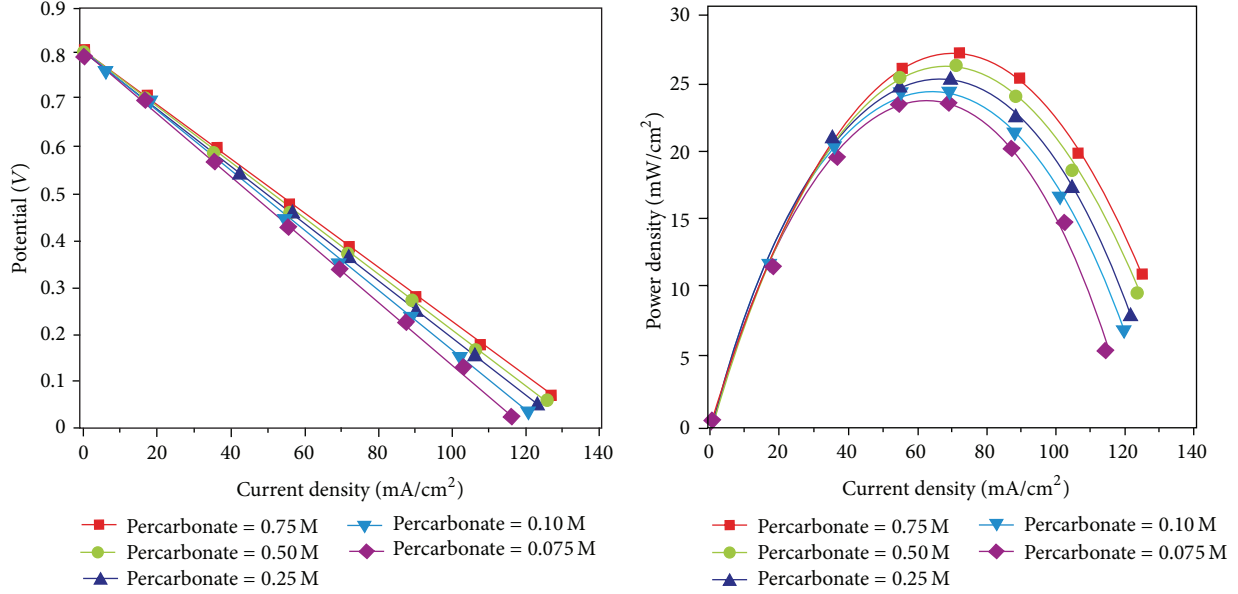
The polarization curves recorded from these membraneless fuel cells have the same characteristic shape of typical fuel cells with the kinetically limited, ohmic, and mass transport limited regions (Figures 3 and 4). Using the corresponding power density curves optimal operation parameters can be deduced: 28.09 mW/cm² at the potential of 0.46 V. In this experiment, the maximum cell potential obtained is 0.799 V and the maximum current density is 127.73 mA/cm² (Table 2). Accordingly, the cell performance will be enhanced intensely if the concentration of oxidant in cathodic stream can be increased to raise the exchange current density at the cathode as high as possible.

As shown in Figures 3(a) and 3(b), five typical concentrations of [Percarbonate]/[NaOH], 0.75 M, 0.5 M, 0.25 M, 0.1 M, and 0.075 M, are considered. It is found that if the fuel concentration reduces to 0.25 M or even smaller 0.075 M, the performance still does not drop significantly. These results are consistent with the experimental findings [7]. While if we can raise the concentration of oxidant in cathodic stream, the cell performance appears to be improved significantly

TABLE 2: Efficiency of the membraneless sodium percarbonate fuel cell (MLSPCFC).

Fuel cathode	Fuel anode	Catalyst	Potential (V)	Current density (mA/cm ²)	Electrochemical efficiency %
[Percarbonate]/[H ₂ SO ₄]	[Percarbonate]/[NaOH]	Pt-black	0.799	127.73	43.71

The molar ratio of [Percarbonate]/[NaOH] = 1 as fuel and [Percarbonate]/[H₂SO₄] = 2 as oxidant. The electrochemical efficiencies were calculated by dividing the measured potential by the maximum achievable potential.



(a) The lines indicate that the current density increases by increasing concentration of oxidant from 0.075 M to 0.75 M

(b) The curved lines indicate that the maximum power density is almost same for the variation of fuel concentration from 0.75 M to 0.075 M

FIGURE 3: Variations of cell performance with five assigned fuel concentrations of sodium percarbonate in sodium hydroxide solution: (a) the polarisation curves; (b) the corresponding power density curves.

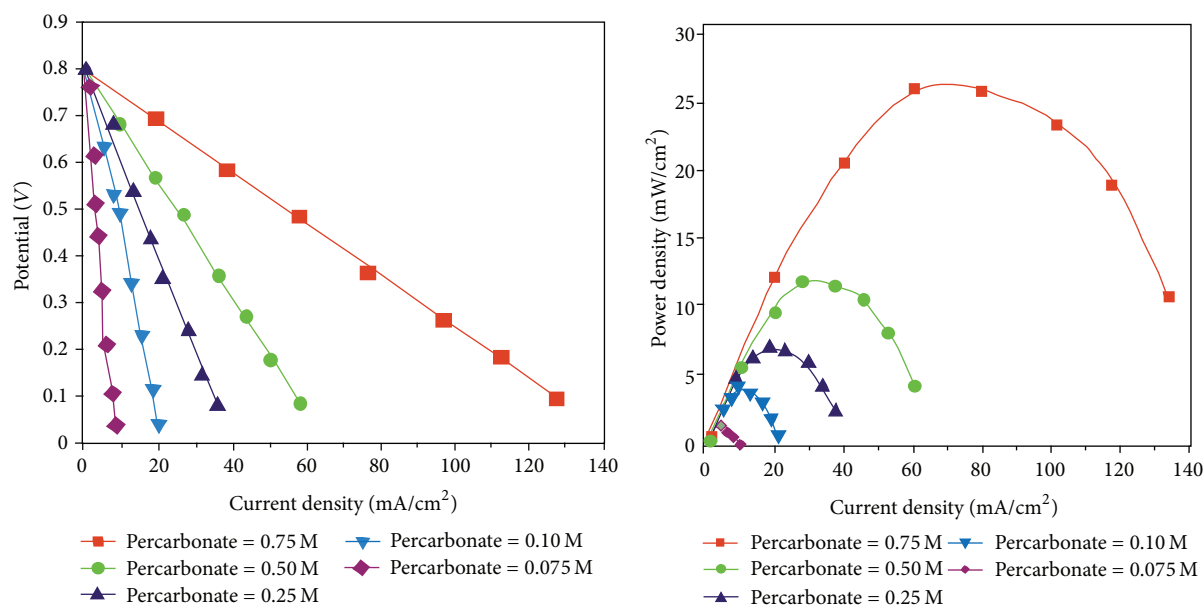
as shown in Figures 4(a) and 4(b), the five assigned values of oxidant concentration 0.75 M, 0.5 M, 0.25 M, 0.1 M, and 0.075 M are investigated. One can see that if the concentration of oxidant is 10 times larger, the current density is also increased approximately ten times. Thus, the present analyses confirm the experimental results that this membraneless microfuel cell is cathodic limited and suggest that it is a crucial factor in improving cell performance to increase the concentration of oxidant in the cathodic stream.

2.2. Thermodynamic Evaluation of MLSPCFC. In this experiment, we examined the thermodynamic properties for the overall reaction of the MLSPCFC by using chemical thermodynamic data [15]. We additionally used the following data: the enthalpy change $\Delta H = -55.9$ kJ/mol and the entropy change $\Delta S = 80.5$ J/Kmol for the reaction of $H^+ + OH^- \rightarrow H_2O$, the $\Delta H = -30.8$ kJ/mol and the $\Delta S = 120$ J/Kmol for $H^+ + HO_2^-(aq) \rightarrow H_2O_2(aq)$. The Gibbs free energy change $\Delta G (= \Delta H - T\Delta S)$, the ΔH , and the ΔS of (5) are estimated as -176, -138, and 128 J/Kmol, respectively. Note that the ΔS is positive value. As a result, the energy term of $T\Delta S$ ($=38.1$ kJ/mol at 298 K) additionally contributes to the ΔG . If all of the ΔG were converted into electrical energy, the maximum efficiency, $\Delta G/\Delta H$, would be 128%. This means that

the MLSPCFC has the potential to convert the $T\Delta S$ as heat. This energy conversion may appear in the MLSPCFC, when we reduce some irreversible energy losses, such as activation losses of electrochemical reactions, resistance to the transport of electrons through the electrodes and interconnections, resistance to the transport of ions through the electrolytes, concentration losses of reactants near the electrode surfaces, and the loss due to the intermediate formation [16].

Using the ΔG value, we estimated the energy density of percarbonate fuel. When we make an acid and an alkaline solution of sodium percarbonate with H_2SO_4 and NaOH (the molar ratio of [Percarbonate]/[H₂SO₄] = 2 and [Percarbonate]/[NaOH] = 1), the energy density is 436 Wh/L estimated at 200 kgf/cm² and 25°C neglecting container volume. Note that our percarbonate fuel was an aqueous solution. Therefore, we can handle the fuel more simply than gaseous hydrogen.

In addition to the experiment with a single cell, we have been carrying out series and parallel integrations of the cell on a microchemical chip. This simple structure can allow an easier integration process than a micro-DMFC [17, 18] and a micro-HFC [19]. Because the integration allows the MLSPCFC to increase the power, it could be utilized for various portable power applications.



(a) The lines indicate that the current density increases by increasing the concentration of oxidant from 0.075 M to 0.75 M

(b) The curved lines indicate that the power density increases by increasing the concentration of oxidant from 0.075 M to 0.75 M

FIGURE 4: Variations of cell performance with five assigned oxidant concentrations of sodium percarbonate in sulphuric acid solution (a) the polarisation curves. (b) The corresponding power density curves.

3. Conclusions

A microscale membraneless sodium percarbonate fuel cell (MLSPCFC) was fabricated and its operating behaviour characterized for the first time. Standard microfabrication techniques were used to create the device. The novel micro-fuel cell utilized a membraneless dual-electrolyte design with sodium percarbonate as both reductant and oxidant. At room temperature, the laminar flow-based microfluidic fuel cell produced a maximum power density of $28.09 \text{ mW}/\text{cm}^2$. The electrochemical performance of the membraneless fuel cell was examined in detail with respect to several critical system parameters.

The results demonstrated that the performance of the developed membraneless fuel cell enhanced profoundly if the concentration of oxidant in cathodic stream is 10 times larger, and the current density is also increased approximately ten times. But a variation of concentration for $[\text{Percarbonate}]/[\text{NaOH}]$ at the anode produced only a small influence on the cell performance. Thus, the present experimental results have confirmed that this membraneless microfuel cell is cathodic limited and suggest that it is a crucial factor in improving cell performance to increase the concentration of oxidant in the cathodic stream. In this experiment, we also examined the thermodynamic properties for the overall reaction of the MLSPCFC.

The flexibility of membraneless fuel cells to function with different media allowed the successful operation of mixed alkaline and acidic fuel cells. The membraneless microfuel cell system investigated in this study seems to be a good candidate for feasible application because its performance is comparable to an air-breathing DMFC without CO_2 emission.

In addition, the development of metal catalysts to accelerate the efficiency of MLSPCFC is in progress. Some further experimental works towards the microchannel and the flow rate of MLSPCFC will be beneficial to verify the present predictions and fulfill the practical utilization in portable power sources.

The MLSPCFC has the advantages of no CO_2 emission, use of aqueous fuel, and good cost-efficiency. Furthermore, percarbonate is a cheap, nontoxic, stable, easily handled, environmental friendly, large-scale industrial chemical and is a convenient source of hydrogen peroxide. We expect that the MLSPCFC may be a promising candidate for practical fuel cells to establish a clean and sustainable energy future.

4. Experimental Section

In the MLSPCFC configuration, we used an E-shaped laminar flow channel with catalyst-coated graphite plates of 1 mm (Graphite India, poco grade EDM-3, 0.0001 in. particle size) employed to act as electrodes [20]. The seed layers for the anode and cathode electrodes were applied via sputtering of 25–50 Å of chromium as an adhesion layer followed by 250–1500 Å of gold as the seed layer. Deposition of platinum black catalyst on these seed layers was performed via contact electrodeposition [21]. Since the reduced diffusion interface inherent in the side-by-side design reduces the amount of reactant wasted to the mixing region, these graphite plates are placed side by side with a specific spacing and form the length of the channel where the fuel and oxidant streams flow next to each other [22, 23].

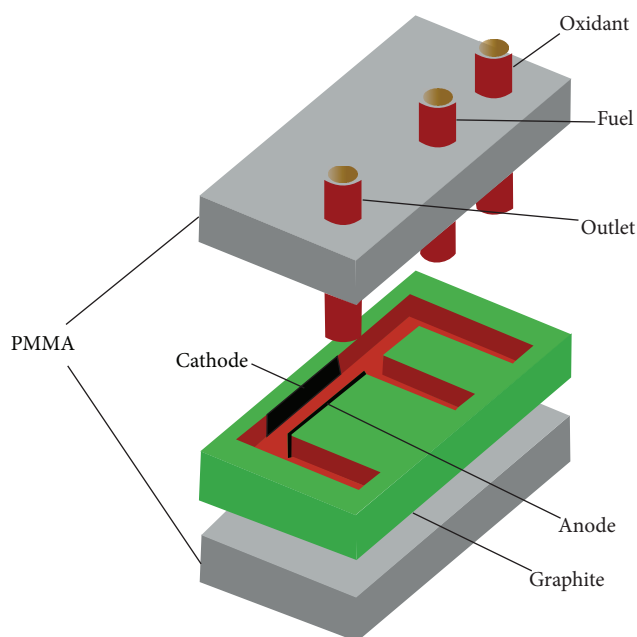


FIGURE 5: Schematic of the E-shaped membraneless laminar flow-based fuel cell with graphite plates molded with PDMS poly(dimethylsiloxane) and sealed with PMMA poly(methylmethacrylate).

Subsequent electrodeposition of catalyst to the cathode and anode, the E-shaped microfluidic channel structure is molded with PDMS poly(dimethylsiloxane), typically 1–10 mm in thickness, and finally sealed with a solid substrate such as 2 mm thick pieces of PMMA poly(methylmethacrylate) to provide rigidity and supportive strength to the layered system (Figure 5).

Silicon tubing (Instech Solomon PE = 205, I.D. 1.0 mm) is placed to guide the fuel and oxidant into the E-shaped channel systems at the top and to guide the waste stream out at the bottom of the channel. The tubing is inserted into holes that are punched exactly at the three ends of the E-shaped channel design and glued into place [24].

The fluid flow is regulated using a syringe pump with typical flow rates of $\sim 17 \mu\text{L/s}$ per stream. Consequently, the microfluidic cell keeps these fluids stable without a separation membrane [25].

All experiments were conducted using sodium percarbonate $2\text{Na}_2\text{CO}_3 \cdot 3\text{H}_2\text{O}_2$ (Riedel) with sodium hydroxide (Merck AR Grade) as the fuel and sodium percarbonate with sulphuric acid (Merck AR Grade) as the oxidant. The experiments were conducted in deionized water at room temperature. For fuel cell characterization the current and potential were measured at different loads using a variable resistor.

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