

Operation of liquid e-fuel cells using air as oxidant

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Abstract

The promising features and capabilities of liquid fuel cells for power generation and further application in electric vehicles have drawn more research attention in recent times. Notably is the recently proposed and demonstrated fuel cell which employs an electrically rechargeable liquid fuel (e-fuel) along with a catalyst-free material at its anode. The smooth and successful functioning of the e-fuel cell generally requires the supply of oxidant at its cathode. However, the space and storage requirement with the use of pure oxygen as oxidant not only limits the energy density of the fuel cell, but as well results in higher cost. Herein, the operation and performance of this e-fuel cell is examined with the use of air as oxidant, in lieu of the commonly used pure oxygen, at the cell cathode. A peak power density of 168.3 mW cm^{-2} was obtained when the cathode is fed with air. Such performance when air is employed as oxidant in the e-fuel cell outperforms most conventional direct liquid fuel cells that are even fed with oxygen

and thereby demonstrates the use of air as a promising alternative for future application. The performance of the cell when operated with air at various flow rates and operating temperatures, and during a constant current discharge test were also examined. This study therefore provides basis and useful insights for future investigations towards utilizing and catalyzing air as oxidant in the e-fuel cell for improved cell performance.

Keywords: Oxidant; Air; Fuel cell; liquid e-fuel cell; Fuel cell electric vehicles

1. Introduction

The continuous increase in the global population, industrialization, and numerous human activities has inevitably resulted in the surge of energy and power demands. In addition, the significant consumption as well as the rapid depletion of inefficient fossil energy has created severe energy crisis and insecurity across the globe.^{1, 2} In a bid to provide clean and sustainable power for the present energy-demanding modern lifestyles and the future generations, renewable energy sources have been largely explored and harnessed.³ Following this, various renewable power generation and energy conversion systems, that are energy efficient as well as environmentally benign, have been developed and deployed for use in several applications. Fuel cells, in particular, are clean and alternative energy provider well known for the smooth conversion of chemical energy stored in form of fuel into electrical energy for use in portable, stationary, and mobile devices.^{4, 5} The progress attained over the years on fuel cell technology has therefore gained the attention of many researchers for further studies, design, and development towards improved performance.

Direct liquid fuel cells are among the common and promising types of fuel cell technology that is currently attracting increasing attention due to their simple structural design, instant recharging, high energy density, safe handling, storage, and transport of fuel.^{6, 7} Liquid alcohols including methanol,⁸ ethanol,⁹ ethylene glycol,¹⁰ and even formic acid¹¹ have thus been commonly considered as choice of fuel to energize fuel cells. However, some of the peculiar challenges and shortcomings with the operation and application of these direct liquid fuel cells include severe fuel crossover, high cost

of catalyst, low power density, and low energy efficiency.^{12, 13} To tackle most of these challenges and ameliorate fuel cell performance and durability, alternative and more efficient liquid fuel has been recently presented and experimentally illustrated.

Recently, a novel concept which involves energizing fuel cells with an electrically rechargeable liquid fuel (e-fuel), in lieu of hydrogen and liquid alcohols, was introduced.^{14, 15} Potential materials for the production of such e-fuel were mentioned to include inorganic and organic materials, as well as suspension of particles. Following this, vanadium ions dissolved in sulphuric acid has been employed as an e-fuel at the anode while using pure oxygen as the oxidant to establish the operation and performance of a liquid e-fuel cell at room temperatures,^{14, 16} low and subzero temperatures,¹⁷ high temperatures,¹⁸ and even as a passive fuel cell.¹⁹ Excellent reactivity, simple storage, broad operating temperature scope, and catalyst-free component at the anode were all cited as few of the various advantages of utilizing this notable liquid e-fuel. The e-fuel cell was therefore described to attain remarkable performances, such as a peak power density of 293 mW cm^{-2} at room temperature and also an energy efficiency around 42 % operated at 80 mA cm^{-2} as the constant current density,¹⁴ surpassing most of the common liquid fuel cells. This therefore highlights the worthwhile potentials of the e-fuel cell not only for stationary applications but also in the automotive industry such as in fuel cell electric vehicles. As a result of the proliferating interest in the design and operation of electric vehicles, the feasibility of employing a power source that is easy to be re-fueled by merely supplying the fuel tank with rechargeable solution is an interesting idea that should receive enormous attention

for further development.

The smooth and successful functioning of the e-fuel cell generally requires the supply of oxidant at its cathode. The oxidant is required to interact with the protons that move across the membrane from the anode and also combine with the incoming electrons from the anode, through the external circuit, to actualize reduction reaction at the cathode.^{20, 21} However, the space and storage requirement with the use of pure oxygen in the course of the practical application of the e-fuel cell not just reduces its energy density, but as well results in higher financial cost. The use of air, on the other hand, would significantly reduce the design complexity and cost of the system. It is thus desired and of significant importance for the e-fuel cell to be workable using air as oxidant. Herein, the operation and behavior of the e-fuel cell is therefore examined with the use of air as oxidant, rather than the use of pure oxygen. The experimental study shows that the cell exhibits a peak power density of 168.3 mW cm^{-2} when air is employed as the oxidant. Such performance when air is used as the oxidant outperforms most conventional direct liquid fuel cells that are even fed with oxygen at the cathode. This therefore demonstrates the use of air as a promising alternative for oxidant in the design, development, and smooth running of the e-fuel cell for future application. The performance of the cell when operated with air at various flow rates and operating temperatures, and during a constant-current discharge test were also examined.

2. Operation principle of the fuel cell

As displayed in **Fig. 1.**, the key components of the cell include a graphite-felt anode, Nafion 117 membrane, platinum/carbon coated carbon paper as cathode, a pair of gaskets, flow fields, current collectors, and end plates. An e-fuel solution which contains V^{2+} ions is supplied into the anode where the V^{2+} is oxidized to V^{3+} during the cell operation according to:¹⁴



The oxidant is delivered into the cathode while the electron from the external circuit and the incoming protons from the anode is transported to the cathode to partake in the reaction according to:¹⁶



The ruling reaction of the e-fuel cell derived by merging the two reactions above is expressed as:



As previously reported,¹⁴ the calculated voltage of this e-fuel cell at 1.49 V, based on theory, is considerably higher in comparison to other types of direct liquid as well as hydrogen fuel cells.

3. Experiment

3.1 Preparation of membrane electrode assembly

The membrane electrode assembly (MEA) adopted for this e-fuel cell was prepared by sandwiching a cationic exchange membrane (Nafion 117) between the anode and a platinum/carbon coated carbon paper used as the cathode. While the dimension of the

membrane is 3.0 cm × 3.0 cm, both the anode and the cathode are of the size 2.0 cm × 2.0 cm. Before use, the membrane was prepared and pretreated by following the standard preparation method reported in a previous study.²² The catalyst-free graphite-felt anode was treated by heating in the air for 5.0 hours under a temperature of 500°C. The cathode, a platinum/carbon coated carbon paper with a metallic loading of approximately 0.50 mg cm⁻², was prepared using the process as previously reported.¹⁴

3.2 Experimental setup and instrumentation

The experimental arrangement for this study involved the assembled cell, an e-fuel tank, delivery pipes, and a peristaltic pump for the delivery of the e-fuel into the cell. The e-fuel solution employed at the anode was produced by dissolving VOSO₄ powder in H₂SO₄ and afterwards the solution is charged using a classic flow cell as reported before.²³ Thereafter, 20.0 mL of the e-fuel was drawn into the e-fuel tank and pumped into the fuel cell using the pump at a flow rate of 20.0 mL min⁻¹. The oxidant pipe is also fastened to the cathode side to deliver air to the cell cathode such that the oxidant flow rate is regulated using a mass flowmeter. The polarization and the constant current discharge tests were carried out using a testing system namely Arbin BT2000. The efficiencies of the cell are thereafter calculated according to the following equations:¹⁴

$$\text{Faradic efficiency (\%)} = \frac{\int i(t)dt}{VFc_{init}} \quad (4)$$

Where $i(t)$, V , F , and c_{init} stand for the discharging current, volume of e-fuel, Faraday's constant, and the initial concentration of the e-fuel, respectively.

$$\text{Voltage efficiency (\%)} = \frac{E_{aver}}{E_{theo}} \quad (5)$$

$$\text{where } E_{theo} = \frac{\int_0^{c_{init}} [E^0 + \frac{RT}{F} \ln(\frac{c_{V^{2+}}}{c_{V^{3+}}})] dc}{c_{init}} \quad (6)$$

E_{aver} and E_{theo} denote the average discharging and average theoretical voltage, respectively.

The energy efficiency is the multiplication of the Faradic efficiency and the voltage efficiency, expressed as:

$$\text{Energy efficiency (\%)} = \text{Faradic efficiency} \times \text{Voltage efficiency} \quad (7)$$

4. Results and Discussion

4.1 General performance and comparison

Fig. 2a. shows the performance characteristics of the e-fuel cell, as regards polarization and power density curves, with the use of air as oxidant in the operation of the cell. While the anode is supplied with an e-fuel solution (0.6 M V^{2+} dissolved in 3.0 M H_2SO_4) at a flow rate of 20.0 mL min^{-1} , air is supplied to the cathode side at a flow rate of 100 standard cubic centimeters per minute (sccm). It is found that the cell attains an open-circuit voltage (OCV) of 1.25 V with the use of air at the cathode as shown in **Fig. 2a**, which is comparable to an OCV of 1.26 V obtained when the cathode was fed with pure oxygen as shown **Fig. S1**. These values are apparently lower than the theoretical voltage of the of the e-fuel cell (1.49 V) as a consequence of the relatively high overpotential at the cathode due to the crossover of the e-fuel to the air side.¹⁴ It can also be seen that a peak power density of 168.3 mW cm^{-2} together with a maximum current density of approximately 450 mA cm^{-2} were obtained when air is applied as the oxidant. As expected, the use of pure oxygen as oxidant in the e-fuel cell produced a

higher peak power density of 254.6 mW cm^{-2} , as depicted in **Fig. S1.**, which is 51 % higher than the use of air at the cathode. This could be majorly ascribed to the fact that the air contains less concentration of oxygen (21 %) compared to that of pure oxygen.²⁴ However, the performance achieved by the e-fuel cell when air is utilized as the oxidant still outperforms most conventional direct liquid fuel cells that are even fed with oxygen at the cathode as shown in **Fig. 2b.** and **Table S1.** For instance, a close look at the comparisons shows that the operation of an ethanol fuel cell using air at the cathode could barely attain a power density of 2.0 mW cm^{-2} at room temperature, despite the higher catalyst loading (Pt/C 4.0 mg cm^{-2}) employed at the cathode.²⁵ A methanol fuel cell which also employs air at the cathode and operated at higher temperature (90°C) could only achieve a peak power density of $110.77 \text{ mW cm}^{-2}$.²⁶ Elsewhere, the peak power density (160.7 mW cm^{-2}) of a formic acid fuel cell that was even operated with oxygen at higher temperature (60°C)²⁷ is still lower than the performance achieved in this study that was even conducted at room temperature. These therefore justify the use of air as a promising alternative for oxidant during the operation and more importantly future application of the e-fuel cell.

4.2 Influence of different flow rates

The flow rate of oxidant into the cathode is one of the significant factors which impact the performance outlook of the cell.²⁵ The successful functioning of an e-fuel cell generally requires the supply of an oxidant at the cathode to accept the electrons arriving from the anode. The availability and mass transport of oxidant at the cathode and the cell performance would therefore be affected by the oxidant flow rate. In this

section, the influence of various air flow rates starting from 10 to 100 sccm were investigated and compared as regards to the cell performance. The performance of the cell is seen to improve with increasing air flow rate from 10 to 100 sccm as shown in **Fig. 3a**. This is primarily because increase in the air flow rate enhances the swift supply and spread of oxidant over the cathode thereby leading to the faster rate of the oxidant to the reaction sites at the cathode catalyst layer.⁸ In more detail, the cell attains a peak power density of 97, 141.4, 159.5, 165, and 168.3 mW cm⁻² at air flow rate of 10, 20, 30, 50, and 100 sccm, respectively. Whereas, as shown in **Fig. 3b**, when pure oxygen is used as the oxidant in the e-fuel cell, serving as the benchmark, little variation in the performance of the cell is obtained at all the flow rates. With the use of air at such low air flow rates of 10 and 20 sccm, limited cell performance is well pronounced. This can be ascribed to the insufficient flow of air to the cathode leading to oxygen starvation and increase in the mass transfer resistance of the oxygen component of air to the active sites of the cathode catalyst.²⁸ In contrast, at higher air flow rates of 50 and 100 sccm, very small difference in the peak power density, between 165 and 168.3 mW cm⁻² is demonstrated, which thereby indicates a further increase in the air flow rate may not be necessary for the smooth running of the e-fuel cell. Furthermore, it is considered that a further increase in the air flow rate would require the consumption of more power for the air delivery, which thereby could reduce the system efficiency.²⁹ While at low oxygen flow rate of 10 sccm, the supply of oxygen to the cathode is seen to be sufficient for achieving a high cell performance, further study is therefore suggested to develop

catalyst with better catalytic reactivity to oxygen reduction reaction so that the e-fuel cell using air as oxidant could match-up with the performance of cell using oxygen.

4.3 Influence of operating temperature

Operating temperature is a major parameter that should be considered in the operation and design of a fuel cell. This is because the operating temperature of the cell poses significant effect on the characteristics of the e-fuel, oxidant, and membrane and the general cell performance.^{30, 31} **Fig. 4a.** displays the effect of operating temperature on the cell voltage and power density with the use of air as oxidant. Obviously, with the rise in the operating temperature from 23°C to 60°C, performance improvement in the e-fuel cell is achieved. The peak power density of the cell when fed with e-fuel/air and subjected to an operating temperature of 23°C is 143.3 mW cm⁻². This power density was increased by 26 % to attain 180.7 mW cm⁻² at 40°C. A further increase in the operating temperature of cell to 60°C exhibit a peak power density of 199.5 mW cm⁻² which is about 40 % more than the performance at 23 °C. The increase in cell performance is accredited to the fact that increasing temperature enhances the reactions kinetics of the e-fuel oxidation reaction at the anode and the reaction at the cathode which therefore reduces activation losses during the cell operation, in accordance with Arrhenius equation.^{25, 32} Increasing temperature also facilitates the transport of reactants at the anode and cathode to their respective active sites, which thus reduces concentration loss.³¹ The performance of the cell using pure oxygen as the oxidant in the e-fuel cell is displayed in **Fig. 4b.** The peak power density obtained are 203.3, 273.9, and 315.9 mW cm⁻² when the cell is subjected to operating temperature at 23°C, 40°C,

and 60°C, respectively. It was observed that when feeding the e-fuel cell with air at the cathode and operated at 60°C, the cell achieved nearly the same performance, a peak power density of approximately 200 mW cm⁻², in comparison to the performance of the cell fed with oxygen but operated at 23°C as shown in **Fig. S2**. This further emphasizes the potential and use of air as a promising option in the future operation and application of the e-fuel cell. However, it is worth noting that, when operated at high temperature, the dehydration of Nafion 117 membrane could happen, which thus would lead to increase in the membrane resistance and thereby induce a larger ohmic loss limiting the cell performance^{31, 33, 34} In addition, the cell is prone to high crossover rate of reactive species through the membrane at higher temperature which would also result in a larger mixed potential at the cathode.¹⁸ Hence, the long-term performance of the e-fuel cell at high operating temperature is an important aspect that still requires further investigations.

4.4 Constant-current discharge performance

Constant-current discharge is an evaluation test method commonly conducted on power generation devices such as fuel cells to determine their performance characteristics during real and practical applications. In addition, constant-current discharge test provides the basis for evaluating the Faradic, voltage, and energy efficiencies of fuel cells to further examine their suitability for real-life operation. In this study, the constant current discharge test of this e-fuel cell is tested at 10.0 mA cm⁻² considering five different flow rates of the oxidants – 10, 20, 30, 50, and 100 sccm with the use of air as oxidant as shown in **Fig. 5a**. For comparison, the constant current discharge test of this

e-fuel cell with the use of pure oxygen, which is the widely accepted basis is displayed in **Fig. 5b**. As it can be seen, there is not much improvement in the voltage plateau with the increase in flow rate of both oxidants, at all the flow rates considered for air and oxygen. With the use of air, the discharge capacity of the cell slightly increases in tandem with the increasing air flow rate. In more detail, an improved discharge capacity performance from 6.44 to 7.16 Ah L⁻¹ was observed when the air flow rate at the cathode was raised from 10 to 100 sccm. With the use of pure oxygen, the discharge capacity of the cell also increases from 6.77 to 7.28 Ah L⁻¹ as the oxygen flow rate increases at the cathode. The primary reason for such increase in discharge capacity is that higher oxidant flow rate promotes fast spread distribution of the oxidant at the cathode catalyst layer which in turn reduces the concentration and activation losses that could affect the discharge capacity.^{35, 36} Using air as the oxidant at all these flow rates, the cell is found to demonstrate its capability for stable operation. This therefore also justifies the use of air as a promising alternative with adequate performance for future application.

4.5 Energy efficiency of the cell

The energy efficiency indicates the competence of the fuel cell regarding the efficient conversion of chemical energy to electrical energy. Here, the Faradic, voltage, and energy efficiencies of the cell considering various flow rates of air were calculated according to the equations (4), (5), and (7), respectively, to further reveal the performance of the cell. As displayed in **Fig. 6a.**, it can be noticed that the energy efficiency of the cell when fed with air at a flow rate of 10 to 100 sccm is between 23.4 % and 26.4 %. In comparison, when the cathode is fed with pure oxygen, the energy

efficiency of the cell as depicted in **Fig. 6b**. is slightly higher and ranges from 26.8 % to 29.6 %. The slight increment of energy efficiency can be ascribed to the reality that pure oxygen possesses more oxygen concentration which brings about the reduction of activation loss.²⁴ However, the usage of air for substituting pure oxygen is found to be able to provide the cell with a similar performance in terms of achieving comparable energy efficiency. Furthermore, the efficiency attained by the e-fuel cell with the use of air as oxidant also outperforms some conventional direct liquid fuel cells that uses air as oxidant and operated under similar testing conditions as summarized in **Table S2**. All these therefore demonstrates the use of air as a promising oxidant for the operation of the e-fuel cell. It is also essential to state that during the long-term running of the e-fuel cell, the oxidant flow rate also plays a major effect on removing the water generated at cathode and therefore still requires further investigation on determining the optimal oxidant flow rate after prolonged operation. Also, it is believed that by enhancing the cathode catalytic reactivity in order to ameliorate the sluggish oxygen reduction reaction at the cathode towards reducing activation loss and thereby achieving a higher voltage efficiency, the energy efficiency of the e-fuel cell employing air as oxidant can be further improved.

5. Summary

In this study, a liquid e-fuel cell was explored using air, rather than the common use of pure oxygen, as the oxidant at the cathode. The experimental result shows that the cell is able to boost of a peak power density of 168.3 mW cm^{-2} which performs better than a number of the common direct liquid fuel cells, including those fed with oxygen at the

cathode. This therefore indicates the potential of air as oxidants in the working of the e-fuel cell and also necessitates the use of air as a promising alternative for future application. The effects of a number of operating variables on the behavior and performance of the cell with respect to using air as the oxidant have also been examined. The cell performance is seen to improve with increasing the air flow rate as high air flow rate enhances the quick supply and spread of oxidant to the reaction sites at the cathode. Likewise, with the increase of the operating temperature from 23°C to 60°C, performance enhancement in the e-fuel cell is achieved. To further improve the cell performance obtainable with the use of air in comparison to pure oxygen, more research attention, such as the fabrication of catalyst with improved catalytic reactivity for the cathodic reaction, should be considered in the future. The influence of the possible water flooding phenomenon at the cathode during the cell operation should also be investigated and studied. A numerical investigation to examine and analyze the influence of the use of air and pure oxygen as oxidant on the cell performance is also a potential direction for future studies.

Declaration of Competing Interest

The authors declare that they have no conflict of interest.

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Figure captions

Fig. 1. Fabrication of the fuel cell.

Fig. 2. (a) General performance of the cell using air as the oxidant, (b) Power density comparison with the data obtained from the open literature.

Fig. 3. Polarization curves and power density of the cell at different (a) air flow rates, and (b) oxygen flow rates.

Fig. 4. Comparison of the cell performance at different operating temperature using (a) air and (b) oxygen as the oxidant.

Fig. 5. Constant-current discharge performance of the cell at various (a) air flow rates and (b) oxygen flow rates.

Fig. 6. Faradic, Voltage, and Energy efficiencies of the cell at various flow rates of (a) air and (b) pure oxygen.

Figures

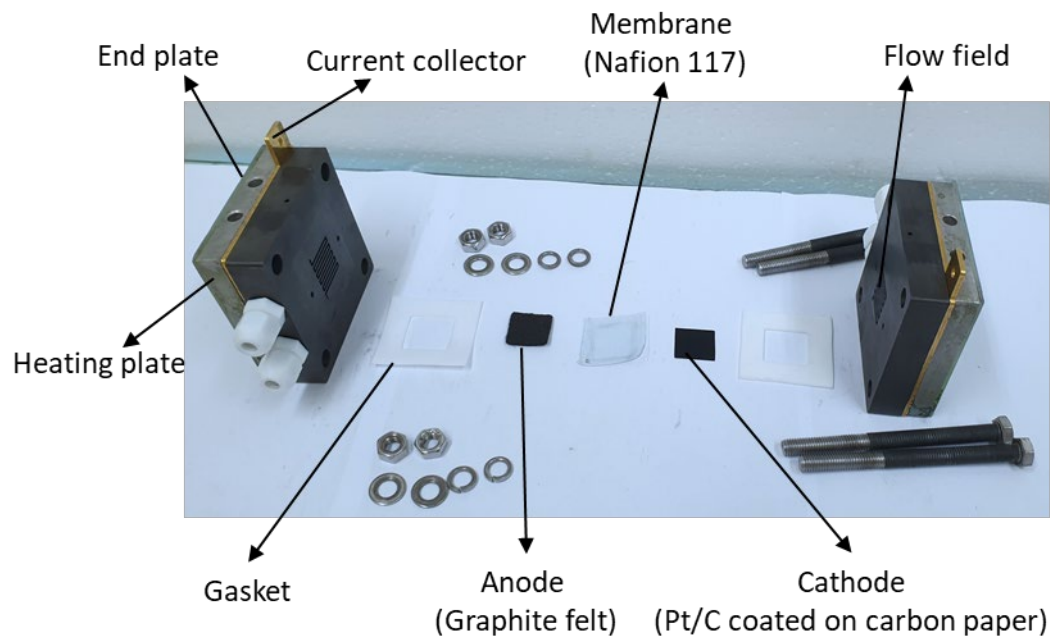


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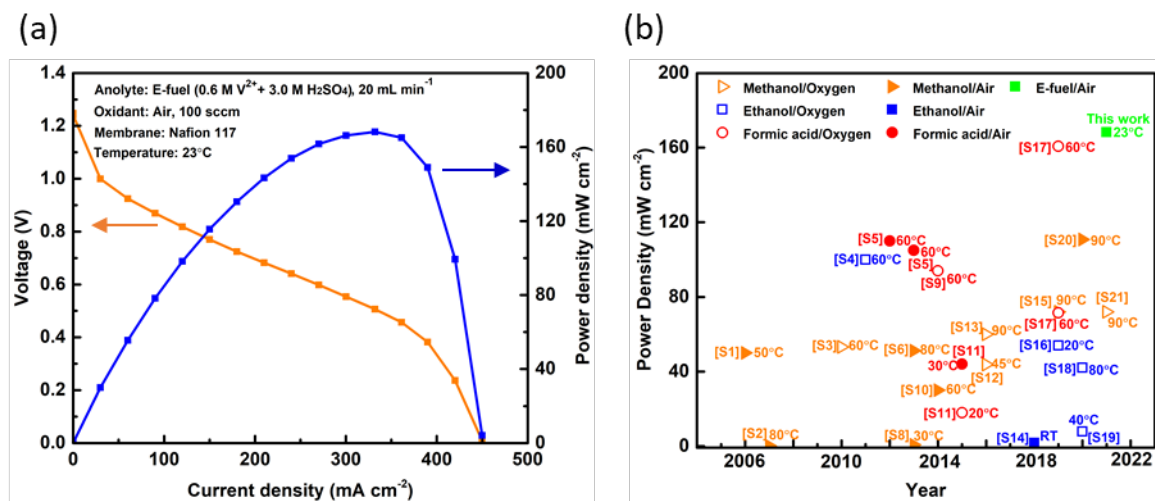


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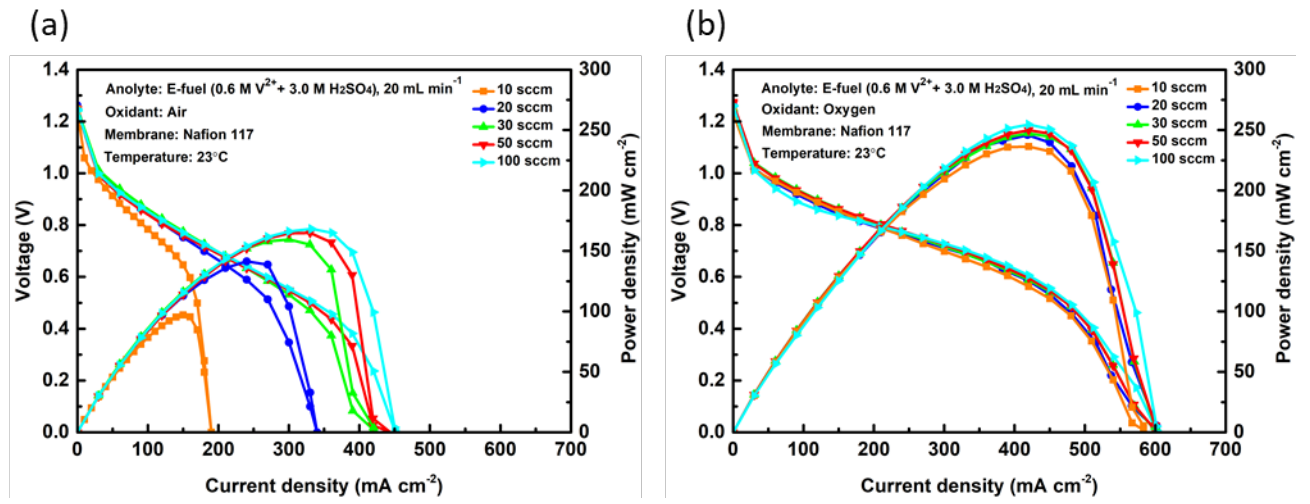


Fig. 3. Polarization curves and power density of the cell at different (a) air flow rates and (b) oxygen flow rates.

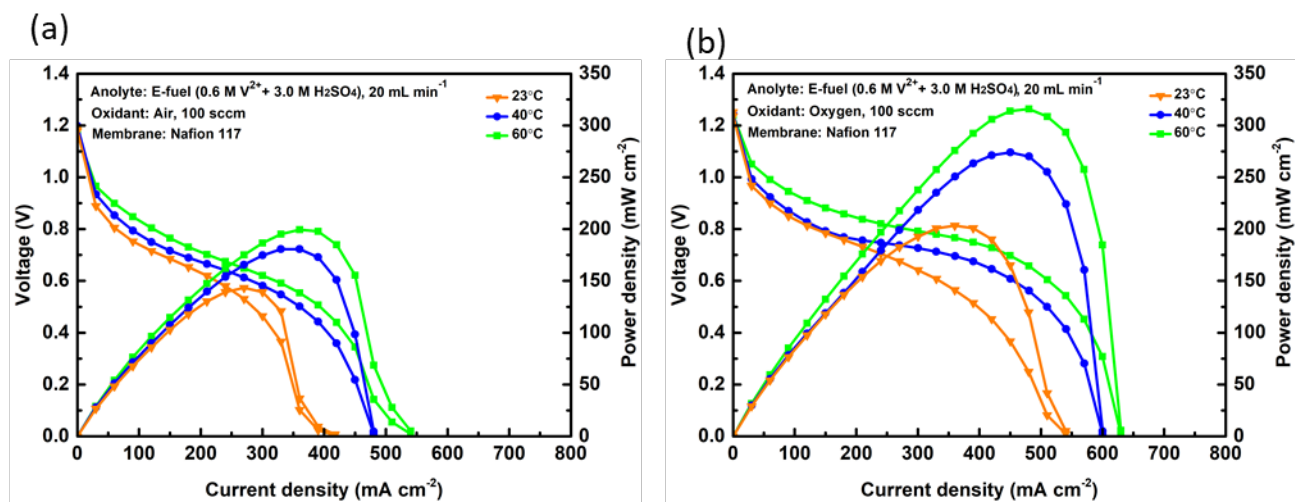


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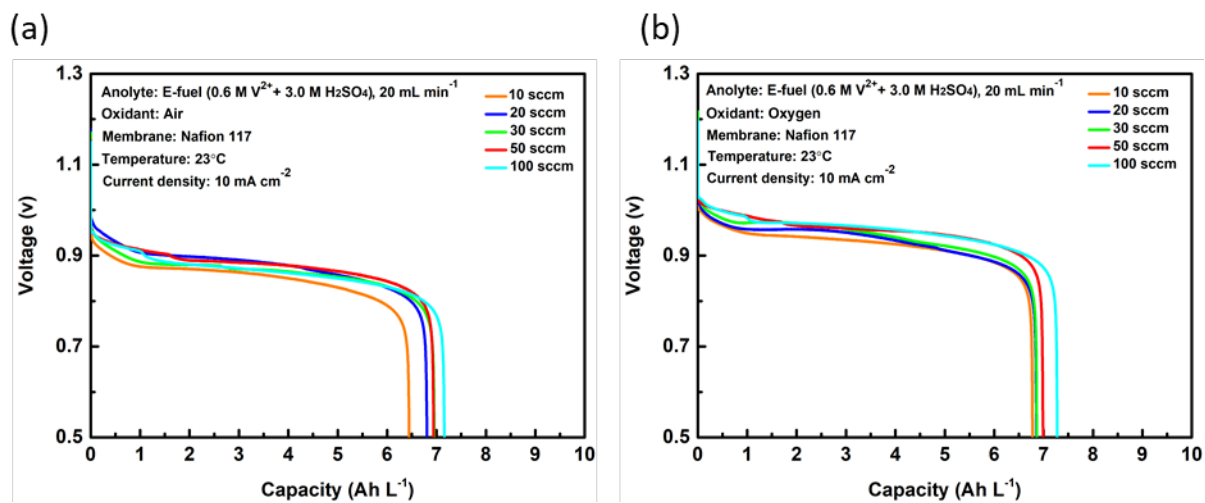


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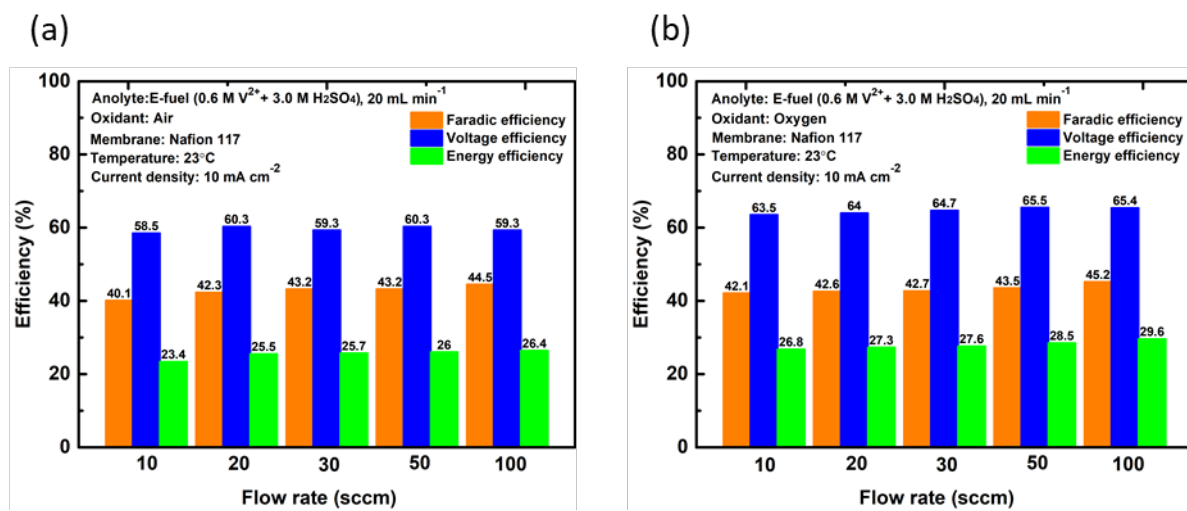


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