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# Passive Direct Methanol Fuel Cell Stack: Design and Performance Characteristics

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**ABSTRACT**

*In this study, passive direct methanol fuel cell (DMFC) stacks consisting of six cells with a porous carbon nanofiber (CNF) anode diffusion layer have been successfully designed, fabricated, and tested. The performance of the passive DMFC stacks connected in series was tested and compared for mono-polar and bi-cell designs. Each stack consisted of six MEAs with an active area of 4 cm² each and was tested at ambient temperature using 3 M diluted methanol. The experimental results indicate that the mono-polar stack exhibited higher performance, with a maximum power of 550 mW, which was 23.8% higher than that of the bi-cell stack. The output voltage can be maintained at approximately 2 V for 7 h with 21 ml of 3 M diluted methanol at a constant current discharge of 60 mA. Furthermore, it can be observed that both stacks display a stable and reproducible performance, which can be restored once the fresh methanol is refilled. Additionally, it can be observed that the difference in cell efficiency between the two stacks is less substantial. Overall, only approximately 35% of the methanol was utilized in each stack.*

**Keywords:** Carbon nanofiber, Efficiency, Passive Direct Methanol Fuel Cell, Stack Design.

## 1. INTRODUCTION

In the past few years, the demand for a clean, renewable power source has increased rapidly due to the rise of environmental issues, such as air pollution and greenhouse gas emissions. Fuel cells have emerged as an attractive alternative to current power sources due to their high efficiency and cleaner operation compared to fossil-fuel power units. There are different types of fuel cells, typically defined by the electrolyte used, that generally work in a similar manner. The direct methanol fuel cell (DMFC) offers certain advantages, including compactness, a simple design, light weight, and low cost [1]. These benefits ensure its application particularly in portable devices, such as cellular phones, notebooks, tablets, and portable music players. The DMFC's passive mode operates without any auxiliary devices (such as a pump and blower), making it even more attractive and competitive in portable applications due to its simplified configuration. However, the output voltage of a passive DMFC single cell is typically lower (±0.5 V) and incapable of performing in practical applications [2]. Therefore, to achieve the desired energy for certain electrical appliances, single cells are typically stacked in series (to increase voltage) or parallel (to increase current) to maximize power output.

In addition to the need for higher power output, a passive DMFC stack with long operation time, quick start-up, and short charging time is necessary to meet the requirements of a usable power device. A few studies have reported to experimental design and analysis of a passive DMFC

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stack in the literature. Achmad et al. [3] developed a stack of six dual DMFCs connected in series; the stack delivered a maximum power output of 600 mW and was successfully used to charge a mobile phone and power an MP3 player. Tsujiguchi et al. [4] demonstrated a high-performance DMFC stack consisting of 8 cells, using PCP and 100% methanol as the fuel. A maximum power output of 1.8 W was achieved using a novel octagonal stack design to ensure uniform output across each unit cell. Conversely, Zhu et al. [5] studied the effects of fuel concentration and stack design on a DMFC stack. They presented a planar 8-cell DMFC twin-stack with a total active area of 32 $cm^2$, and a maximum power output of 540 mW was obtained at 1.8 V. Their results indicated good stability of the DMFC stack, enabling the powering of a sensor node for 39 h when equipped with a designed fuel-feed device. Feng et al. [6] evaluated the practical operating performance of a new small passive DMFC stack. Combined with supercapacitors, it provided a large current-pulse discharge; when combined with a DC-DC converter, the stack provided a much more stable voltage output. However, after long-term operation, the stack showed degradation, which is an important problem that needs to be solved before it can be realized as a portable application for passive DMFCs. Additionally, Wang et al. [7] designed, fabricated, and tested a 4-cell modular passive DMFC stack that was flexible enough to be integrated to numerous electronic devices. The modular stack achieved a maximum power output of 27 mW cm-2 using 5 M diluted methanol. The results exhibited a good response and reproducibility at high currents (>100 mA). Hashim et al. [8] developed a multi-cell stack of passive DMFCs. The micro stack, with an active area of 6 cm2, achieved an optimal power output with 4 M diluted methanol (12.05 mW at 1.08 V and 11.2 mA). In addition to research centers and universities, several companies have been actively involved in developing DMFCs as an alternative power source, including NEC, Sony, Toshiba, and Fujitsu [9].

The fuel cell prototypes primarily support and improve current power sources, which are dominated by battery-based technology, due to their high energy density, long life, and reliability. However, unresolved issues need to be addressed before they can be considered commercially competitive. One of these issues is the stack's hardware and design configuration. Generally, there are three basic types of fuel cell stack design: bipolar, mono-polar, and bi-cell [10,11]. The most common design for passive DMFCs is based on mono-polar and bi-cell stack configurations. Hence, in the current study, the relationship between stack structure designs and operational performance was evaluated and analyzed. A simple passive DMFC stack consisting of six MEAs with a CNF porous layer in the anode electrode was designed and fabricated in both mono-polar and bi-cell configurations. Considering the output voltage desired by an electrical device, the 6 MEAs are connected in series. The objective of this study was to evaluate the performance of the passive 6-cell DMFC stack with an integrated serial cell, comprising mono-polar and bi-cell stacks, operating at ambient temperature to ensure the DMFC is adoptable for portable applications. The current-voltage characteristic and constant-current discharge of the designed stack were analyzed to assess the effectiveness of the stack design. Additionally, to evaluate stack stability, a long-term test was conducted in a constant-current discharge mode, with five start-up and shut-down cycles using fresh refueled methanol. Furthermore, the effects of cell efficiency were investigated, including Faradic and energy efficiencies.

## 2. EXPERIMENTAL PROCEDURE

### 2.1 Stack Design and Test

In this report, a miniature 6-cell DMFC stack with two different stack designs (mono-polar and bi-cell) was proposed, as illustrated in Figure 1. The mono-polar stack was fabricated with dimensions of 82 × 65 × 34 mm, whereas the bi-cell stack was 85 × 38 × 57 mm. In the mono-polar stack, the cells were arranged in line on the same plane. The anodes faced the fuel reservoir, while the cathodes faced the other side. Six cavities were used as a fuel reservoir in the anode compartment; two of them were interconnected (up and down) and independent of the other two interconnected reservoirs. In the bi-cell stack, the dual cell was arranged in-out, with two anodes on the inside, through which the fuel passed, and two cathodes on the outside. The fuel reservoir was interconnected across each array, and there were 3 arrays in the 6-cell bi-cell stack, with the dual MEAs sharing a reservoir. The volume capacity of each cavity was 4.2 $cm^3$, resulting in a total volume of 25.2 $cm^3$ for the stack reservoir and approximately 21 ml of fuel. At the top of each cavity, two circular holes (∅ 5 mm) were drilled for easy fuel entry and $CO_2$ removal.

**Figure 1:** Miniature 6-cell passive DMFC stack developed in this study: (left) mono-polar and (right) bi-cell design.

In the cathode compartment, an opening was designed to allow oxygen from the environment to enter. Figure 2 illustrates schematics of the two assembled 6-cell stacks. Similar series connections were applied for a banded principle structure [12], where the anode current collector of the odd cell was connected to the cathode current collector of the even cell, and the anode current collector of the last cell (cell number 6) and the cathode current collector of the first cell (cell number 1) were free to connect to the external circuit for further performance tests.

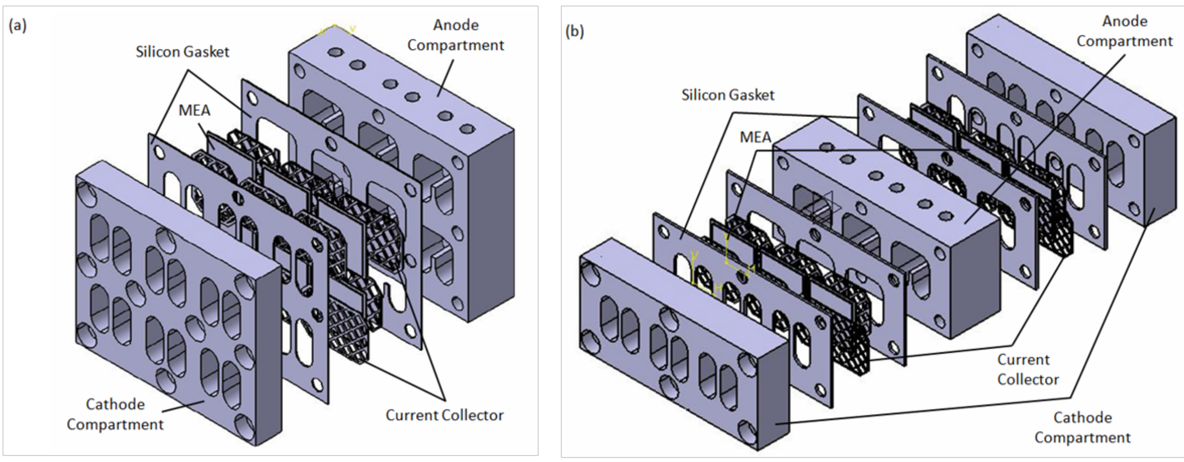


**Figure 2:** Schematic of 6-cell passive DMFC stack (a) mono-polar and (b) bi-cell.

The performance of the DMFC stack in passive mode was evaluated using current-voltage polarisation curves and a long-term discharge test. The stack was conditioned and activated overnight with 2 M diluted methanol at ambient temperature before proceeding to the performance test. All measurements of stack performance were controlled and recorded using a Potentiostat/Galvanostat (WonATech, Korea). A diluted methanol solution at 3 M was fed into the fuel reservoir using a syringe, and oxygen was supplied to the stack by air spontaneously entering through the opening in the cathode compartment. After I-V characterization, the stacks were further tested for long-term discharge in constant-current mode, with the current fixed at 60 mA and the voltage response recorded.

### 2.2 Membrane Electrode Assembly

The unsupported catalyst Pt-Ru Black (HiSPEC 6000, Alfa Easer, USA) and Pt black (HiSPEC 1000, Alfa Easer, USA) were used as the catalysts for the anode and cathode layers, respectively; additional details on the MEA preparation and the characteristics of CNF can be found in our previous report [13,14]. The active area of one single MEA was 4 $cm^2$ with a catalyst loading of 8 mg $cm^{-2}$ in each electrode, and Nafion® 117 was used as the electrolyte membrane. The MEAs were fabricated by sandwiching the electrodes with the electrolyte membrane by hot-pressing at 135 °C at 12.5 kgf $cm^{-2}$ for 180 s.

## 3. RESULTS AND DISCUSSION

### 3.1 Stack Performance

Figure 3 depicts the voltage-current and power output of the 6-cell stack operating at ambient conditions with 21 ml of 3 M diluted methanol. The overall effective area of the stacks (mono-polar and bi-cell) is the same, i.e., 24 $cm^2$. The mono-polar stack achieved a maximum power output of 550 mW at a current of 300 mA, whereas the bi-cell stack produced a maximum power of 444 mW at 292 mA, corresponding to stack power densities of 22.9 mW $cm^{-2}$ and 18.5 mW $cm^{-2}$ for the mono-polar stack and bi-cell stack, respectively. The maximum power density of the mono-polar stack is 23.8 % higher than that of the bi-cell stack, indicating that the in-line stack structure is more useful for maintaining the high power density of the single cell. The one-plane fuel reservoir of the mono-polar stack could provide a higher and more uniform contact pressure distribution to the six MEAs, thus reducing the cell-to-cell power deviation [15].

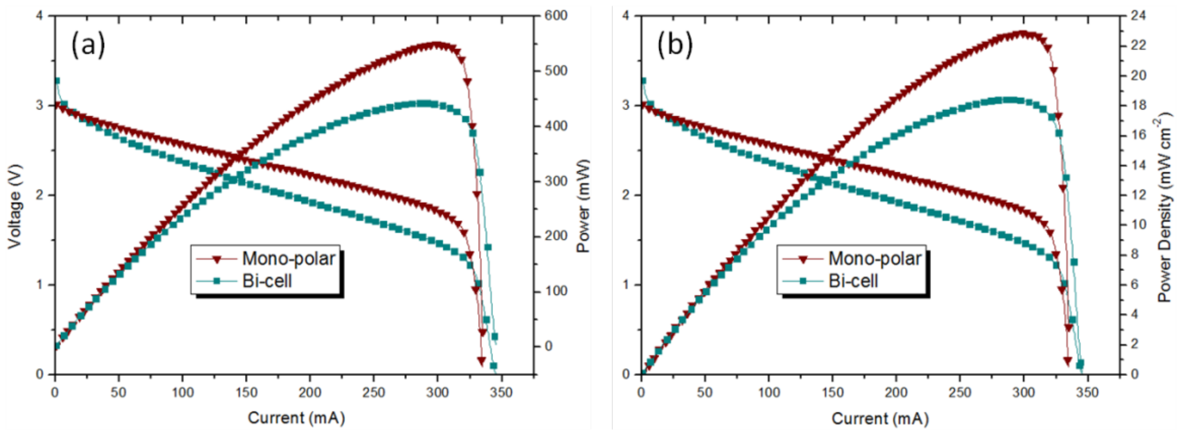


**Figure 3:** Performance curves of 6-cell passive DMFC with different stack designs; (a) power, (b) power density.

Furthermore, the increased temperature in the stack is advantageous for proton transport through the electrolyte because it lowers the internal cell resistance. However, one negative effect was that it increased the methanol crossover rate, leading to potential mixing at the cathode and reducing cell performance, especially at low current density.

### 3.2 Stack Stability and Reproducibility at Constant Discharge Current

A comparison of stack stability under a constant discharging current of 60 mA for both 6-cell stacks, when feeding 21 ml of 3 M diluted methanol, is shown in Figure 4a. The stacks were able to maintain high stability during an 8-hour test with relatively stable voltage outputs. In the case of the mono-polar stack, the voltage dropped from 2.6 (initial 5 min of OCV) to 2.0 V after applying the load and increased gradually and remained at 2.3 V between 3 to 7 h. The increasing trend in voltage from the beginning to the 3-h mark may be due to the abundant supply of methanol fuel for the reaction; however, the voltage retained after the 3-h mark is due to the temperature effect, which compromises other limitations, such as water flooding, fuel crossover, and mass transfer resistance. Lastly, the voltage rapidly dropped from 2.3 V to 0.03 V in the last 1 h. The last significant decrease in performance can be attributed to mass transfer polarisation loss, caused to methanol depletion in the fuel reservoir. The voltage output of the bi-cell dropped sharply from an OCV of 2.7 V, reached a steady voltage of approximately 1.8 V for 7 h, and then decreased again to 0.02 V in the last 1 h. There is a slight decrease in the voltage of the bi-cell between 4 to 7 h; one possible explanation is that the accumulation of water in the cathodes blocks the gas pathway and affects the overall performance. Thus, the fuel-consuming time for both 6-cell stacks were approximately 8 h when 21 ml of limited fuel was fed into the reservoir.

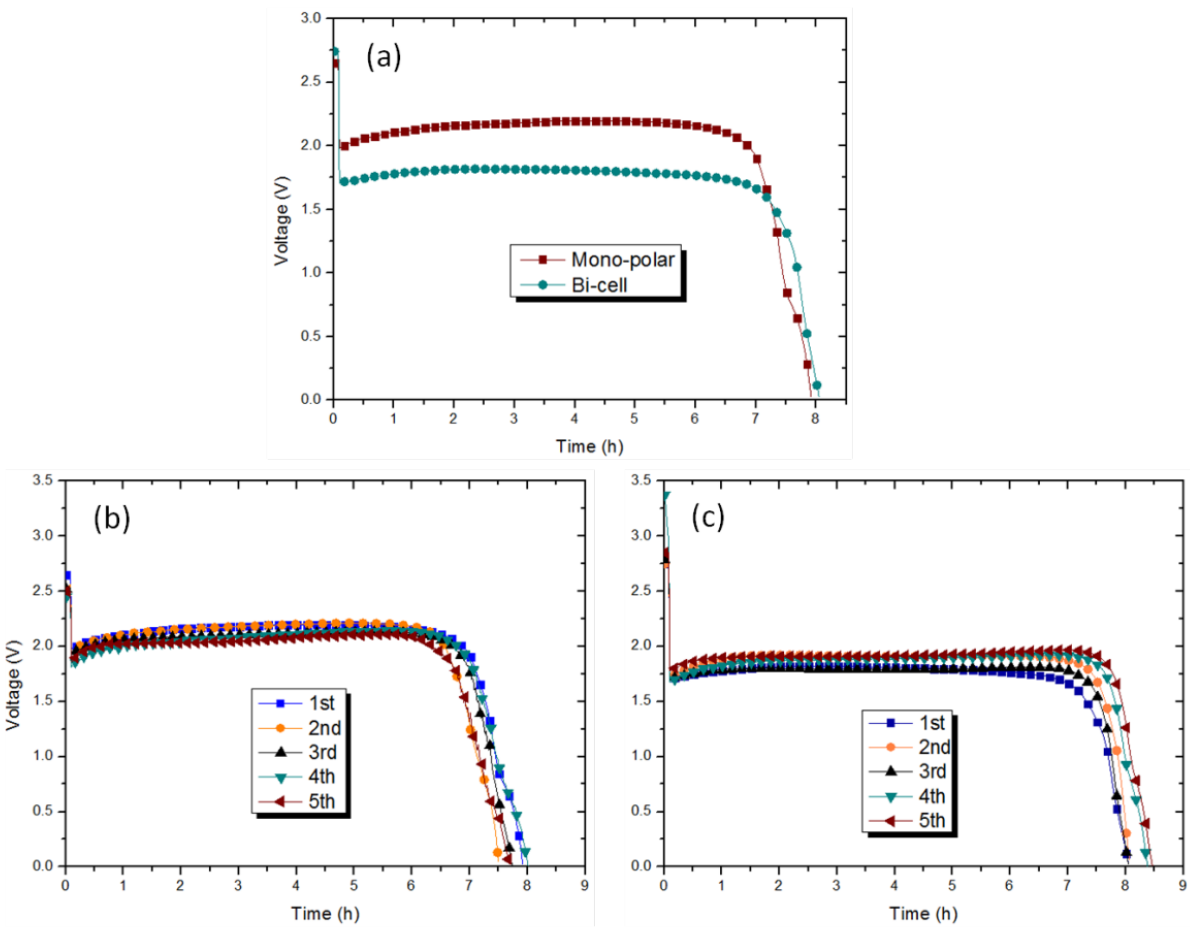


**Figure 4:** Transient voltage discharged at a constant current of 60 mA for a passive DMFC at room temperature (a), and reproducibility test with 5 refueling cycles; (b) mono-polar, (c) bi-cell. Fuel amount: 21 ml (3 M diluted methanol).

A practical passive DMFC stack should be capable of delivering reproducible and stable performance as long as it is provided with sufficient fresh fuel. The reproducibility of the designed stacks was evaluated by conducting a five-cycle long-term constant current discharging test at 60 mA. Fresh methanol solution was refilled into the fuel reservoir to replace the used methanol at each cycle, and the voltage started at OCV and dropped to zero at the end of the test cycle. The results are presented in Figures 4b and 4c. In the case of the mono-polar stack (see Figure 4b), it can be seen that the stack could be discharged for approximately 7.5-8 h, and a similar trend in the output voltage was generated for each cycle. The stack voltage exhibited a considerable decrease at the beginning of the discharging test and then slightly increased and reached a stable voltage of approximately 2.2 V. For the bi-cell stack (see Fig. 4c), the discharging time is approximately 8-8.5 h, which is slightly longer than that of the mono-polar design; however, the stable output voltage of the bi-cell was only 1.9 V. Furthermore, it can be observed that the output voltage and discharging time for the bi-cell stack increases for each additional cycle. Heat generation and accumulation in the stack may cause an increase over time during prolonged operation. After one cycle of the constant-discharging test, the methanol in the anode compartment is extracted and measured with a refractometer. After the reproducibility test, the concentration of the methanol was determined to be 0.654 M and 0.523 M for the mono-polar and bi-cell stacks, respectively.

Based on the reproducibility results, it can be concluded that the limited fuel supply was the primary reason behind the significant decrease in performance for the passive DMFC stack. After replacing the reservoir with fresh methanol at the end of each cycle, performance could be recovered and returned to its original state. Additionally, a nearly constant voltage output was observed during the 5-cycle reproducibility test, indicating the high stability of the stack performance in the current study.

### 3.3 Efficiency Output

The Faradic efficiency and the energy efficiency ($\xi$) were determined according to references [16-19]. The Faraday efficiency is the ratio of the actual discharge capacity to the theoretical capacity of fuel used in the stack. The Faradic efficiency is used to describe the efficiency of the fuel that is converted to useful current, which is also known as current efficiency. The Faraday efficiency can be calculated using Eq. (1) as follows:

$$\eta = \frac{Actual\ discharging\ capacity\ (Ah)}{Theoretical\ discharging\ capacity\ (Ah)} = \frac{it}{6\Delta C_m V_m F}\ .n.100 \quad (1)$$

where $i$ is the discharging current (60 mA); $t$ is the time for the discharging process; $n$ is the number of cells in the DMFC; $\Delta C_m$ is the methanol concentration difference between the beginning and the end of the long term test; $V_m$ is the volume of the methanol solution in the reservoir (21 ml); and $F$ is the Faraday constant ($F$ = 96485.3365 sA $mol^{-1}$).

Energy efficiency is the percentage of electric energy generated in the stack relative to the theoretical electric energy of the total injected fuel. Furthermore, the energy efficiency of the stack can be described in terms of energy (watt-hours) using the specified voltage and current. The energy efficiency can be defined in Eq. (2) as follows:

$$\xi = \frac{Discharging\ energy\ (Wh)}{Theoretical\ discharging\ energy\ (Wh)} = \frac{vit}{6\Delta C_m V_m F E}\ .100 \quad (2)$$

where $v$ represents the transient discharging voltage, and the theoretical voltage $E$ is 1.21 V for the DMFC.

The Faradic and energy efficiencies calculated using Eqs. (1) and (2) with different refueling times (1st-5th) for the mono-polar and bi-cell stacks are depicted in Figure 5. The results corresponded with those of Liu et al. [17]; in a passive DMFC system using 3 M diluted methanol, the Faradic and energy efficiency was 37.1% and 11%, respectively. It is observed that there is no substantial difference in cell efficiency between the mono-polar and bi-cell stacks; the Faradic and energy efficiency differences are 0.17% and 15.4%, respectively. In particular, only approximately 35% of the methanol was utilized in each stack; the remainder of the methanol solution permeated to the cathode and reacted directly with oxygen, generating heat. As reported in previous studies, most of the higher losses in the Faradic efficiency were due to the fuel crossover [17-19]. Table 1 summarizes the literature for passive DMFC stack experimental study.

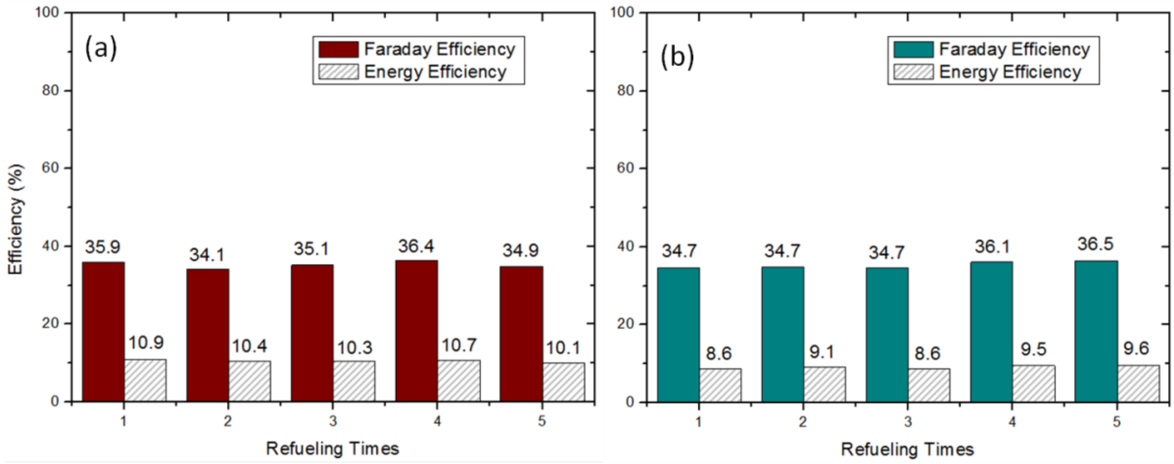


**Figure 5:** Faradic efficiency and energy efficiency of passive DMFC fuelled with 21 ml of 3 M diluted methanol at different stack designs; (a) mono-polar, (b) bi-cell and discharged at a constant current (60 mA).

**Table 1:** Summaries of the literature for passive DMFC stack experimental study.

| Stack Design | Methanol Concentration | No. of Cell, Active area | Power Output | Researchers |
|---|---|---|---|---|
| Dual stacks (Bi-cell based) | 5 M | 12, 48 cm$^2$ | 600 mW, 12.5 mW cm$^{-2}$ | Achmad et al. [3] |
| Twin-stack (Bi-cell based) | 4 M | 8, 32 cm$^2$ | 540 mW, 16.9 mW cm$^{-2}$ | Zhu et al. [5] |
| Small planar stacks (Bi-cell based) | 4 M | 10, 50 cm$^2$ | 1300 mW, 26 mW cm$^{-2}$ | Feng et al. [6] |
| Modular stacks (Bi-cell based) | 5 M | 4, 20 cm$^2$ | 545 mW, 27 mW cm$^{-2}$ | Wang et al. [7] |
| Bipolar stacks | 4 M | 6, 6 cm$^2$ | 12.05 mW, 2.01 mW cm$^{-2}$ | Hashim et al. [8] |
| Bi-cell stacks | 3 M | 6, 24 cm$^2$ | 444 mW, 18.5 mW cm$^{-2}$ | This study |
| Mono-polar stacks | 3 M | 6, 24 cm$^2$ | 550 mW, 22.9 mW cm$^{-2}$ | This study |

Due to voltage losses, the energy efficiency of the DMFC stack is generally lower than the Faradic efficiency, resulting in a deviation from the theoretical voltage. Additionally, the energy efficiency is affected by the discharge voltage, Faraday efficiency, and the number of cells in the stack [16]. Although the energy efficiency of both stacks is only approximately 10 %, i.e., 610 W h kg$^{-1}$ (based on the theoretical specific energy of methanol), it is still comparable to that of batteries (lithium-ion theoretical specific energy, 410 W h kg$^{-1}$) [17, 20]. Therefore, a passive DMFC is a good alternative for supporting and replacing batteries as a primary power source for portable applications. However, there are still several serious problems that need to be resolved for passive DMFCs to be the best alternative for portable power sources. Finally, Table 1 presents a comparison of the power output with other researchers. It can be observed that this study has successfully achieved the highest power output and power density for a passive DMFC, namely 550 mW and 22.9 mW cm$^{-2}$, respectively, at a very low methanol concentration of 3 M.

## 4. CONCLUSION

In summary, a 6-cell passive DMFC stack was successfully fabricated in two configurations. The stacks were analyzed based on their power generation and efficiency contributions. The results indicated that the power density of the mono-polar stack is 23.8% higher than that of the bi-cell stack. Furthermore, the experiments revealed that the one-sided arrangement is considerably better than the double-sided arrangement. Both stacks were able to maintain high stability during an 8-hour test with relatively stable voltage outputs. Based on the durability results, it can be concluded that a limited fuel supply was the primary reason behind the significant decrease in performance for the passive DMFC stack. After replacing the reservoir with fresh methanol at the end of each cycle, performance could be recovered and returned to its original state. The cell efficiency of both stacks indicated that the methanol utilization was only approximately 35%, with an energy efficiency of 10%. This report was written to analyze a stack design configuration suitable for passive DMFCs.

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**Author contributions statement:** Conceptualization, A.M. Zainoodin; Methodology, A.M. Zainoodin; Investigation, A.M. Zainoodin; Resources, S.K. Kamarudin & M.S. Masdar; Writing – Original Draft Preparation, A.M. Zainoodin; Writing – Review & Editing, A.M. Zainoodin, S.K. Kamarudin & M.S. Masdar.

**Data Availability Statement:** All data generated or analyzed during this study are included in this published article.