

A Review on Solid Oxide Fuel Cell Stack Designs for Intermediate Temperatures

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ABSTRACT

Solid oxide fuel cell (SOFC) has significant advantages of clean and quiet operation while providing a relatively high efficiency owing to enhanced reaction kinetics at high operating temperature. The high operating temperature of SOFC, typically around 800 – 1000°C helps to enable internal reforming of hydrocarbons and negate effects of impurities in small quantities in the fuel. However, this limits the application of SOFC only to stationary applications due to the long period needed to reach this temperature range. A high temperature operation is also not ideal in terms of cost reduction and long-term stability of the cell components. Hence, lowering the operating temperature of SOFC is crucial for reduction of cost production and commercialization, which enables SOFC to have a wider range of application areas inclusive of portable and mobile ones. Building a high-performance SOFC with small volume is essential as the underlying criteria for these small-scale portable applications. Therefore, careful design and fabrication methods of SOFC operating on intermediate temperatures with high power outputs need to be considered. The intermediate temperature operation of the fuel cell not only increases the overall lifespan of cell but also allows for longer operation with a lower degradation rate compared to high temperature operation. Furthermore, a modified intermediate temperature stack design can accommodate a wider range of applications compared to the tubular and planar stack designs. This paper reviews the development of SOFC stack designs aimed at intermediate temperature operation towards achieving high performance and the benefits of each design.

Keywords: Solid oxide fuel cell; stack design; intermediate temperature

ABSTRAK

Sel fuel oksida pepejal (SOFC) mempunyai kelebihan yang ketara kerana beroperasi secara senyap dan bersih serta dalam masa yang sama mempunyai kecekapan yang tinggi untuk mempertingkatkan reaksi tenaga kinetik pada suhu operasi yang tinggi. Suhu operasi tinggi bagi SOFC adalah secara amnya dalam lingkungan 800 – 1000 °C di mana membantu dalam pembaharuan dalaman kepada hidro karbon dan mengetepikan unsur kesan bendasing yang kecil dalam bahan bakar. Walau bagaimanapun, aplikasi SOFC menjadi terhad kepada aplikasi pegun kerana masa yang lama diperlukannya untuk mencapai suhu ini. Suhu operasi yang tinggi ini juga bukan lah sesuatu yang ideal dari segi pengurangan kos dan kestabilan dalam masa yang panjang untuk komponen. Oleh itu, merendahkan suhu operasi pada SOFC adalah penting untuk pengurangan kos pengeluaran dan pengkomersialan, di mana dapat membolehkan SOFC mempunyai aplikasi yang lebih meluas termasuklah dalam aplikasi mudah alih. Pembinaan SOFC yang mempunyai prestasi tinggi dalam kadar yang kecil adalah penting sebagai kriteria asas untuk aplikasi mudah alih berskala kecil. Oleh itu, reka bentuk dan cara fabrikasi SOFC yang teliti pada suhu pertengahan dengan penghasilan berkuasa tinggi perlu dipertimbangkan. Suhu operasi pertengahan kepada sel fuel bukan hanya meningkatkan jangka hayat keseluruhan sel tetapi juga membenarkan untuk operasi lebih panjang dengan kadar degradasi yang lebih rendah berbanding pada suhu operasi yang tinggi. Tambahan pula, reka bentuk jenis timbunan untuk suhu pertengahan yang telah diubah suai boleh menampung aplikasi yang lebih luas dan pelbagai berbanding reka bentuk jenis tiub dan planar. Artikel ini mengulas semula pembangunan SOFC dalam reka bentuk jenis timbunan yang menyasarkan pada suhu operasi pertengahan dimana bertujuan untuk mencapai prestasi yang tinggi dan kelebihan pada setiap reka bentuk.

Kata kunci: Sel fuel oksida pepejal; reka bentuk tindanan; suhu sederhana

INTRODUCTION

The development in solid oxide fuel cell (SOFC) technology is widely known in the area of energy conversion, with applications involving energy conversion from chemical to electrical (Bierschenk et al. 2011; Graves et al. 2011; Jensen et al. 2010). Majority of the development on SOFCs have been focused on stationary power generation due to its high operating temperature leading to high efficiency of energy conversion, high scale of production energy and ability to provide co-generation. Various simulations conducted on stationary power plants using SOFC predict that higher flexibilities of fuel consumption are possible to operate the SOFC through the utilization of hydrocarbons, biomass, or coal as the fuel. Furthermore, considerably lower outputs of carbon emissions, pollutants and a significant reduction in water utilization for power generation serve as a major benefit of SOFCs (Cordiner et al. 2007; Kivisaari 2004; Panopoulos et al. 2006; Wachsmann et al. 2012) gave rise to the idea of potential applications in many sectors including base-load power plants, dispatchable generation to offset intermittent renewable power sources, and distributed co-generation of electricity and heat (or cooling). The development of a <1 kW portable electrical generator utilizing liquid hydrocarbon fuels opens the possibility of diversification in portable applications (Zhan and Barnett 2005). SOFCs use fuel to generate electricity but electrolysis operation mode for SOFCs are currently in development to convert excess energy to fuels (Bierschenk 2011; Graves 2015; O'Brien 2007). SOFC operate in electrolysis mode through the conversion of accumulated excess renewable electricity to fuel, vice versa in fuel cell mode (Bierschenk et al. 2011; Graves et al. 2015). The importance of an on and off grid battery storage system will be highlighted when there is a need to even out irregularities in supply and demand with the growth of renewable energy generation in total electrical grid generation capacity (Dunn et al. 2011; Yang et al. 2011). SOFC systems have reached a point where manufactured 100 kW systems are in use for stationary power applications.

However, fabrication cost, structural integrity and operating temperature remain significant challenges in the commercialization of SOFC in a wide array of applications. The fabrication cost is largely considered and determined by the balance of system and the characteristics of the cells where they dictate the operating temperature and SOFC stack design. This paper reviews on the modifications done to SOFC

stack designs that help in achieving intermediate temperatures and the outcome of such modification.

OPERATING TEMPERATURE REGIMES

SOFC systems generally operate in the range of 800-1000°C to provide better efficiency and thermal stability. The primary drive in the attempts to decrease the operating temperature of SOFCs towards intermediate temperatures are to obtain a balance between performance, durability and overall life span of the stack and to minimize the operating and fabrication cost. A brief comparison between high temperature and intermediate temperature SOFCs can be summarized in Table 1.

The relationship between possible industrial commercialization based on applications of SOFCs and other fuel cells with the operating temperature regime of the types of fuel cell can be seen in Figure 1. In view of this diagram, a distinguishment of multiple pairing of products can be observed and the need for decreasing the operating temperature is highly dependent on the stack design principle and market application. The current scene in the usage and applications of SOFC are: (1) a high operating temperature (800-1000°C) for commercial and industrial cogeneration with an combined option of a gas turbine (GT), (2) an intermediate operating temperature (550-700°C) with a focus on micro CHP applications of SOFC with the utilization of cost-effective ferritic steels in the build of the SOFC stack (Steele 1994) and (3) a potential usage of SOFC in portable applications at temperatures below 500°C (Steele 1994).

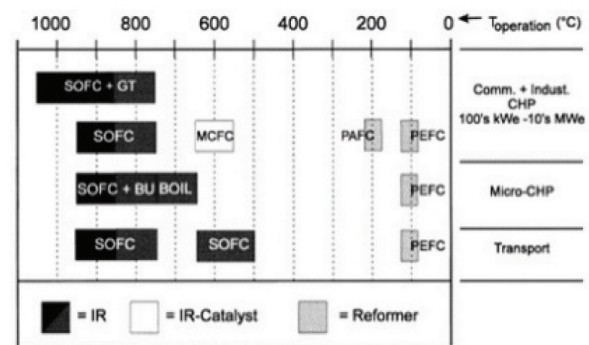


FIGURE 1. Opportunities in industrial commercialization based on applications of SOFCs and other fuel cell types vs. the current operating temperature regime
Huijsmans et al. 1998

TABLE 1. Comparison between High temperature and Intermediate Temperature SOFCs.

Property	High Temperature	Low Temperature	References
Operating Temperature	800-1000°C	550-700°C	Huijsmans et al. 1998
Operating Cost	High	Low	Hacker & Mitsushima 2018
Fabrication Cost	High	Low	Hacker & Mitsushima 2018; Irshad et al. 2016; Wan Yusof et al. 2018
Life Span	Short	Long	Irshad et al. 2016
Suitability for mobile applications	No	Yes	Huijsmans et al. 1998

SOLID OXIDE FUEL CELL (SOFC) PRINCIPLE OPERATION

Unlike the combustion process that produces reaction products and heat through chemical energy conversion, a fuel cell generates electric power with the utilization of chemical potential energy in the form of fuel. Water (H_2O) and carbon dioxide (CO_2) are whereby given as waste products. A single SOFC comprises of a membrane-electrode assembly (MEA) and a current-collection system. Figure 2 shows the operating principle of a SOFC. Oxygen reduces into oxygen ions at the cathode. Then, the oxygen ions diffuse to the anode through the solid oxide electrolyte where the oxidation of fuel occurs. The electrolysis of the fuel at the anode gives off a water byproduct and two electrons that flow via an external electric circuit as electricity. As the electrons travel and re-enter the cathode material, the process repeats. Thus, the electrical power generated through the electrochemical process of fuel oxidization is delivered to the external load.

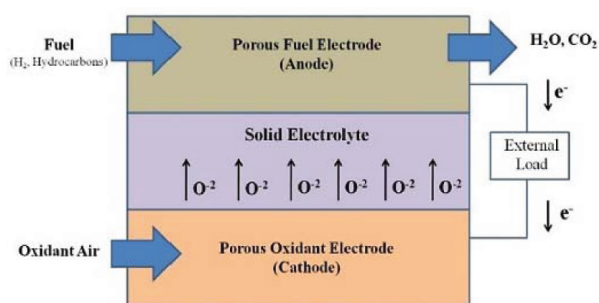


FIGURE 2. Solid Oxide Fuel Cell (SOFC) Schematic and Operating Principle Irshad et al. 2016

SOFC COMPONENT AND FUNDAMENTAL STACK DESIGN

The basic components needed for a SOFC are an electrolyte and electrode. The YSZ electrolyte is the pioneer electrolyte used in SOFC application due to high ionic conductivity properties (Nurul Akidah 2018). An electrolyte with high ionic conductivity is needed to maintain high efficiency and thermal stability. However, YSZ electrolyte shows excellent ionic conductivity in high operating temperature in the range of 800-1000°C. The simplest way to reduce the operating temperature without sacrificing the ionic conductivity is to modify to the anode supported cell design configuration to reduce operating temperatures to intermediate temperature ranges. Anode supported cells have a higher likelihood to

experience redox cycling due to the employment of thin electrolyte layers in the cell (Sarantaridis et al. 2008; Dikwal et al. 2008; Hatae et al. 2010; Vedaari et al. 2010). Besides that, Scandia stabilized zirconia (ScSZ) or magnesium doped lanthanum gallate (LGSM) would serve as a suitable replacement for YSZ to operate at intermediate temperatures due to their relatively high ionic conductivity, but studies show that transitional phase shift in ScSZ to rhombohedral from cubic can lead to a significant reduction in ionic conductivity below 650°C (Lange et al. 1986; Dougherty et al. 1995). Previous studies also show that LGSM reacts with the anode material, Nickel Oxide (NiO), causing a La-Ni phase formation that would deactivate the anode (Huang and Goodenough, 2001; Huang and Goodenough, 2000). Stability issues of ScSZ can be avoided with the introduction of alumina or ceria in small quantities (Wang et al. 2005; Lee et al. 2005; Qadri et al. 1990; Varanasi et al. 2005). SOFCs that generally operate below or around 500°C are called as Low-Temperature SOFCs (LT-SOFCs), often utilizing Gd/Sm doped ceria as an electrolyte material. However, doping electrolytes with ceria may cause complications during the sintering process and adversely affect electronic conductivity causing a power loss due to short-circuiting of the cell (Mogensen et al. 2000; Timurkutluk et al. 2011).

Cell structures can also be modified with different selections of materials and arrangement of cells to suit applications based on temperature regime. In Figure 4, two standard SOFC designs shown to be either tubular or planar. In Table 1, listed a comparison can be seen between tubular and planar designs of SOFC. SOFC tubular designs are more popular whereby although fabrication is difficult and has a lower power density as compared to the planar type, both gas sealing and interconnection application of single cells during stack manufacturing can be achieved easily. The planar design structure however not only makes fabrication simpler but displays higher power density

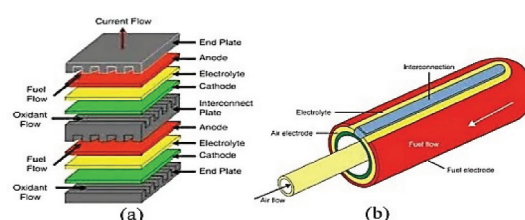


FIGURE 3. Schematic of a planar (a) and a tubular SOFC (b) Ramadhani 2017

TABLE 2. Comparison between Tubular and Planar Designs

Property	Planar Design	Tubular Design	References
Power density	High	Low	Singhal 2004; Nagel et al. 2008; Ryu T 2007
Volumetric power density	High	Low	Pi et al. 2014; Yang et al. 2013; Suzuki et al. 2008
High temperature sealing	Difficult	Easy	Sammes et al. 2005 ; Du & Sammes 2004
Startup duration	High	Low	Kendall 1998; Nguyen et al. 2004)
Fabrication cost	Low	High	Kendall 1998; Nguyen et al. 2004
Thermal cycling stability	Low	High	Yang et al. 2013

readings. Furthermore, the tubular design cannot achieve higher power density than planar designs due to its structural disadvantage of current collection path.

MODIFICATION OF CELL DESIGNS FOR SOFC INTERMEDIATE TEMPERATURES

CONE SHAPED CELL SYSTEM

The cone-shaped stack principle is based off a segmented cell concept where the flow of gas and electricity is connected in series. The connected single cells are lined up to follow the segmented cell design where this can provide high thermal stability. Stacking the one cone on top of the other forms a modified self-supporting tubular structure. Furthermore, the interconnectors act as both a seal and current collector. This design makes it possible to achieve a flexible and compact stack structure with a higher power. Therefore, lightweight and flexible stacks with cone-shaped design for portable applications can be achieved. On the other hand, the interconnector integrity and fabrication methods pose as an issue due to geometric complexity. Figure 4 displays a 3-cell cone-shaped stack design (Sui J 2007). Sui and Liu first introduced the Cone-shaped design SOFC with the fabrication and testing of a 3-cell stack (Sui J 2007; Liu 2013). The cell comprised of a 2 cm long slipcast cone-shaped 8 mol % Ytria-stabilized zirconia (8YSZ) electrolyte support, LSCF/GDC cathode on the outside and NiO/GDC on the inside of the cone-shaped electrolyte support. The cone-shape Samarium doped Ceria (SDC) electrolyte supported cell has an active area of 0.85 cm² which allowed it to provide 300 mWcm² peak power density at 700 °C. However, the usage of SDC as the electrolyte can limit its use in a SOFC stack due to its rather poor mechanical ability (Sui 2012). Another cone-shaped SOFC was fabricated with Gadolinium Doped Ceria (GDC) electrolyte support by Jing et al. This support was produced through the slipcasting technique and measured 0.85 cm in diameter, 1 cm in length and 0.026 cm thickness after sintering for 4 hours at 1400 °C. At a temperature of 700 °C, the low ohmic resistance measured through the employment of the thick electrolyte was able to generate the highest

power density value at around 300 mWcm⁻². Zhang et al. were able to show significant cell performance improvement by employing the cone-shaped anode supported structure (Zhang et al. 2008).

Ding et al. (2011a) studied the overall electrochemical performance and stability under various densities with a NiO/YSZ cone-shaped anode supported SOFC under direct operation with methane. Results showed that under the presence of high current density, the anode was able to function safely due to the presence of miniaturized coking formed on the anode after the operation with methane at 700 °C. They also performed tests on the durability of the cone-shaped anode-supported cells under direct methane fuel with a current density of 0.4 W cm⁻² and temperature of 600 °C for 140 hours (Ding 2011b). The fabrication of a 11-cell stack with cone-shaped NiO/YSZ anode-supported cells was achieved by Bai et al. The 11-cell stack managed to display the highest volumetric power density at 0.9 W cm⁻³ during operation under methane fuel (Bai 2010). Further analysis of the microstructure of the stack showed that varying amounts of coking were present at multiple positions. Ding et al. (2011c) and Liu et al. (2013) performed tests on the cone-shaped SOFC under different fuel: primarily dimethyl ether (DME) and carbon monoxide fuels where the utilization of DME as a fuel instead of hydrogen caused a performance drop of 30 % at 600 °C.

A summary of studies for cone-shaped SOFCs are shown in Table 3 and classified by cell materials, fabrication techniques, and test results under different fuels.

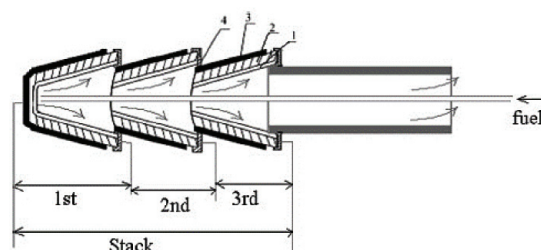


FIGURE 4. Cone Shaped 3-cell Stack elements: 1. Anode, 2. Electrolyte, 3. Cathode, 4. Interconnect
Sui 2007

TABLE 3. Summary of Coned Shaped SOFC Designs and Performance Study Results.

Ref.	Cell Materials			Technique: Operational cell support	Operational Temp (°C)	Type of Fuel	Maximum power density (mWcm ⁻²)
	Electrolyte	Anode	Cathode				
Sui 2007	Ce _{0.8} Sm _{0.2} O _{1.9}	NiO-Ce _{0.8} Sm _{0.2} O _{1.9}	Sm _{0.5} Sr _{0.5} CoO ₃	Slipcasting	700	Humidified Hydrogen	297
Sui 2012	Ce _{0.9} Sm _{0.1} O _{1.95}	NiO-Ce _{0.9} Sm _{0.1} O _{1.95}	Sm _{0.5} Sr _{0.5} CoO ₃	Slipcasting	700	Humidified Hydrogen	300
Zhan 2008	8YSZ	NiO-8YSZ	LSM-YSZ	Slipcasting	700	Humidified Hydrogen	140
Ding 2011b	Gd _{0.1} Ce _{0.9} O _{1.95}	NiO-Gd _{0.1} Ce _{0.9} O _{1.95}	La _{0.6} Sr _{0.4} Co _{0.8} Fe _{0.2} O ₃ -Gd _{0.1} Ce _{0.9} O _{1.95}	Slipcasting	600	Humidified Methane	414
Bai 2010	Gd _{0.1} Ce _{0.9} O _{1.95}	NiO-Gd _{0.1} Ce _{0.9} O _{1.95}	La _{0.6} Sr _{0.4} Co _{0.8} Fe _{0.2} O ₃ -Gd _{0.1} Ce _{0.9} O _{1.95}	Slipcasting	600	Dimethyl Ether	350

HONEYCOMB SOFC STRUCTURE

In 1992, the honeycomb SOFC structure was invented and patented and was described by Wetzko et al. (Wetzko 1999) as a “condensed-tubes” like structure. The honeycomb design offers the best of both worlds where it borrows certain advantages of the tubular and planar SOFC designs. In brief, honeycomb designs benefit with high physical property values in terms of mechanical strength and durability and also possess high volumetric power density. Figure 5 shows the structure of a honeycomb SOFC where its channels are used as fuel and oxidant routes. The honeycomb structure can be supported by either electrolyte or electrode depending on the configuration. Main issues of this design that need to be looked into are the interconnection and the electrical lead (Kendall K 2010; Yang 2008; Droushiotis 2009). Yamaguchi et al. used quadratic and circular structures to develop cathode supported honeycomb SOFCs. This not only reduced the ohmic losses by employing a thin electrolyte but also improved the total volumetric power density (Yamaguchi et al. 2008; Yamaguchi et al. 2009a; Yamaguchi et al. 2009b; Yamaguchi et al. 2009c).

The LSM cathode honeycomb support was fabricated through extrusion cell used extrusion and employed a 10 mol% Sc_2O_3 stabilized ZrO_2 (10ScSZ) electrolyte and NiO/GDC anode. Upon testing, results show that cells exhibit high volumetric power density above 1 Wcm^{-3} at 600°C and 2 Wcm^{-3} at 650°C under hydrogen fuel respectively (Yamaguchi et al. 2008).

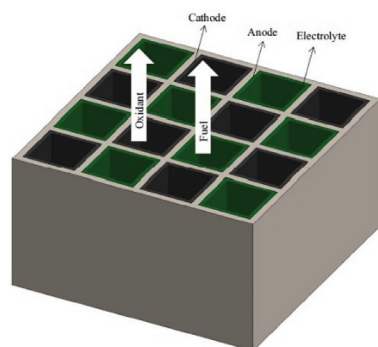


FIGURE 5. Honeycomb SOFC design
Wetzko 1999

A summary of studies for honeycomb SOFCs are shown in Table 4 and classified by cell materials, fabrication techniques, and test results under different fuels.

MICRO-TUBULAR SOFC DESIGN

Micro-tubular SOFCs are designed in such a way that they will undergo a reduction in terms of the tube diameter until it reaches the millimeter scale. The two benefits of this modification are a relatively high-power density and high resistance against thermal shock which causes the heating of the micro-tubes to the operating temperature without experiencing any damage. These advantages are suitable for mobile applications due to being able to startup rapidly along with a high-power density. Poor current collection for this stack design is mainly due to the small diameter of the tubes and can be viewed as one of the disadvantages of this design.

In the early 90s, the micro-tubular SOFC concept was the brainchild of Kendall (Kendall K 2010). Yang et al. (2008) were considered to be one of the very first few to publish a paper regarding a SOFC structure with hollow fibers. (Droushiotis et al. 2009), with the help of a single step via co-extrusion process, were able to fabricate hollow fibers with a dual layer design in a phase inversion and showed that there exists a relationship between the extrusion rate and the thickness of the electrolyte layer (Droushiotis et al. 2009). The microstructural, electrical and mechanical properties of NiO/8YSZ hollow fibers were studied based on the effects of the initial NiO content. When the porosity and electrical conductivity experienced an increment proportionate to the NiO content, the bending strength of the fiber structure decreased. (Yang et al. 2008 ;Othman MHD 2010) evidenced that one of the strong functions of sintering temperature is electrical conductivity. Consequently, they successfully manufactured a dual-layer hollow fiber of 10 μm thickness, dense GDC electrolyte with an extrusion rate of 0.5 ml min^{-1} was successfully manufactured (Meng XX 2015). Although this fiber exhibited low values in terms of mechanical strength and gas tightness property among all the cases, at 600°C , the electrochemical performance displayed was the highest at 1.11 W cm^{-2} . Through the usage of the single step phase

TABLE 4. Summary of Honeycomb SOFC Designs and Performance study results.

Ref.	Cell Materials			Technique: cell support	Operational Temp ($^\circ\text{C}$)	Type of Fuel	Maximum power density (zWcm^{-2})
	Electrolyte	Anode	Cathode				
Yamaguchi 2008	ScSZ	NiO-GDC	LaSrMnO3	Extruding	600-650	Humidified Hydrogen	1 to 2
Yamaguchi 2009b	ScSZ	NiO-GDC	(La _{0.8} Sr _{0.2}) _{0.97} MnO3	Extruding	600	Humidified Hydrogen	0.3
Yamaguchi 2009c	ScSZ	NiO-GDC	LaSrMnO3	Extruding	600	Humidified Hydrogen	1.2
Shimizu 2009	ScSZ	NiO-GDC	LaSrMnO3	Extruding	600	Hydrogen	2
Shimizu 2007	Sc _{0.9} Ce _{0.1} ZrO ₂	NiO-GDC	(La _{0.8} Sr _{0.2}) _{0.97} MnO3	Extruding	600	Humidified Hydrogen	0.67

TABLE 5. Summary of Micro-tubular SOFC Designs and Performance Study Results.

Ref.	Cell Materials			Technique: cell support	Operational Temp (°C)	Type of Fuel	Maximum power density (Wcm ⁻²)
	Electrolyte	Anode	Cathode				
(Yang et al. 2008)	GDC	NiO-GDC	LSCF-GDC	Extruding	600	Humidified Hydrogen	1.11
(Meng et al. 2015)	GDC	NiO-GDC (17-33µm)	LSCF-GDC	Extruding	600	Hydrogen	1.21

inversion assisted co-extrusion technique, an electrolyte-anode functional layer-anode triple-layer hollow fibers for microtubular SOFCs was successfully fabricated by (Li T et al 2015; Meng XX 2015). The thickness of the anode functional layer ranging between 17-33 µm was found to show significant improvements in terms of cell performance. It achieved its highest performance which peaked at 1.21 W cm⁻² at 600 °C with the utilization of pure hydrogen fuel. Table 5 summarizes the studies done by researchers which include the cell materials, fabrication techniques and test results of the micro-tubular SOFCs.

CONCLUSION

This review paper shows the evidence that changing the structural design of a SOFC is useful in reducing the operating temperature to intermediate temperature regimes. Furthermore, changing its design can also offer many other benefits such as for the case of micro-tubular SOFCs, high-power density and effective shock resistance against thermal shock which allow the small tubes to reach to the optimum operating temperature through heating without any damage. Other challenges that might be faced by the stack designs discussed in this paper can be found in Table 6.

TABLE 6. Summary of advantages and challenges of stack design.

Stack Design	Advantages	Challenges
Cone-shaped	Improved performance with high thermal shock resistance and high-power output.	Fabrication issues due to complex design and interconnector integrity problems
Honeycomb	High durability and mechanical strength with volumetric power density.	Fabrication issues due to complex design, interconnector integrity problems and electrical lead.
Microtubular	High volumetric power density, output and thermal shock resistance with rapid start up ability	Poor current collection

A stack design that is too complex can cause fabrication problems and incur high manufacturing costs. Other factors that can influence the performance are the ionic conductivity of an electrolyte and type of fuel used. The microtubular design shows much promise and it can be certainly be improved with further research into solving its

current collection issue. Overall a higher performance in terms of power density and other benefits as seen in Table 6 make employing the microtubular cell design a better option compared to the other designs. Furthermore, usage of additional materials such as ferritic steel plates would also help in bringing down the operating temperature to intermediate ranges and further help to increase its operating period and lifespan of the cell. Thus, reducing the operating is certainly possible with modifications done to the stack but coupling this another measure to lower the temperature would benefit greatly in future development of SOFCs toward low temperatures regimes.

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