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Review

Review of the micro-tubular solid oxide fuel cell Part I. Stack design issues and research activities

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ABSTRACT

Fuel cells are devices that convert chemical energy in hydrogen enriched fuels into electricity electrochemically. Micro-tubular solid oxide fuel cells (MT-SOFCs), the type pioneered by K. Kendall in the early 1990s, are a variety of SOFCs that are on the scale of millimetres compared to their much larger SOFC relatives that are typically on the scale of tens of centimetres. The main advantage of the MT-SOFC, over its larger predecessor, is that it is smaller in size and is more suitable for rapid start up. This may allow the SOFC to be used in devices such as auxiliary power units, automotive power supplies, mobile electricity generators and battery re-chargers.

The following paper is Part I of a two part series. Part I will introduce the reader to the MT-SOFC stack and its applications, indicating who is researching what in this field and also specifically investigate the design issues related to multi-cell reactor systems called stacks. Part II will review in detail the combinations of materials and methods used to produce the electrodes and electrolytes of MT-SOFC's. Also the role of modelling and validation techniques used in the design and improvement of the electrodes and electrolytes will be investigated. A broad range of scientific and engineering disciplines are involved in a stack design. Scientific and engineering content has been discussed in the areas of thermal-self-sustainability and efficiency, sealing technologies, manifold design, electrical connections and cell performance optimisation.

Contents

1. Introduction to SOFCs

Fuel cells are electrochemical devices that convert chemical energy in hydrogen enriched fuels into electricity electrochemically. In the most widely used energy extraction mechanisms of today, such as engines and hydrocarbon fuelled turbines etc., a process of producing heat and converting this heat energy into mechanical work results in thermodynamic limitations such as Carnot efficiency limitations. During the combustion process pollutants are also exhausted and these pollutants add to global warming and are harmful to human health.

In the future fuel cells will be a means of producing electricity more efficiently and they have the potential of producing envi-

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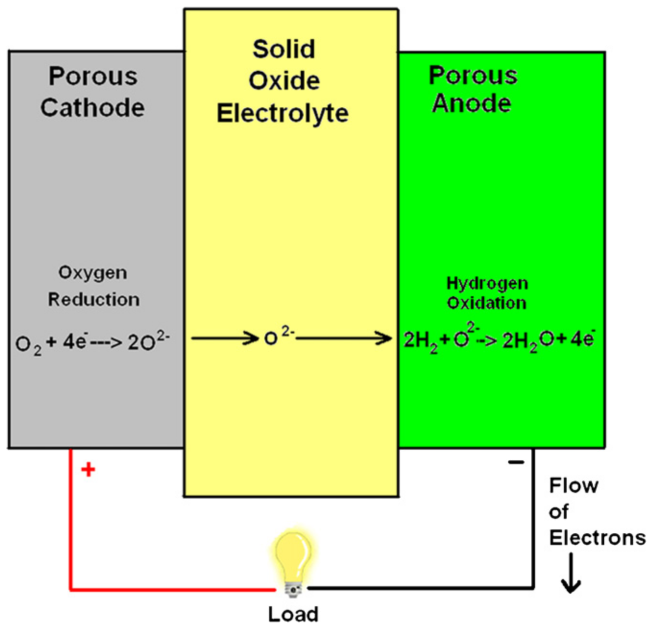

Fig. 1. Operation of a SOFC where three phase conditions in the cathode and anode allow the red–ox reactions.

ronmentally friendly energy with much lower pollutant and CO_2 emissions. One particular type of fuel cell is the solid oxide fuel cell (SOFC). It has some important characteristics that other types of fuel cells cannot provide. Within a fuel cell, hydrogen rich fuel is fed to an oxidising anode electrode and an oxidant, air or pure oxygen, is fed to a reducing cathode electrode. Sandwiched between the anode and cathode is an ion conducting device called an electrolyte that electrically isolates the anode from the cathode. Fig. 1 depicts an illustration of this mechanism.

An important concept in the operation of the SOFC is the triple phase boundary regions, which are places where the oxygen reduction and hydrogen oxidation (red–ox) can only take place. More specifically these are areas where oxygen ions being transported through the electrolyte meet hydrogen atoms flowing through pores in the anode. At these points there should also be a nickel conduction path to an external circuit. Thus oxidation can occur on the anode forming water and releasing electrons that can be used for work in an external circuit. These electrons then flow back to the cathode to facilitate reduction of oxygen, producing the ions for transportation through the electrolyte.

These electrons are driven by the voltage generated between the negative and positive electrodes. This voltage is a function of the Nernst equation, which provides a relationship between the ideal standard potential for a cell reaction and the ideal equilibrium potential at other partial pressures of reactants and products. For the overall cell reaction, the cell potential increases with an increase in the partial pressure, gas concentration of the reactants and/or a decrease in the partial pressure of products. A more detailed introduction to the inner workings of the SOFC can be found in Part II of this review series.

The three main advantages of the SOFC over many other types of fuel cells are firstly, that losses associated with the cells that result in heat can be integrated into the heat provision system for the SOFC and can make it thermally self sustaining or at least very efficient once the cells are operational. The second advantage of the SOFC is that at these elevated temperatures internal fuel reforming can also take place on nickel in the anode. The internal reforming capability of SOFCs means that a variety of fuels such as hydrocarbons can be used without using external reformers. These devices require power themselves to run and they add mass, which reduces the overall efficiency of a fuel cell power plant/pack. The third advantage is that the materials to make good quality cells are not as expensive as those for other types of fuel cells.

2. The micro-tubular SOFC stack and possible applications

The demonstration in the early 1990s of the extrusion of thin YSZ ceramic tubes, with wall thicknesses of between 100 and 200 μm, which could be used as the electrolyte for SOFC operation, by Singhal and Kendall [1], opened up the micro-tubular SOFC (MT-SOFC) research field. Soon after this achievement Kendall and his group [2] demonstrated the first operational 20-cell, 200-cell and 1000-cell reactors. Described by Alston et al. [2] is the 1000-cell reactor which was used to produce electricity from the cells directly and to heat water with the heat losses. The reactor is made up of 40 modular stacks that are joined together to make a master stack. They reported that the hot air alone fed to the reactor was sufficient to heat up the cells to their operating temperature and that these cells could withstand temperature increases of 200 °C min^{-1}. The reactor produced only 82 mW cm^{-2} but this achievement proved that the MT-SOFC system was feasible.

Currently the most commonly used cell architecture for the MT-SOFC is a deviation from the original electrolyte supported type called the anode supported MT-SOFC. To reduce the ohmic losses and impedance to ionic transport, a thin electrolyte layer is spread upon the anode. In this case the anode is much thicker than the electrolyte and it is the support body for the electrolyte and cathode. This form of cell was also pioneered by a group including Kendall [3]. More detail about the cell development and the methods used to improve the electrode and electrolyte structures while also improving the power output of the cells will be supplied in the Part II of this review. Cutting edge experimental techniques such as impedance spectroscopy, X-ray computed tomography in combination with modelling techniques, oxygen stable isotope labeling and subsequent secondary ion mass spectrometry analyses are some examples of the novel approaches for improving the cell performance that will also be discussed in detail in Part II.

In 2000 a paper by Van Herle et al. [4], investigated the application of the SOFC for vehicles. It was suggested by the authors (which included esteemed members of the field) that the over-looking and presumption, at the time, that SOFC technology could not meet the requirements needed by a vehicle power plant, i.e. a massive reduction in size, enhanced strength and resistance to fast thermal heat up, may be short-sighted and incorrect. They recommended that cells based on thin small tubular ceramics had the potential to overcome the problems listed above and that the inherent advantages of the SOFC over engines and other types of fuel cells (mainly economic) made the MT-SOFC very attractive for this scale of power plant.

Several designs for the use of SOFCs for automotive use have since been suggested for example by Winkler and Lorenz [5]. Their suggestions include a SOFC-GT (gas turbine) power train system, a SOFC-GT power train system with external cooling and a SOFC–Stirling cycle. The authors specifically point to the MT-SOFCs as the most feasible type of SOFC for mobile applications coupled with a Stirling engine because of the expected high system efficiency, low specific volumes and the present availability of small units.

The heat extraction of the SOFC by a Stirling engine does not affect the combustion reaction because the coolant is the separated working fluid of the Stirling engine. They also recommend that the exhaust temperatures of the SOFC–Stirling engine should be much lower than that of a SOFC-GT system indicating better heat recovery. These initial studies indicated that the design problems of such cycles should be solvable. The micro-GT is a very simple device compared to the Stirling engine and the inclusion, for example, of turbo-chargers in the exhaust flow of the Stirling engine system, to increase its efficiency, also increases its complexity. However, the authors pointed out that more research in this area could be valuable in the long term.

Tompsett et al. [6] in another application for MT-SOFCs describe the integration of a micro-SOFC stack and standard gas burner to produce both electricity and heat by putting a cell inside a three-catalyst burner. Fig. 2 illustrates the concept where the platinum combustion catalyst provides heat (900 °C) for the operation of the MT-SOFCs and the pre-heating of the ruthenium pre-reforming catalyst to 700 °C. The catalytic reformer produces hydrogen gas for the fuel cell operation and the mixing of some air into the fuel gas prevents carbon build up on the fuel cell anodes, by oxidising the carbon. Finally the air that is utilised in the fuel cells is fed back to the pre-burner fuel mix to preheat the fuel giving a faster reaction time.

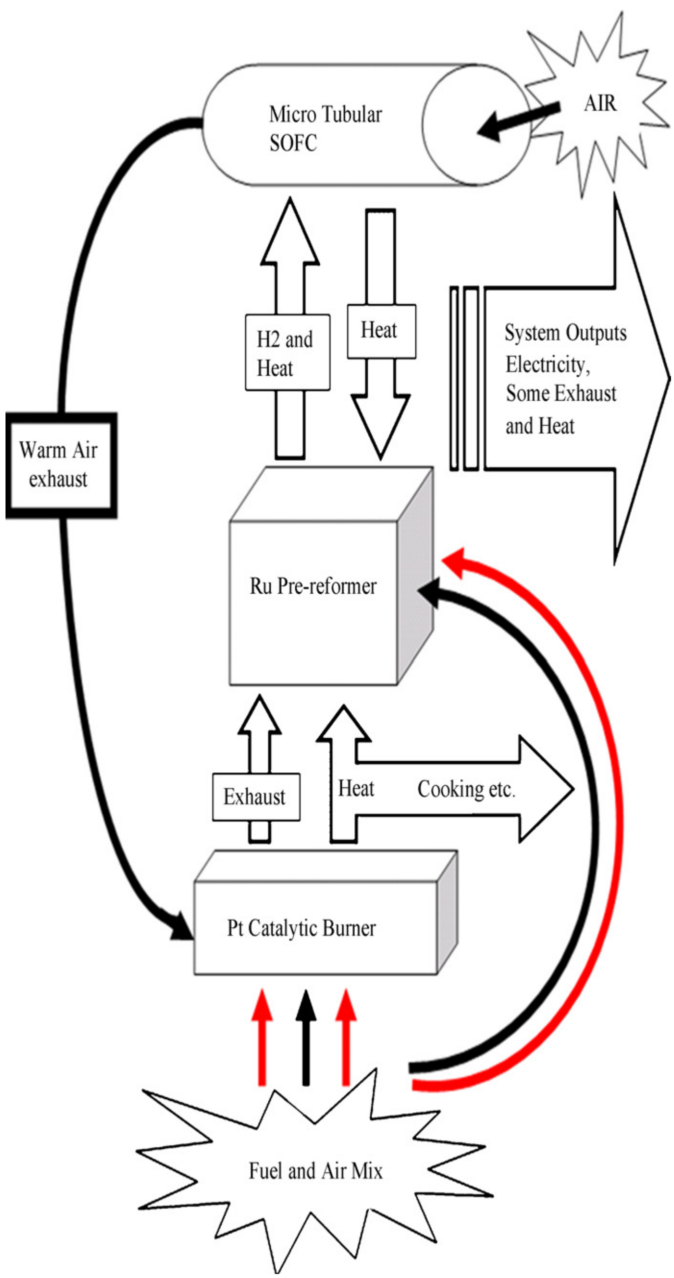

Fig. 2. Depiction of the gas and heat flows in a triple layer catalyst–SOFC-catalyst system proposed by Tompsett et al. [6].

This prototype unit produced a very meager 1.5 W, but the demonstration alone showed the versatility of the MT-SOFC stack. The authors also pointed out other possible applications such as a vehicle battery charger. In this case the fuel cell could be used to recharge a battery and the battery life would be extended due to smaller discharge depth, ultimately reducing the costs of the number of replacements. The usable time of the vehicle would be increased due to the continuous recharging from the fuel cell, when in use. They also point out how a power pack from a MT-SOFC would not intrude on such products as golf trolleys, bicycles and wheel chairs.

Instantaneous start-up, similar to that of an engine or battery, is an unavoidable condition, for the commercial success of MT-SOFCs, not only in the automotive field but also in the auxiliary power supply sector. Systems using battery and super capacitor start-up heating mechanisms have been envisaged for the start up of the MT-SOFC stacks, but the volume occupied by such a system, that generates an equivalent power, is prohibitive for many applications. Originally Kendall and co-workers [2] demonstrated with his 1000-cell reactor that MT-SOFCs could be heated up to their operational temperature of 850 °C at a rate of 200 °C min^{-1} and other authors have since shown much faster heat up times.

In 1996 Prof. Kendall also set up the first MT-SOFC company called Adelan (UK) Ltd. and Bujalski et al. [7] with these cells have shown that MT-SOFCs could be consistently ramped to operational temperature at a rate of 4000 °C min^{-1} without any structural damage or power degradation. This would mean that the cells would be at their operational temperature after 12 s. An important result of these tests was the finding that these cells could also be allowed to cool extremely fast to 300 °C within a few seconds and not become damaged or in cur losses of performance after a 50 cycle test. Of course it should be noted that the apparatus used to perform these tests was specifically designed for single cell tests and a stack of these cells will have its heating up and cooling down rates further limited by the thermal stresses induced by the reactor housing and seals of the cells. Nonetheless it has been shown that the cells themselves can be very thermal shock resistant.

Very recently Du et al. [8] showed with their cells (Nanodynamics USA, Inc.) that regardless of single cell tests or stack tests that thermal cycling of their cells did not cause any degradation of them. They recommended that their stacks can be heated up at rates of 550 °C min^{-1} and that their cells produced the normal operational power as soon as it reached the operational temperature of 800 °C. It was very interesting that their single cell tests just comprised of putting the cells into preheated ovens for heating up and simply removing them for cooling down. This is another example of how robust these cells can be. Du et al. [8] have also performed experiments where they heated up different generations of their cells in stacks. In these experiments the start-up heating rate was 550 °C min^{-1}. They found that no performance losses or structural damage occurred even though the cell size was scaled up 2.7 times. The sizes of these cells were not found in any of the published literature associated with the company. However, this is a demonstration the robustness of stacks containing MT-SOFCs.

3. Reports of micro-tubular SOFC stack designs and considerations

In the literature there are only a few design concepts described for MT-SOFC stack design. The most standard design is an array of MT-SOFCs with hydrogen flowing inside each cell in the array and air flowing around the outside of the cells. The lack of stack designs and optimisation of the arrays in the literature is probably for two reasons. The first being that the problems associated with single cells have not been totally eradicated by most groups. Thus they will be focusing their efforts on single cells. The sec-

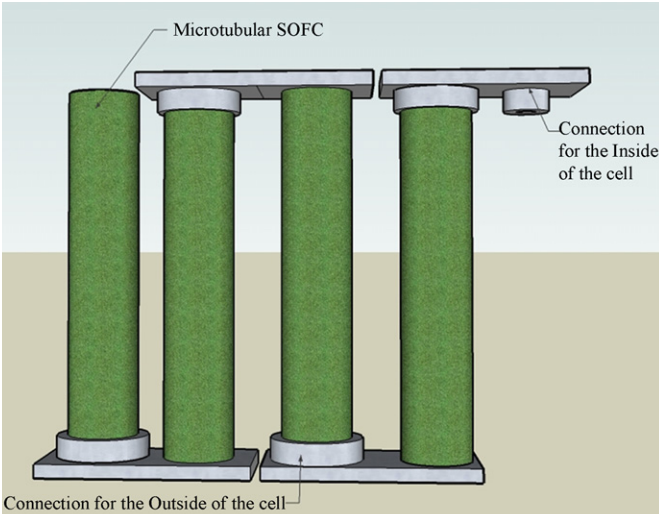

Fig. 3. Depiction of the current collectors and cell configuration by Sammes et al. [9].

ond reason is that groups associated with industrial partners will be guarding their stack designs carefully to keep their innovations hidden from their competitors. However, there are some interesting reactor designs described in both the literature and patents for MT-SOFCs.

To begin with Sammes et al. [9] show a concept for a 40-cell stack where the cells are connected together with current collectors in planar multi-cell arrays. The main feature of this design is that these single rows of tubular cells can be electrically connected in series or parallel and physically connected as parts of a larger stack comprised of these individual modules. Silver is used to braze the cells to the current collectors and the brazing process takes place inside a furnace. Fig. 3 illustrates an example of the current collection and sealing mechanism for serially connected cells where the terminal on the left of the flat plate connector contacts the cathode and the terminal on the right to the anode providing a serial connection that builds up the voltage.

Funahashi et al. [10] describe the fabrication of a cube shaped MT-SOFC bundle that can be seen in Fig. 4. The reactor components were successfully assembled into a cube shaped MT-SOFC bundle (3 cm × 3 cm ×3 cm) with 36 tubular cells each having a diameter of

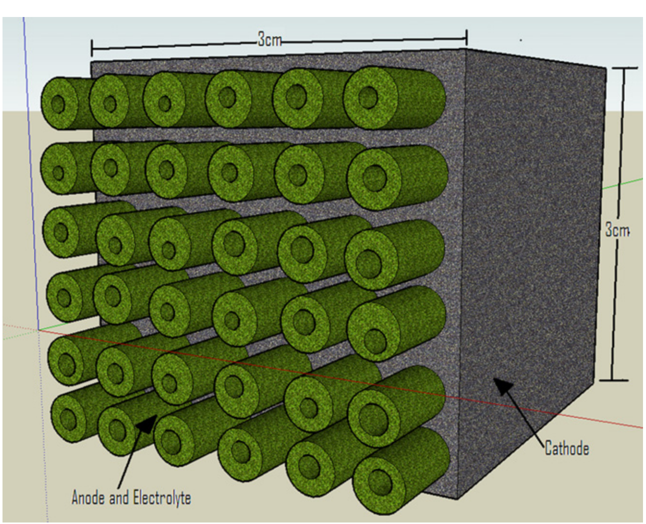

Fig. 4. Illustration of the cube shaped SOFC cathode structure with embedded electrolyte-anode tubes by Funahashi et al. [10].

2.0 mm. In this type of MT-SOFC stack design, the cathode consists of blocks of matrices with several grooves that hold the MT-SOFCs. The cathode matrices are used as the current collectors, the guide for cell arrangement and the oxidant flow path. This original design is also an example of a stack where cells are connected in parallel and recently the same group [11] have shown an innovative technique to connect these cells in series.

They recommend that when the cathode matrix has a gas penetration coefficient of 1.68×10^{-4} ml cm^{-2} s^{-1} Pa^{-1}, it is possible to flow 600 ml min^{-1} of air in a cubic cell (1 cm^3) under 0.1 MPa difference between the inlet and outlet. The air flow rate of 470 ml min^{-1} was estimated to meet the target power density of 2 W cm^{-3}, and indeed these cathode matrices appeared to be suitable components for the cube to achieve targeted performance.

The flow of fuel to the anode is also an important area to be optimised in a MT-SOFC reactor design. Currently most groups send the fuel in one end of the micro-tubular cell anode and out the other end. The electrical connections are made via wires lodged inside the anode, meshes lodged inside or brazed connections to areas of the anode not covered by the electrolyte. Recently Lee et al. [12] have demonstrated their 700 W anode supported SOFC stack for APU applications. It has a novel combined approach of supplying the fuel and electrical connection by using the red conductive inlet pipe and orange wires as seen in Fig. 5. Their cells are 10 mm in diameter and they are 160 mm long, which is at the larger end of the MT-SOFC scale.

A novel feature of this design is that the inclusion of a metallic pipe, nickel wires and layer of nickel felt inside the anode should enhance the fuel utilisation efficiency by reducing the volume of the gas flow channel inside the anode. Importantly this system may simplify the fuel inlet and exhaust manifolds because both the fuel and exhaust enters and leaves the cell at the same end. It may also be the case that the heat transfer from the hot exhaust to the cooler inlet gas in this system may also improve the overall system efficiency. They report a dramatic increase in the power output of their cells from 0.24 up to 0.54 W cm^{-2} with a fuel utilisation of 45%. The reasons suggested for this increase in performance are attributed to this new fuel supply system, a reduction in ohmic losses and a reduction in contact resistance between the anode and the current collector.

A very interesting part of this paper examines the gas supply to a 6-cell stack. They use a computational fluid dynamics program to design their gas supply systems and by varying the size of the gas entrance slit within the fuel supply chamber and the angle of the inclined wall of the fuel exhaustion chamber, they make sure that each cell in the reactor receives the same amount of fuel. The length of the current paths within their stack was also designed so that the length of the current collection wires in hot zones was minimised. This was because of the increased power losses from these wires at higher temperatures. Their final stack containing 36 cells produced a total of 700 W at 750 °C with a fuel utilisation of 49% and 0.39 W cm^{-2}. This method may be applicable to the smaller diameter cells as there are micro-sized capillary pipes available on the market made from pure silver and indeed nickel from various manufacturers.

The prior mentioned stack designs only relate to the physical structure of the reactors and there are some examples of detailed accounts of whole systems that include the key topic of thermal-self-sustainability. For example Alberta Research Council [13] have issued a patent which includes the design for the operation of a MT-SOFC stack and heating system. The system is actually quite simple. The un-reacted fuel is used to supply the required burn temperature from the exhaust to control the temperature inside any of the reactor modules and this is electronically controlled. These burners can also be used to maintain any individual module at a standby temperature.

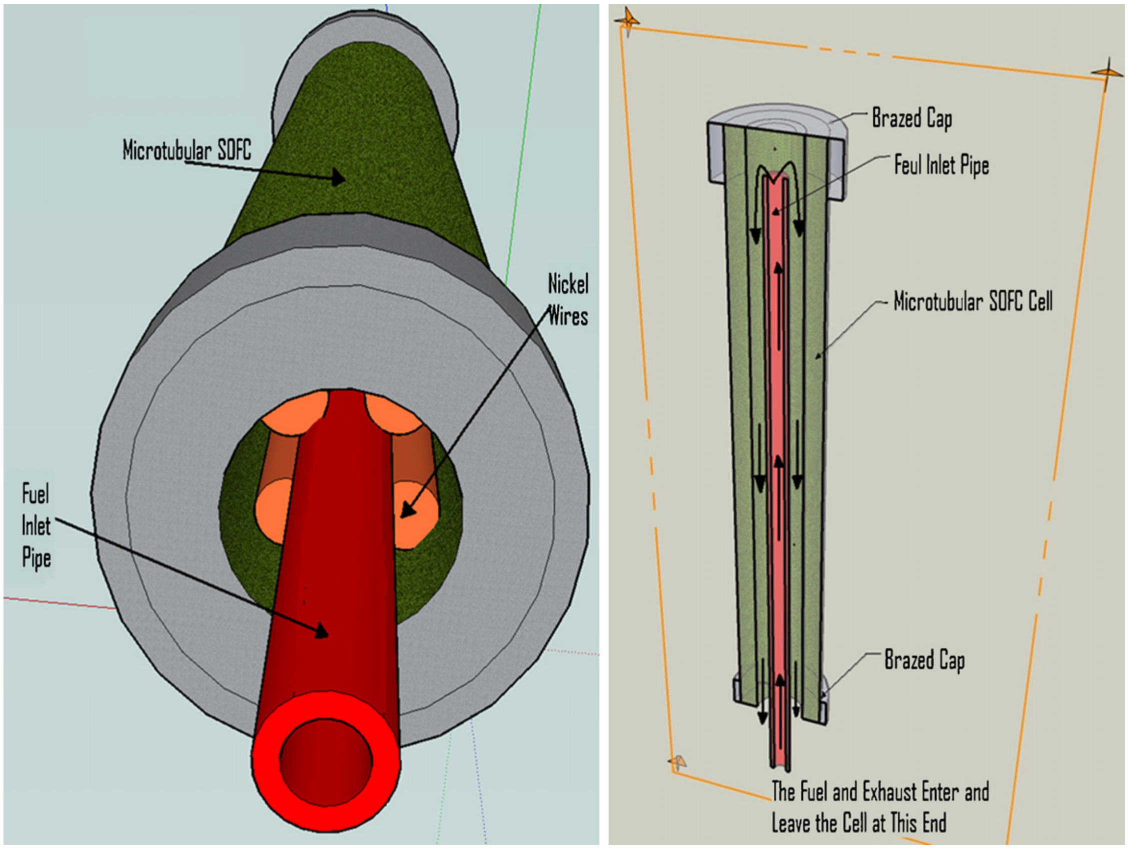

Fig. 5. Illustration of the gas supply system invented by Lee et al. [12].

The invention contains four modules of cells or sub-stacks with different numbers of cells in each, any of which can be activated or deactivated in any combination to satisfy the power demand. Also a novel type of insulation from Aspen Aerogels (USA) Inc. [14] is utilised to thermally insolate the system. This seems to have a very low thermal conductivity, which would be extremely advantageous for holding heat within the modules. In the patent application there is a lot of detail about the workings of the stack and sub-stack systems and the reader is referred to the patent [13] for more information.

Another patent also by Alberta Research Council [15] also describes the operation of a MT-SOFC stack with a novel heat exchanger design as seen in Fig. 6, where it should be noted that the oxidant outlet from the top of the stack is connected to the oxidant outlet in the heat exchanger. The system comprises of a cylindrical array of SOFC cells encased by a heat exchanger that wraps around the fuel cell stack and comprises of a flexible, thermally conductive first layer and an overlapping flexible, thermally conductive second layer spaced from the first layer. The first and second layers define adjacent annular oxidant supply and oxidant exhaust channels when wrapped around the stack. The exhaust channel is communicative with heated exhaust oxidant flowing from the stack. The oxidant supply channel is communicative with supply oxidant that flows to the stack and is heated by hot exhaust oxidant in the adjacent oxidant exhaust channel and from heat radiating from the stack.

This is just a design and there is no performance data available on how well the heat exchanger works but it seems that it could work quite well. Indications of the advantages of similar coiled heat exchangers have been shown elsewhere by [16–19].

Another addition of this system could be to burn the exhaust hydrogen to also heat the incoming oxidant and fuel lines, which should increase the overall efficiency. A big question with this design would be, because the flow all exits at one point inside the MT-SOFC holding chamber, does this cause areas inside the reactor where the inlet gas is not refreshed? A possible solution could be to reverse the direction of the flow and allow the gas to exit at the porous material rather than the single point. A very interesting additional adaptation of this design would be to supply the fuel rather than the oxidant to the cells trough this heat exchanger with the anode layer on the outside of the cells. This may make it much

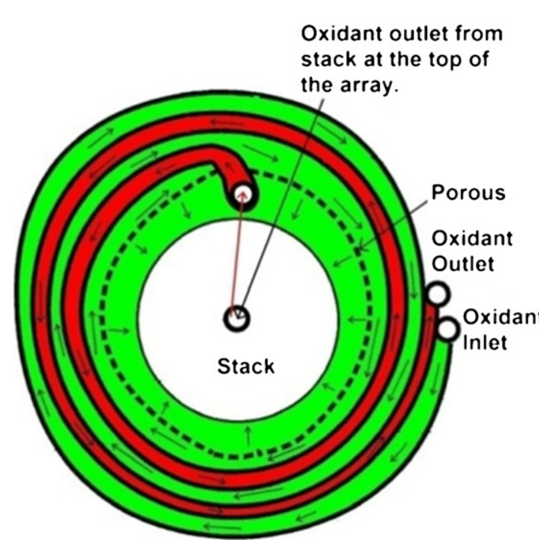

Fig. 6. Looking from the top of the heat exchanger design by Alberta Research Council (CAN) Inc. [15].

easier to make electrical connections to the anode and the fact that the hydrogen would be confined inside this chamber could also be advantageous and a novel approach.

Most of the previous reactor concepts are designed to operate at high temperatures but many groups are working to reduce the operating temperature of the cells. The use of intermediate temperature MT-SOFCs, operating under 650 °C, can decrease material degradation, prolong stack-lifetime and reduce stack material costs by utilizing common metal materials. In the past few years results published by several authors [20–25] have reported promising intermediate temperature MT-SOFCs and how this was achieved will be covered in more detail in Part II. The reduction of the operating temperatures also reduces the diversity of the fuels that can be used in these cells. Internal reforming requires temperatures in excess of 700 °C and so it may not be possible without external reformers operating at higher temperatures than the cells themselves. So there is a possibility that high temperature cells will always be needed for hydrocarbon fuel utilisation.

A very important part of a MT-SOFC reactor design will be the temperature management of the stack. This has been studied recently by Hashimoto et al. [26] experimentally and with some analytical techniques. The principle behind the technique is that the reduction in electrical potential of a MT-SOFC, as the amount of current drawn from the cell is increased, can be accounted for by mainly ohmic and polarisation losses associated with the electrolyte. The ohmic part of these losses should obey Ohm's law and thus be a linear function. However, if the resistance of the cell is relatively high compared to the current level, or if the cell has difficulty in radiating heat away fast enough, the cell will lose the linearity between ohmic potential drop and current. This is due to heat generation within the cell, which lowers the resistance of the electrolyte. This would imply that the electrical potential drop due to ohmic losses is related to the temperature of the cell.

To measure the ohmic losses and polarisation losses two methods were used. The current interrupt method is an AC technique that very briefly interrupts the fuel cell current and rapidly measures the terminal voltage before and during the interruption as seen in Fig. 7. A current pulse generator is used as an electric load and pulse source. An electrometer that has a high input resistance is connected to the potential sensing terminals of the cell under test. Using the electrometer the potential pulse can be measured by using an oscilloscope and thus the ohmic drop of the cell can be measured at any cell current.

The second method to measure the ohmic losses was electrochemical impedance spectroscopy (EIS). This method involves imposing the AC perturbation over a broad range of frequencies normally from 10 kHz to 1 Hz or lower. By monitoring the resulting variations in magnitude and phase of the cell voltage and current with a frequency response analyser the complex impedance (Z' and Z'') of the fuel cell can be found as seen in Fig. 8. This produces a detailed data set and several parameters may be extracted including the ohmic losses from the cell, kinetics and mass transport within

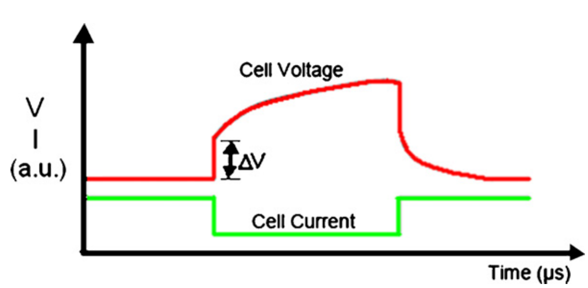

Fig. 7. The current produced by the fuel cell is very briefly interrupted using a current pulse generator and the initial change in cell voltage, due to losses is measured.

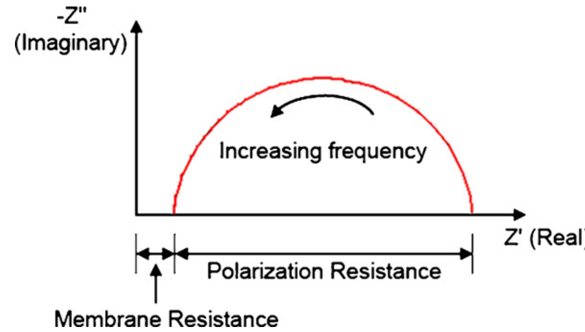

Fig. 8. Example EIS result where the amplitude and phase of the signals may be plotted in Nyquist format for analysis and modelling.

the cell. They report no differences in ohmic measurement results between the two methods indicating that the much less expensive current interrupt method is just as effective as the costly EIS method.

The steps to define the cell temperature as a function of the current drawn from the cell are as follows. Firstly, polarisation of the cell during power generation is measured by EIS or the current interruption method. The ohmic potential drop as a function of current is obtained and the ohmic resistance at each current is calculated. Secondly, the temperature dependence of the cell resistance at OCV or in the low current density region, where the ohmic potential drop follows Ohm's law, is measured. By applying the change in ohmic resistance in the cell, the cell temperature as a function of current can be estimated.

While experimentally validating their theory they found a dramatic reduction in the ohmic resistance of the electrolyte of a cell as the current density was increased. This would mean that there should also be a large increase in temperature around the cell. Compared to their electrochemical technique for measuring the temperature, the experimental results matched quite well, where there was the same slope in temperature gradients, but 10 °C less in the experimental result. This was most likely caused by the distance of the thermocouple away from the cell.

The methods used by this group could be a very useful non-intrusive method to measure the temperature of cells within bundles or stacks where the temperature at points within the reactor is related to the current being drawn from a cell. The authors make suggestions for the improvement of the measuring methodology. These include, better positioning of the potential sensing terminals, the addition of other potential sensing terminals and making a regular measurement of the dependence of the testing cell's electrode resistance to temperature.

Table 1 summarises some of the qualities of the MT-SOFC stacks that were found during the course of this research paper. Probably the most advanced micro-tubular reactor has been made by Nanodynamics (USA) Inc. which is already being commercialised. This system runs on propane/natural gas and reforms the fuel via partial oxidation (POX) reformation. There are currently 50 and 60 W systems for battery recharging. While there are some papers dealing with improving the performance and optimisation of the micro-tubular cells from this company, there is very little information available on the stack design and system operation.

A key component in MT-SOFC reactor design for commercialisation has been addressed by Sammes et al. [27]. Once the performance of the stack has been decided the choice of the optimal active surface or number of MT-SOFCs has to be decided based upon economic reasoning. This is divided into investment/buying cost versus operating cost. Increasing the current density should lead to a lowering of the initial investment cost because less cells will be needed. However, at the same time the increased current

Table 1
Comparison of the researched micro-tubular SOFC stack designs.

Name of group	Cell diameter (mm)	Support	Stack power (W)	Development stage	Operational temperature (°C)	Heat up time (°C min^{-1})
Adelan (GBR)	∼2	Electrolyte/anode	500 (1000 cells)	Demonstration model	850	200
AIST (JPN)	0.8	Cathode	2 (20 cells)	R&D	500	83
Alberta Research Council (CAN)	∼2	Anode	20 (∼30 cells)	Thermally self sustaining model	800	N/a
Korea Institute of Technology (KOR)	10	Anode	700 (36 cells)	Demonstration model	750	6
Nano-dynamics (USA)	N/a	Cathode	250	50 and 60 W models in the market	800	550
TOTO (JPN)	N/a	N/a	43 (14 cells)	N/a	700	N/a
University of Connecticut (USA)	13.2	Anode	100 (40 cells)	Demonstration model	850	40
Upper Austria University of Applied Sciences (AUT)	∼2	Anode	3.5 (7 cells)	Early experimental stacks	850	N/a

density reduces the fuel efficiency resulting in an increased operating cost. So a trade off will be required between the buying cost and the running costs. This will determine the optimal size of the active surface and consequently the number of MT-SOFCs. Due to the fact the MT-SOFC stack is at such an early stage in its development there are probably very few groups considering this. But it is a critical reactor design consideration when stacks for the consumer market should be made.

4. Sealing of micro-tubular SOFC stacks

A MT-SOFC sealing is more difficult than the sealing of the larger planar, tubular and monolith types. In these larger types the seal would be located in a specific large area. However, with the MT-SOFCs, sealing occurs in small amounts to connect each of the individual cells to its gas feed and exhaust ports. This results in many more possibilities of seals being or becoming defective and thus a much higher risk of fuel leaks. For example groups such as Serincan et al. [28] have already shown the extent of possible temperature variations across the MT-SOFC. So the possibility of a substantial temperature gradient across the position where the seal may be is a very important consideration. This thermal gradient is likely to occur at the sealing location because the active heat producing cells will be joined to passive manifolds at this position.

Typically hydrogen travels through the inside of the cell and air will travel around the outside of an array of cells, because this is a much easier way to contain the hydrogen and control the fuel supply to the cells. Flowing the oxidant through the inside of the cell could mean that very high flow rates of air inside these thin tubes would be required because of the high concentration of N_2 in the air. Leaks of hydrogen into the air flow can be catastrophic for the performance and durability of a MT-SOFC stack. When hydrogen leaks at high temperatures it causes a flame, which is both safety-wise unacceptable and also causes damage by overheating and vaporisation of seals as seen in Fig. 9. The flame removes oxygen from the stack and can also melt the metallic current collectors attached to the cathodes. This leak also increases the possibility of the re-oxidation of the anode which is fatal for a cell's power producing capability.

As sealing is such an important component in the operation of a MT-SOFC, companies seem to guard their sealing methods and so the only literature available at present is not specifically for MT-SOFCs but may be applied. One of the only cases where the sealing was explicitly described for MT-SOFCs was by Hashimoto et al. [29]. They describe the sealing techniques that they used for pressurised tests on MT-SOFCs. They used a combination of Aron C and CC (Toagosei Japan Ltd.) ceramic glues to seal their cells to aluminium oxide tubes and then tested their cells and seals under pressures of 0.7 MPa. They found that the seal worked very well. Detailed information on the process they used can be found in the paper [29].

In many instances the sealing will be between a zirconia electrolyte and a high temperature resistant metal frame, which has the advantage that the electrolyte acts as an electrical insulator. Seals can also be from an electrode, normally the anode to a metal interconnect, which can act as a flow path for the free electrons produced by the cell if the seal is metallic. Depending on the design, this seal can be exposed to operating temperatures of about 750–850 °C in MT-SOFCs that still work best in this range. This seal will always be exposed to an oxidising atmosphere on the cathode side. On the anode side the seal will be exposed to a wet fuel gas, containing various ratios of hydrocarbons (e.g., CH_4, H_2, CO, CO_2, and H_2O) and the seal will always be between these two conditions. The housing material can also be as important as the sealing material itself. Crofer E-brite (26Cr–1Mo) and heat treated 430 stainless steel are some of the materials being considered.

Strong metal to ceramic seals seem to be quite difficult to realise even at ambient temperatures and the extreme temperatures required by MT-SOFC stacks make the sealing problem substantially more difficult. Glasses are most widely used to make metal–ceramic seals [30] because they can be modified to have a very close match of thermal expansion with other fuel cell components. These seals show good hermeticity and good thermal and environmental stability.

In a very comprehensive review paper Raj [31] breaks down the different sealing approaches into groups. Outlined in his own research are alloys like FeCrAlY (Fe 22% Cr, 5% Al, 0.2% Y) as a metal for sealing by brazing to YSZ. Other methods that use glasses that crystallize either during processing or during use in an SOFC stack, thereby forming a more rigid glass seal are also discussed. The author states that these seals are hermetic because they have a very small crystalline phase but require a close match of the coefficient of thermal expansion (CTE) and may be less forgiving against seal fracture under thermal transients. Self healing glass was studied by creating cracks in seal samples and then observing the healing at elevated temperatures. It was found that some of the glass seals performed quite impressively even after as many as 300 thermal cycles between room temperature and 800 °C. Along with the stability, the glass seals retained their ability to self-heal even after very long exposure times of more than 3000 h at higher temperatures.

Raj also talks about a number of more novel sealing concepts that are being investigated by groups, such as using polymers of

Table 2
Comparison of the type of electrical connections used by MT-SOFC development groups.

Name of Group	Cathode connection	Anode connection	Connection profile and architecture
Adelan (GBR)	Silver wire wrapped around the cells	Internal wire-mesh contact to the anode	Modules containing cells in parallel
AIST (JPN)	a. Silver wire wrapped around the cells b. Cells impressed in cubic cathode matrix	Connection to one end of the anode	a. Modules with serially connected cells connected in parallel b. Parallel or serial connection of all cells within the module
Alberta Research Council (CAN)	Gold or silver layers applied by a sintering technique	Gold or silver layers applied by a sintering technique	Modules with cells, connected in parallel connected in series
Korea Institute of Technology (KOR)	Silver wires wrapped around the cells	Capillary pipe provides fuel and current collection	Parallel
Nano-dynamics (USA)	N/a	N/a	N/a
TOTO (JPN)	N/a	N/a	N/a
University of Connecticut (USA)	Brazed seal at one end of the cell which also collects the current	Brazed seal at the other end of the cell which also collects the current	Serially connected cell modules connected in serial or parallel thereafter
Upper Austria University of Applied Sciences (AUT)	Silver wire wrapped around the cells	Internal wire contact to the anode	Several methods under test

Si–C–N to make seals to SOFC components and a novel approach of mica and mica–glass hybrid compressive seals.

In review paper, Lessing [30], discusses some types of sealing methods. Glass–ceramics are a common type of sealing method, where compositions of glasses that are amorphous when liquefied but are specifically designed to partially or fully crystallize and become opaque when held in a high temperature range that is below the melting/solidification temperature range. Lessing also gives an in-depth analysis of this type of sealing stating its problems.

Metal seals are also a very interesting and a plausible solution to the sealing problem for MT-SOFCs but the types of metals that are suitable are limited because of their oxidation at high temperatures or insufficient melting points. Silver is a possible solution, however, based on experimental observations of Singh et al. [32] it was concluded that silver may not provide adequate long-term structural stability under a dual oxidation reduction environment. It is suggested that porosity formation in the silver seals is related to the formation of gaseous H_2O bubbles due to simultaneous diffusion of hydrogen and oxygen followed by subsequent interaction resulting in the formation of steam. Abernathy et al. [33] attached silver wire to a patterned $La_{0.8}Sr_{0.2}MnO_3$ cathode. This cathode was put into a regulated temperature and atmosphere chamber that was filled with Cr-containing vapor. Using Raman spectroscopy they found silver chromate, Ag_2CrO_4, on the surface of the silver at 500 °C and at 625 °C the Ag_2CrO_4 was deposited by vaporisation across the LSM surface. The evidence of this contamination means that choosing the type of metallic interconnect is a serious consideration to be made especially in MT-SOFC systems with silver components.

Tucker et al. [34] have shown that Ticusil (68.8Ag–26.7Cu–4.5Ti) even after complete oxidation could be a sealing solution. The brazed strength, sealing ability and conductivity of this material remains sufficiently high after application. This material oxidises rapidly in air above 700 °C and sample Ticusil joints have been exposed to fuel/air dual atmospheres. After exposure the joints remained robust and free of voids. They found, even after very rapid cycling, that no de-lamination or cracking was observed in the vicinity of the joint. This enabled 30 rapid thermal cycles of a braze-sealed metal-supported tubular cell with no loss of open circuit potential. Based on these observations they concluded that silver-based brazed alloys filled with low CTE particulates are strong candidates for use as the seal or interconnect in SOFCs operating at 700 °C. These types of seals could be suitable for MT-SOFC stack designs because of the ease of application and dual role as a gas tight seal and current collector. Recently Kim et al. [35] have shown the high temperature tolerance of silver–copper oxide air braze filler metals under dual oxidising reducing atmospheres at 800 °C. Regardless of the filler metal composition or brazing temperature employed, none of the specimens that were tested for 100 h displayed any signs of hydrogen embrittlement. Specimens exposed for a longer period of time, 1000 h, did exhibit evidence of internal porosity and the extent of this was limited to less than half the width of the joint. According to Kim et al. while the issue of hydrogen embrittlement remains a concern with respect to the long-term stability of silver-based brazes under dual oxidising/reducing environment, its development in the Ag–CuO filler metals is much slower than was previously reported in high-purity silver. This

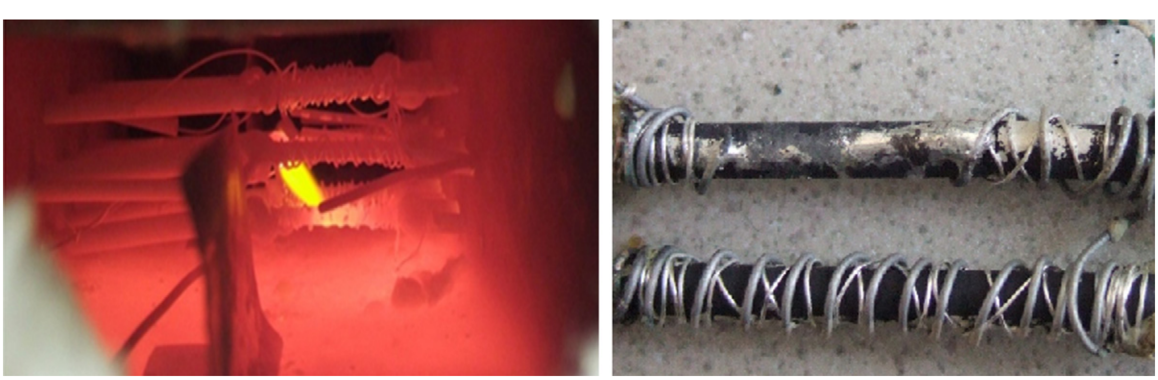

Fig. 9. An array of cells (left) and the damage caused (right) from a small flame in a reactor [55].

is both due to the longer diffusion path in the braze joints and the buffering effects that CuO reduction appears to afford. Kim et al. [36] have also investigated the possibility of a Ag–Al–Cu and Ag–Al brazes. , they report that while the Al improves the atmospheric tolerance it also weakens the joint strength. Lessing in his review [30] also points to the work of ceramic-composite seals being investigated by Sandia National Laboratory (USA) and NexTech Materials (USA Ltd). Sandia's stated composite approach is to produce a deformable seal based on using a glass above its normal glass transition point with control of the viscosity and CTE modified by using ceramic powder additives. The aim is that the composite should be rigid enough to remain in the joint, but have a low enough viscosity to slightly flow to relieve stresses and heal cracks. Lessing reports that NexTech Materials are using $BaO–CaO–Al_2O_3–SiO_2$ candidate glass compositions (e.g., 15 BaO 25 CaO 7.5 Al_2O_3 45 SiO_2) together with powders of mica, talc, alumina, or zirconia fibers as an additive to increase the seal viscosity.

Historically, both alumino-silicate cements and alumino-silicate cements plus glass were used during the early 1990s by various developers [30]. High temperature cements are easily available from a variety of commercial sources. However, after curing and firing, they are typically very porous and lack good wetting/bonding with metal interconnects and the ceramic surfaces that are needed to produce hermetic seals. Therefore, in some cases, glass was added to the cement in an effort to fill the porosity and aid wetting. This combination then becomes a "glass–ceramic" seal, when the mixture is fired to sufficient temperature to melt the glass. Sealing cements can also be produced by combining ceramic powders (e.g., zirconia) with chemically bonded ceramics.

5. Modelling and micro-tubular SOFC stack design

Producing electricity at high temperatures electrochemically has been studied by a large number of authors by modelling the coupled electrochemical and physical phenomena. Computation fluid dynamics (CFD) packages such as FLUENT (ANSYS, Inc. USA), CFD-ACE (ESI Group, Inc. USA) and Star-CD (CD-adapco Ltd. USA) all have their own inbuilt electrochemical models and indeed some academic groups have built their own models such as DETCHEM [37,38] to work with FLUENT or stand alone CFD codes. While some information can be gathered by using models without the electrochemical and mass transfer of the cells, the best models will certainly contain the electrochemical and mass transfer reactions.

For example Lockett et al. [39] have used some very simple features in the FLUENT modelling package to characterise heat transfer inside arrays of MT-SOFCs in counter flow. The purpose of these simulations was to identify possible stack designs and methods for efficient stack thermal management. This paper describes a 20-cell test system and claims to be the first of its kind for modelling flow around MT-SOFCs in a stack. This was a very basic attempt to use CFD to model the flow regimes within a stack. This model neglected radiation modelling, species removal and all of the electrochemistry. With a constant heat flux of 0.5 W on each cell it cannot be certain how the cells would work in reality because of the missing modelling variables.

While some studies such as Jasinski et al. [40] try to discover how these processes affect the cells through experimental approach, the vast majority of authors are developing models on the nano scale/molecular dynamics Lehnert et al. [41] and Qi et al. [42] macro scale/fluid dynamics Tanaka et al. [43] and Bhattacharyya et al. [44] and system scale/system dynamics Ivers-Tiffée et al. [45] and Xue et al. [46]. A paper by Hong [47] has shown how all the afore mentioned scales of models and their associated dynamics, can culminate into a real time simulation package for a vehicle fuel cell power plant.

Some of the fundamentals of the electrochemical processes in SOFCs are still not understood. Indeed the effects of the multi physical phenomena surrounding the electrochemical processes on a high temperature electrode have not been entirely resolved in the literature. The electrodes on either side of the electrolyte reduce the concentrations of the fuel and oxidant through red–ox reactions, producing thermal and electrical energy. The oxidant and fuel concentrations and thermal gradients surrounding MT-SOFCs and the power output of them should ideally be uniform for a cell that does not experience any other mass transfer or thermal disturbances. It is unspecified and not fully resolved in the literature to what degree the fluid, thermal, mass transfer and geometrical dynamics affect the performance of the electrodes.

An example of how models are ever evolving and giving a more accurate account for what is happening in the nano scale can be seen in the improvement in the modelling of the anode. Older studies which include models such as Lehnert et al. [41] have shown that there is a concentration gradient within the anodic electrode. It is suggested that the reaction zone size is 50 μm out stretched from the electrolyte. This layer is treated as a boundary condition and the fuel concentration through the anode appears as an exponential function. Hussain et al. [48] also suggested that the anode reaction zone is 50 μm but they consider the reaction zone as a volumetric continuum rather than a boundary condition and their result is more like a bi-linear function. They also show that considering the reaction zone as a boundary condition leads to over predicting the concentration over-potential at all loads.

The effect of radiation within reactors is a very interesting topic and is discussed by VanderSteen and Pharoah [49] who have shown that for both planar and tubular models that neglecting radiation effects in modelling results in overestimating the reaction rates, material properties and also the maximum theoretical voltage. Tanaka et al. [43] also concur with this statement and add that modern CFD packages are easily capable of including radiation models. However, Daun et al. [50] state that due to the minor effect of thermal radiation on the temperature field in planar SOFC cells, that thermal radiation can be excluded from a detailed CFD analyses of a planar anode supported SOFC. To the contrary Murthy and Fedorov [51] found that ignoring radiation effects caused errors of 100 °C in the surrounding temperature fields.

Damm and Fedorov [52] studied radiation with a two-dimensional model of an anode supported SOFC. This study found, in contrast to that of Daun et al. [50] that the effect of radiation accounts for a few degrees in the cell temperature field and states that heat is predominately removed from the cells by convection. The authors remark that their opposite hypothesis to that of Murthy and Fedorov [51] is because of the different SOFC geometries involved. So it can be concluded that the geometry within a MT-SOFC reactor can have a huge bearing on the radiation effects.

Qi et al. [42] model with finite volume methods the effects of transport dynamics of the fuel and oxidant. Their main objective was to measure how all of the macroscopic transport phenomena affect a cell's power producing performance. They only model a small 3D section of a tubular cell and apply step responses to inlet temperatures and velocities. The results include dynamics from the diffusion, inherent impedance, flow and heat exchange processes including radiation and internal shift processes. The results clearly show that fuel flow inlet pressure and temperature have large effects on the dynamic performance of a SOFC, especially with regard to its temperature response. It was also shown on the cathode side that the temperature of the incoming air has the most significant effect on SOFC solid phase temperature and performance when compared to other inlet flow properties. They report that the effect of flow velocity is not as significant as inlet pressures and temperatures. Their results also pointed out that the transient response of a SOFC was mainly controlled by the temperature dynamics. This would be a critical inadequacy of models that just apply a heat flux to the cells. For example Lockett et al. [39] recommend an air inlet temperature of 400 °C, which may well be correct for dummy cells outputting a constant flux, but this air temperature will have a huge affect on the thermal flux that the cells output. This is an example of why electrochemical models are a necessity when modelling a MT-SOFC stack design.

Comparisons in the literature of the co-, counter- and cross-flow methods, can only be found for big SOFC tubes and planar cells. Recknagle et al. [53] examined the effects of these three different methods with a validated electrochemical code and combined it with CFD software.

However, for the MT-SOFC, it is interesting to note that Xue et al. [46] and Lockett et al. [39] both assume a counter flow configuration. In both simulation results it seems that the counter flow operation results in lower cell structure temperatures at the inlets and outlets. This would be advantageous for the MT-SOFCs because high temperature seals are required in these positions. An area which is totally neglected in the literature is the performance of the MT-SOFCs or its larger predecessor mounted in the cross flow position. The tubular SOFC in cross flow is a very interesting research topic because of the wakes at the rear of the tubes.

The role of the flow regime between and around cells within arrays and also through them internally is a very important consideration in a MT-SOFC stack design. This issue has been addressed to some degree by several authors. For example recently Serincan et al. [28] have investigated the steady state behaviour of a MT-SOFC inside an oven in a very comprehensive and enlightening 2D model.

The fuel modelled in this paper is a mix of 25% H_2 in N_2 having a flow rate of 100 ml min^{-1}. The temperature of the fuel in the anode rose by about 110 °C between OCV and 0.2 V. Such a temperature response should affect the system response. Also they found temperature gradient field ranges of 18 and 40 °C on the cathode and anode sides. In their model the electrolyte temperature is slightly higher than the electrodes, ranging from 0.1 °C at OCV to 0.9 °C at 0.2 V. This is probably because of the reactions taking place in that vicinity. Fuel mixtures of H_2 and N_2 are not expected to have dramatic effects on the temperature distribution across a cell, even with different ratios of each because of their similar molar (volumetric) heat removing capacities. However, the use of hydrocarbon fuels flowing inside the anode containing extra un-reacted mass should be an issue to be considered for the temperature profile across a cell.

Making a MT-SOFC stack that is thermally self sustaining is a very important characteristic to have for high system efficiency. A critical consideration is how close should the tubes be stacked together and what are the effects on the flow regimes and heat transfer through bundles of cells? In an attempt to answer this question Lawlor et al. [54,55] performed a numerical analysis on high temperature flows through an array of cylinders. It was found that for a particular arrangement of MT-SOFCs exposed to flow rates expected inside a reactor that buoyant flows could be an issue for extremely low flow rates. However, these effects could be negated by forced oxidant flows over a single mm s^{-1} through this array of cells.

Chouikh et al. [56,57] studied the spacing distances between cylinders and the effects of heat transfer by natural convection. They concluded that the distances between the cylinders are paramount to the heat transfer and flow regimes of heated cylinders causing natural convection. Even though there are no detailed models in the literature on this topic Suzuki et al. [58] made manifold design and stack housing system which did not have a forced flow of oxidant between the cells. This reactor was placed inside an oven for the tests and natural convection and/or diffusion of oxygen from the surroundings must have been the oxidant refresher to the cathodes. From visual observations of the images of the cell stacking it in the paper, similar to Fig. 10, it seems that these cells are very close together, which is interesting with regard to the refreshing of oxygen to the cells.

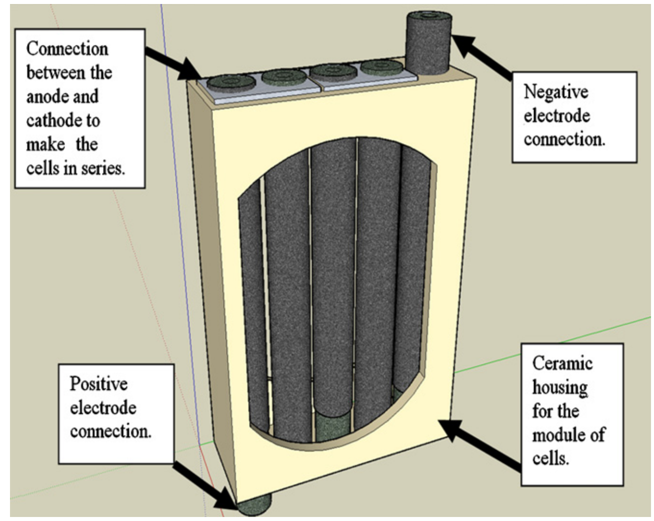

Fig. 10. Depiction of a single module in a multi module system used by Suzuki et al. [58].

The range of literature for flow around arrays of cylinders is wide and varied but none of it deals specifically with high temperature, laminar flow, around arrays of cylinders wrapped with wires, with electrical contacts, being affected by mass transfer.

6. Electrical connection and efficient power extraction from stacks

Current collection from the MT-SOFCs is critical for achieving high power densities from the cells and high fuel efficiency. There are many variables that effect the collection of current from the cells including, contact resistance, temperature, amount of current in the contact wires, means of connecting cells together (parallel or serial), choice of materials and geometry and scale of the cells. Most groups generally use a winding of silver around the cathode and nickel meshes or wires as the means of connection to the anode. These materials have high melting points, resistance to oxidation and corrosion and high electrical conductivity. To reduce the contact resistance on the cathode side, silver paste is normally used as a transition between the silver wire wrappings on the cathode and the cathode itself. Indeed Serincan et al. [59] have performed some numerical simulations where coating the entire cathode surface with silver was suggested as a viable current collector for MT-SOFCs.

The crucial question is, are there mass transport limitations when a layer of silver is coated over a whole cell? To find this out the silver coating on the cathode was given different porosities using a simple model which incorporates mass, momentum and charge balances as well as the species transport. The results suggest that for a broad range of porosity values, mass transport losses are negligible due to the small scale of the fuel cell. They also compared this design with a more common type of current collector arrangement with silver paste on the cathode that utilises several pasted strands of silver, length ways along the cathode as seen in Fig. 9.

In this case the potential losses were found to be negligible for the same reason related to the small scales of the system. A non-uniform current density distribution inside the cathode was predicted by the model, which would lead to a non-uniform reaction rate and temperature distribution along the cathode.

The authors conclude that such a non-uniform temperature distribution maybe severe enough to cause mechanical failures such as warping and distortion of the cathode and the separation of the silver paste from the cathode. However, there is no experimental data to back this up. Table 2 presents a summary of the different methods of electrical connections used by the main groups in the field.

Connecting the cells in parallel or series has advantages and disadvantages. When cells are connected in series the voltage is increased incrementally, while the current is limited by the worst performing cell. However, a break in the connection, for example by a damaged cell or broken wire, leads to a failure of current collection among all the cells in serial connection. Connecting the cells in parallel of course will not cause all the cells to stop providing current if one malfunctions unless there is an electrical shortcut. In this case the current rather than the voltage increments up. This is undesirable because of the associated wires losses. A combination of both serial and parallel connections should be used to extract the current from the reactor. Current collection is an area in the literature which has not received any meaningful attention with regard to what combinations work best. Some authors [2,13,58] have presented the cells connected in series within smaller modules and then these modules are connected in parallel to make larger stacks. The other approach is to connect modules of cells initially in parallel and then in series or parallel. Initially the AIST (Japan) Inc. group connected their cells in parallel [24] but recently they have invented a serial approach [11] which they hope to use to increase the power production of their stack modules by reducing the losses through resistance. Of course the numbers of cells in the modules will be determined by the size of the stack, but there should be a balance between, efficiency, costs and probability of a cell becoming defective to decipher the most optimised stack electrical configuration.

For example if every cell is connected individually to external power conditioning hardware a lot of wires will be needed, thus increasing the total system cost. If all the cells are just connected in series to build up a high voltage, then the amount of wiring is reduced but the probability of a cell failure causing the failure of the system or a part failure is increased. This is an area which deserves addressing and is essential for the cost of these systems versus reliability. Keeping the electrical wires and contacts out of the hot zones or at least as short as possible in them is advantageous because in these hot zones the wires will have higher resistances and also sufferer degradation faster. So an appropriate geometry design with consideration for the electrical current collection should also improve the system efficiency and reliability. The attempt by several groups to reduce the operating temperature of the MT-SOFCs should also reduce the electrical losses.

The power coming from a MT-SOFC reactor will have to be conditioned through the use of an inverter unit, especially if it is used as an auxiliary power pack or for an electric motor. The inverter should be compact and highly efficient. Since driving an EV motor requires a large current, the heat generated by the power module in the inverter, has been a problem impeding the achievement of compactness and high efficiency. The transistor switching losses increase and free wheel diode (FWD) reverse recovery losses increase linearly with the switching frequency while the total loss increases quadratically as the current increases. In other words, a higher switching frequency and current both induce more power module loss and heat generation and this reduces the efficiency of the system.

To cope with this heat generation requirement a larger power module and a cooling system is required, which is also undesirable. Attempting to suppress the current would compromise the performance. Isolated gate bi-polar transistor switching (IGBT) technology is used for breakdown voltages above 1000 V, while the metal oxide semiconductor field effect transistor (MOSFET) is used for device breakdown voltages below 250 V. Between 250 and 1000 V, there are many technical papers available from manufacturers of these devices, some preferring MOSFETs and some IGBTs. However, choosing between IGBTs and MOSFETs is very application-specific and cost, size, speed and thermal requirements should all be considered. To optimise the total system conversion chain, the output voltage and current of the MT-SOFC stack, which will be determined by the parallel and serial arrangement of the cells within the stack, should be also considered.

Also to be considered is the costs. The size of the components involved in the power inverter depends on the output frequency and current. As the frequency is reduced the size of the capacitance and inductive components must be increased to store larger amounts of power between charge and discharging. It is also desirable to keep the frequencies above 20 KHz so that the switching is not audible. Increasing the voltage would seem to be the solution but then undesirable ripple voltages occur and wire insulation can become an issue.

7. Level of research in micro-tubular SOFCs and what is being researched in the field

To compare the quantity of research in larger SOFCs with the micro-tubular version the Compendex database on the Engineering Village web site [60] was used in November 2008. This site contains a data base of scientific and engineering publications from the most popular journals around the world. It was found that while comparisons can easily be made between the levels of research between larger SOFCs and the micro-tubular type, when these are subsequently broken into paper themes it is difficult to make a direct comparison. This is because the larger SOFC papers tend to be more specific while the papers relating to the MT-SOFC tend to include several topics in a single paper, especially in stack design papers.

The following results were found for the number of publications which had solid oxide fuel cell or SOFC in their abstracts, titles or subject fields. Using different inclusion and exclusion parameters in the advanced search engine on the site, these results were broken down into their main themes. It is clear that there has been a recent surge in the interest in general SOFC research as shown in Fig. 11. These results are also broken down into the themes of the research papers and indicate a good spread of research

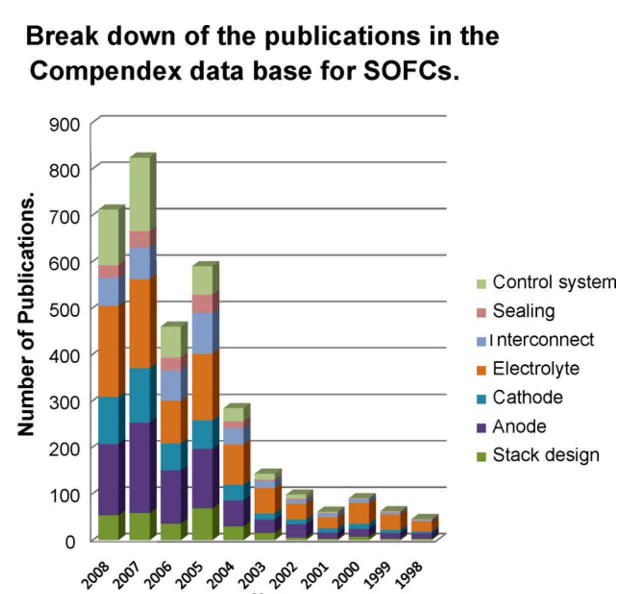

Fig. 11. Search results from [60] for articles with the terms {solid oxide fuel cells (OR) SOFC} (AND/OR/NOT) a theme in their titles abstracts or subject fields.

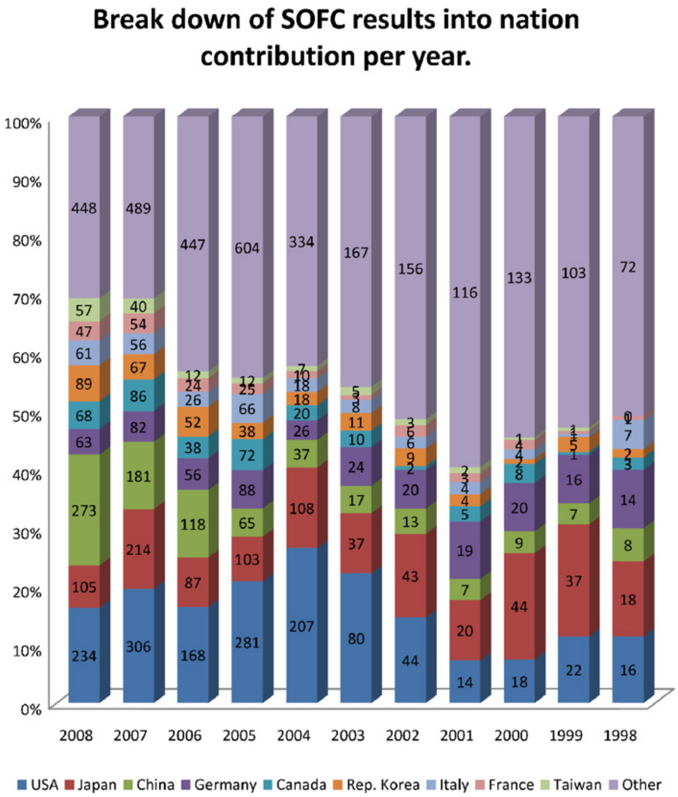

Fig. 12. Break down of the search results in Fig. 11 into the countries of origin of the major contributors.

over the whole topic. It should be noted that while these papers are not directly related to the MT-SOFC, many of them may be applied such as control systems, sealing and interconnects. It seems that as the interest in SOFCs increases so also has the interest in the peripheral devices such as sealing, interconnects, control systems and stack design. It may be the case that these peripherals become of more interest to SOFC groups and groups of other disciplines as the quality of the cells improve and the role of the different peripherals become more necessary for fully functioning stacks.

Fig. 12 illustrates the percentage of the total for each year of publications from each of the major contributing nations. It is clear

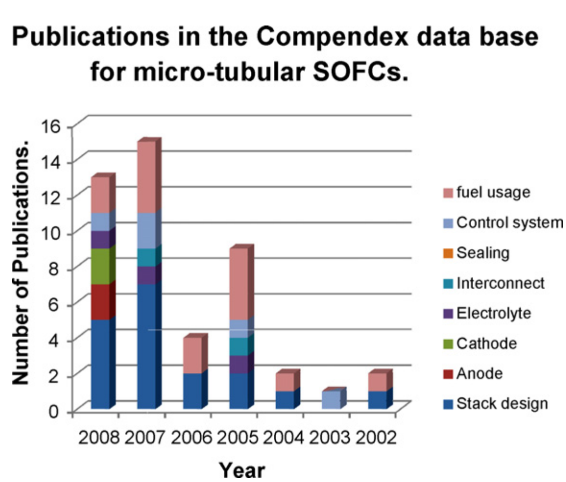

Fig. 13. Break down of the search results in [60] using an advanced search more specific to MT-SOFCs.

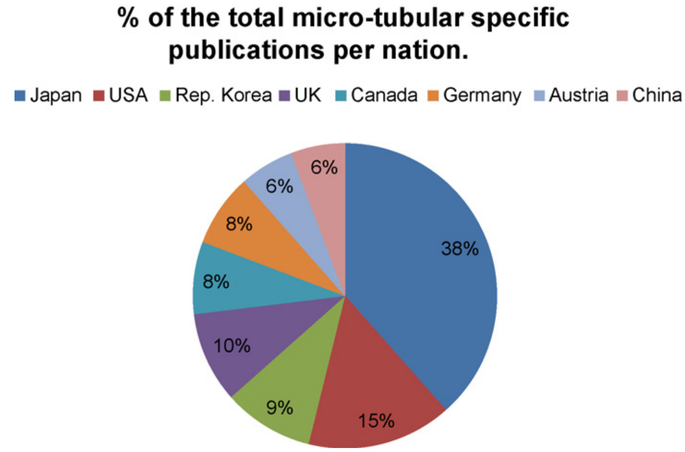

Fig. 14. Break down of the search results from Fig. 13 into percentage publications per nation since 2002.

from this plot that China and Korea seem to be continually expanding their research. Also the contribution numbers of all the major contributors in the last 7 years has continually eaten into the percentage contributed by the non major contributors. This could be an indication that the major contributors, because of their previous research activities, are becoming much more specialised and therefore will be the dominant proprietors of the SOFC know how and intellectual property.

This review has focused more specificaly with the micro-tubular SOFC and so a more sutible search query was inserted into the Engineering Village's Compendex data base search. The result can be seen below in Fig. 13, which is broken down into the main topic of the publications. It was found that the MT-SOFC publications tend to be much more general. For example there is not any literature that deals specifically with the sealing of these cells. However, some information about sealing can be found within texts based on other themes. So for example a single paper can include several topics such as fabrication of the anode cathode and electrolyte, with some manifold design, sealing techniques and stack arrangment.

It can be seen from the results Figs. 12 and 13 that only a small proportion of the overall publications related to SOFCs deal specifically with the MT-SOFC. It should be remembered that some of the topics included in the general SOFC do not also apply to the micro-tubular version. Because of the much lower numbers of publications it was also noticed that many papers for the MT-SOFCs dealt specifically whith the theme of running the cells on different fuels.

From Fig. 14, which is a breakdown of these results into their nation of origin, it can be seen where most of the development is taking place in this field. This does not account for non-published research but it should still be a good indication. Breaking these results down shows that 38% of the publications in the past 7 years came from Japan alone, 21% from America and Canada and 20% came from EU members.

Winkler [61] researched a worldwide survey and review of research taking place dealing with micro scale SOFCs. Also outlined in this document are substantial projects for micro-tubular and micro-planar SOFCs. Four were found in the US, two each in the EU zone, Japan, Canada and Republic of Korea. It is very clear that the points and advantages raised by Van Herle et al. [4] in 2000 about the potential of MT-SOFCs for portable applications have been certainly proved. While the larger type of SOFC may be closer to large scale commercialisation than the MT-SOFC the potential of the micro-tubes seems to be over looked by the broader SOFC community.

8. Conclusions

This paper has discussed the topics of MT-SOFC stack design and application. In the process a broad range of scientific disciplines, including materials sciences, chemistry, thermo-fluid engineering and mechanical engineering and of course electro-chemistry have been included. Scientific and engineering content has been discussed in the areas of the optimisation of the cell performances within reactors, sealing within reactors, rector design and a review of some of the modelling taking place, which can be applied to MT-SOFC stack design.

An important conclusion of this study is that while there are some products on the market using MT-SOFCs, their potential as power producing devices for a whole range of applications has yet to fulfil its potential. With so many technological advances possible and the diverse range of scientists and companies researching in the field it is highly possible that this type of SOFC may have a bright future as more efficient power generating mechanisms become in demand. But also this form of SOFC could turn out to be extremely profitable, when alternative inexpensive methods of producing APU and vehicle power are required, because of the relatively small scientific community and industrial partners advancing it.

However, to reach this goal it is suggested that targeted research is required in the area of sealing, manifold design and optimisation of the fuel and oxidant flow regimes. A key area that has not received quality research is the electrical connection regimes. For example should these cells be connected in parallel or series or which combinations of these is most effective in reducing power losses while also reducing the effect of a single cell failure on the whole stack? While the reduction of the operating temperature of these cells offers the advantages of less thermal stresses and a wider range of materials suitable for sealing, frame material and electrical connections. However, lower operating temperatures negates the internal reforming attribute of the MT-SOFC and reduces the system efficiency when hydrocarbon fuels are to be used. High temperature operation of MT-SOFC stacks that run on hydrocarbons should certainly not be forgotten amidst the recent trend of lower temperature stacks.

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