

## 1

## Introduction

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The current and future energy scenarios faced by the international community are discussed in this chapter, starting with a brief presentation of the energy landscape and related issues, including the increase in demand and environmental aspects. A list of possible solutions to existing and foreseen problems is presented and discussed, setting the framework to highlight the significant potential of fuel cells for future power generation. Following on from this, the performance characteristics of fuel cells are introduced, including an analysis of their different types and corresponding differential features. Additionally, attention is devoted to hybrid systems based on the coupling of high-temperature fuel cells and microturbines (mGTs).

### 1.1 World Population Growth, Energy Demand and its Future

A study carried out by the United States Census Bureau (USCB) [1] estimated that the world population exceeded 7 billion on

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Mario L. Ferrari, Usman M. Damo, Ali Turan, and David Sánchez.

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12 March 2012. Now, at the time of writing in August 2016 with the global population standing at about 7.4 billion [2], this figure is expected to continue rising over the coming decades [2]. As the world population grows, in many countries faster than the global average of 2%, the need for more and more energy is intensifying in a somewhat similar proportion, thus putting pressure on the natural resources available and existing infrastructures. This higher energy consumption is not only due to the growth in world population, but also to the improved lifestyles leading to a greater energy demand per capita (two features that inevitably come together). This is best exemplified by the fact that the wealthy industrialized economies comprise 25% of the world's population but consume 75% of the world's energy supply [3]. A recent study (from ref. [4]) shows that the total world consumption of marketed energy is expected to increase from 549 quadrillion British thermal units (Btu) in 2012 to 629 quadrillion Btu in 2020, and to 815 quadrillion Btu in 2040 – a 48% increase from 2012 to 2040 [4].

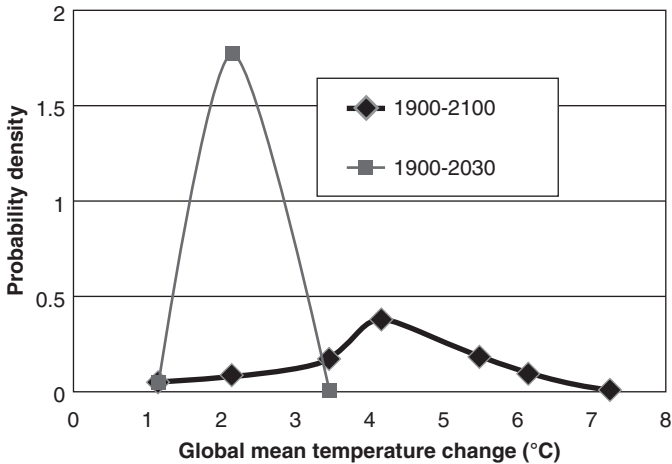
Indeed, the landscape of future energy demand and generation projected for the world seems rather bleak, as most nations, including the most developed ones, depend primarily on conventional energy sources such as oil, coal and gas to generate power not only for the domestic and industrial sectors but also for transportation. This dependency results in global warming, contributes to rises in fuel prices that constitute a burden on economies, and can lead to delays in energy production and supply [5, 6]. Furthermore, even if the global production of fossil fuels is currently sufficient to cover the world's needs, the exponential rise in the exploitation rate of this finite, fast-depleting resource would pose a risk to the future energy demand and generation balance [7–9]. In the long run this global dependence on conventional fuel sources for power production will prove problematic because the world will eventually fall short or run out of these resources. Renewable energy sources are often set forth as a feasible alternative to this fossil-fuel dominated world [10], although many of their inherent features, such as their low energy density, intermittency and geographical distribution, pose a number of challenges that remain to be solved today.

## 1.2 World Energy Future

Due to the heavy reliance of most nations worldwide on fossil fuels for power generation and transportation, the atmospheric concentrations of carbon dioxide and methane have increased by 36% and 148% respectively, compared with pre-industrial levels [11]. These levels are indeed much higher than at any time during the last 800,000 years, the period for which reliable data have been extracted from ice cores. This observation is further confirmed by less direct geological observations that also show that carbon dioxide concentrations higher than today were last seen about 20 million years ago. These findings suggest that the root cause for such high concentrations is anthropogenic, mainly hydrocarbon-based fuel burning (responsible for three-quarters of the increase in CO<sub>2</sub> from human activity over the past 20 years) and deforestation [11]. Other environmental factors, including air pollution, acid precipitation, ozone depletion and emission of radioactive substances, are also of concern and raise awareness of the negative impact of human activity on the environment [3].

As a consequence of this massive production of anthropogenic carbon dioxide and other greenhouse gases (trace gases in particular [12]), global temperatures in 2016 were 0.87°C above the long-term 1880–2000 average (the 1880–2000 annually averaged combined land and ocean temperature is 13.9°C), which translates into a warming rate of around 0.61°C/century over the last few decades. In particular, the average temperature of the Atlantic, Pacific and Indian oceans (covering 72% of the Earth's surface) has risen by 0.06°C since 1995. The situation regarding global warming is far from being under control. As stated by the US Department of Energy's forecast, carbon emissions will increase by 54% above 1990 levels by 2015, making the Earth likely to warm by 1.7–4.9°C over the period 1990–2100 (see Figure 1.1). Such observations demonstrate the need for efforts towards alleviating energy-related environmental concerns in the near future [3].

Achieving higher efficiencies and, if possible, the utilization of renewable energies in power generation technologies will



**Figure 1.1** Global mean temperature probability changes, for the years 1990–2100 and 1990–2030. *Source:* Omer (2008) [3]. Reproduced with permission of Elsevier.

be vital steps in mitigating or reducing these environmental problems, whilst meeting the expected rise in energy demand in the future. With increasing fuel prices and significant pressure to reduce emissions, increasing energy efficiency is considered amongst the most feasible and cost-competitive approaches for reducing CO<sub>2</sub> emissions. For instance, Britain wastes 20% of its fossil fuel and electricity which, if used efficiently, would translate into a potential £10 billion annual reduction in the collective fuel bill and a reduction of some 120 million tonnes of CO<sub>2</sub> emissions [3]. Unfortunately, even if energy is currently recognized globally as being at the centre of the sustainable development paradigm, the industrial and social development paths favour energy consumption rather than conservation [3].

The significant fuel consumption and CO<sub>2</sub> emission issues have to link with the fact that conventional thermal power plants (regardless of the type of fuel used) cannot convert all of the thermal energy supply into useful (mechanical) work. In most cases, more than 50% of the heat added to the working cycle is rejected to the environment. Combined heat and power (CHP) installations are able to use a part of this heat, which would otherwise be wasted in a conventional power plant,

to raise the overall first law efficiency to values higher than 80% for the best available technology [3]. This concept enables drastic reductions of the primary energy consumption and cost compared with the independent production of both forms of energy (electricity and thermal energy).

Complementary to energy conversion at high efficiency, substituting fossil fuels with renewable energy sources is envisaged as another means to tackle the aforementioned social, economic and environmental problems. Renewable energies are broadly regarded as energy sources that are naturally replenished over a short timescale (i.e. in comparison to the lifetime of a human being), such as sunlight, wind, rain, tides, waves and geothermal heat. They have shown the potential to replace conventional fuels in various distinct areas, such as utility-scale electricity generation, hot water production/space heating, fuels for transportation, and rural (off-grid) energy services [13, 14]. Renewable energy sources have the potential to constitute the future energy sector's backbone, despite some evident shortcomings such as low density and inherent intermittency.

According to the REN21's 2014 report [15], renewables contributed 19% to the world's energy consumption in 2012, and 22% to electricity generation in 2013, using both traditional (biomass) and more innovative renewable energy technologies such as solar power, large wind farms and biofuels [10]. The importance of renewable energy sources has been disseminated widely, and several nations worldwide have decided to invest large sums of money in renewable technologies; such is the case in the US with a total investment of more than \$214 billion in 2013, whereas other countries like China are following close behind [15].

Hybrid systems based on the coupling of a microturbine with a high-temperature fuel cell are highly regarded as a solution for future power generation due to their high efficiency, ultra-low emissions and their ability to run on fuels such as hydrogen produced from renewable sources. These systems can achieve very high efficiencies: more than 60% electrical efficiency using natural gas (depending on the low heating value). This efficiency is virtually independent of plant size due to the modular nature of these devices. Hybrid systems based on solid oxide fuel cells (SOFCs) are of particular interest, because they have the

potential to overcome the main limitations of traditional power plants, and furthermore to meet the hurdles posed for the world's future energy need without worsening environmental issues.

## **1.3 Introduction to Fuel Cells and Associated Terms**

A fuel cell is a device that converts the chemical energy in a fuel into electricity through an electrochemical reaction with oxygen. Hydrogen is commonly used in a fuel cell, but hydrocarbons such as natural gas and alcohols like methanol are also used. In contrast to batteries, fuel cells require a constant source of fuel and oxygen/air to sustain the chemical reaction and thus produce electricity as long as this input flow is supplied [16, 17].

A fuel cell typically consists of an anode (negative electrode), a cathode (positive electrode), and an electrolyte that allows charges to move between the two sides of the fuel cell [16, 17]. Direct current electricity is produced when electrons are drawn from the anode to the cathode. Typical layouts and choices of materials vary between fuel cell types. A classification of layouts/materials used in different types of fuel cells is shown in Table 1.1.

### **1.3.1 Background for Fuel Cells and Thermodynamic Principles**

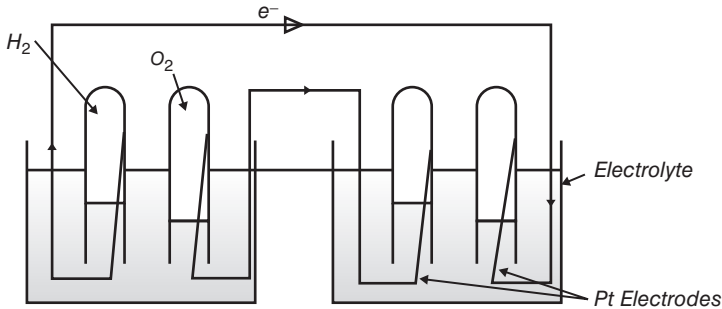
Fuel cells as power systems were first conceived and realized by Sir William Grove in 1839, using the experimental setup shown in Figure 1.2 [18]. His demonstration is the reverse of electrolysis: an electric current is produced through the process of combining hydrogen and oxygen to form water. This process is an 'electrochemical burning' reaction (although no real combustion is present in a fuel cell) which consumes hydrogen as fuel and produces electricity instead of heat.

Despite the fact that a large proportion of those with an interest in fuel cells are professionals with a background in heat engines, it is wise to revisit some fundamental concepts in chemical thermodynamics. To this aim, a brief introduction to the Gibbs potentials is provided below.

**Table 1.1** Comparison of fuel cell technologies [17].

	PEFC	AFC	PAFC	MCFC	SOFC
Electrolyte	Hydrated polymeric ion exchange membranes	Mobilized or immobilized potassium hydroxide in an asbestos matrix	Immobilized liquid phosphoric acid in SiC	Immobilized liquid molten carbonate in LiAlO <sub>2</sub>	Perovskites (ceramics)
Electrodes	Carbon	Transition metals	Carbon	Nickel and nickel oxide	Perovskite and perovskite/metal cermet
Catalyst	Platinum	Platinum	Platinum	Electrode material	Electrode material
Interconnect	Carbon or metal	Metal	Graphite	Stainless steel or nickel	Nickel, ceramic, or steel
Operating temperature	40–80°C	65–220°C	205°C	650°C	600–1000°C
Charge carrier	H <sup>+</sup>	OH <sup>-</sup>	H <sup>+</sup>	CO <sub>3</sub> <sup>-2</sup>	O <sup>=</sup>
External reformer for hydrocarbon fuels	Yes	Yes	Yes	No, for some fuels	No, for some fuels and cell designs
External shift conversion of CO to hydrogen	Yes, plus purification to remove trace CO	Yes, plus purification to remove trace CO and CO <sub>2</sub>	Yes	No	No
Prime cell components	Carbon-based	Carbon-based	Graphite-based	Stainless-based	Ceramic
Product water management	Evaporative	Evaporative	Evaporative	Gaseous product	Gaseous product

Source: US Department of Energy.



**Figure 1.2** First demonstration of a fuel cell by Grove in 1839. Source: Srinivasan (2006) [18]. Reproduced with permission of Springer.

Let  $\Gamma$  be a chemical system whose absolute pressure, temperature, volume and entropy are denoted by  $p$ ,  $T$ ,  $v$  and  $S$  respectively. The following four Gibbs potentials are defined to calculate the work yielded by the system: internal energy ( $U$ ), Helmholtz free energy ( $A$ ), enthalpy ( $H$ ), and Gibbs free energy ( $G$ ), as shown in equations 1.1–1.3.

$$A = U - TS \quad (1.1)$$

$$H = U + pV \quad (1.2)$$

$$G = H - TS \quad (1.3)$$

These Gibbs potentials are used in the analysis of a wide range of processes.

The total work yielded by the system  $\Gamma$  during an infinitesimal process,  $W$ , is higher than the amount of work that can actually be employed by a potential user,  $W_u$  [19]. The difference between both works is that done by the system on its surroundings:

$$dW = dW_u + d(pv) \quad (1.4)$$

Based on this consideration, the four state functions previously listed are used to equate the First and Second Laws of thermodynamics applied to a closed system (Eqs 1.5 and 1.6), where work  $W$  and heat  $Q$  are considered positive when they flow out of and into the system respectively:

$$dU = dQ - dW \quad (1.5)$$

$$dS = dQ/T + dS' \quad (1.6)$$



Combining equations 1.2, 1.4 and 1.5 yields the following alternative form of the First Law:

$$dH = dQ - dW_u \quad (1.7)$$

Equations 1.5 and 1.6 can be interpreted in the following terms:

- If the process followed by the system  $\Gamma$  does not perform work, internal energy change equals heat added to the system ( $dU = dQ$ ).
- If the process followed by the system  $\Gamma$  does not perform useful work, enthalpy change equals heat added to the system ( $dH = dQ$ ).
- If the system undergoes an adiabatic process:
  - Total work equals internal energy decrease ( $dW = -dU$ ).
  - Total useful work equals enthalpy decrease ( $dW_u = -dH$ ).

Equation 1.6 evidences that the entropy gain of the system  $dS$  comes about due to the heat added to the system at constant temperature  $T$  plus a certain amount of unbalanced entropy change  $dS'$ . As stated originally by Clausius, this unbalanced entropy change is either positive in the case of an irreversible process, or null in the case of a reversible process [20]. Again, the combination of equations 1.1–1.3 and the First and Second Laws provide the following useful interpretations:

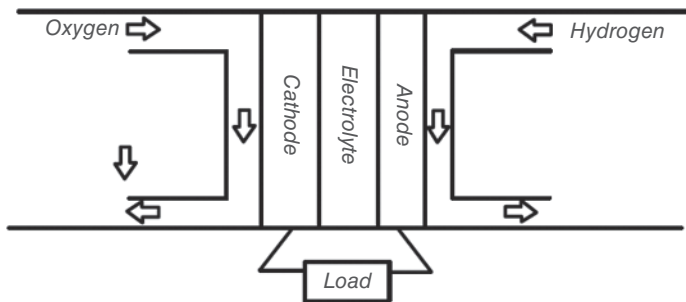
- If the system undergoes an isentropic process:
  - The total work done by the system equals the internal energy drop minus the energy dissipated to the surroundings ( $dW = -dU - TdS'$ ).
  - The useful work done by the system equals the enthalpy drop minus the energy dissipated to the surroundings ( $dW_u = -dH - TdS'$ ).
- If the system undergoes an isothermal process:
  - The total work done by the system equals the Helmholtz free energy drop minus the energy dissipated to the surroundings ( $dW = -dA - TdS'$ ).
  - The useful work done by the system equals the Gibbs free energy drop minus the energy dissipated to the surroundings ( $dW_u = -dH - TdS'$ ).

This set of useful thermodynamic relations provides a means to calculate the total and useful work through the four Gibbs potentials in a variety of processes. These relations are summarized in Table 1.2 for clarity.

Despite a thermodynamic analysis utilizing the considerations above showing the promise of the experimental setup shown in Figure 1.2, the current generated is usually very small due to the unfavourable characteristics of the three-phase interface (contact area between the gas, the electrolyte, and the electrode) and the high ion-transport resistance of the electrolyte [21]. These issues have driven the development of fuel cell technology towards flat and porous electrodes with a thin layer of electrolyte, as shown in Figure 1.3. Layouts of this kind result in the contact area being maximized and the resistance kept to a minimum, increasing the current produced [21].

**Table 1.2** Useful relations based on work and heat potentials.

Process	$-dU$	$-dA$	$-dH$	$-dG$
$dW = 0$	$-dQ$			
$dW_u = 0$			$-dQ$	
$dQ = 0$	$dW$		$dW_u$	
$dS = 0$	$dW_{max}$		$dW_{u,max}$	
$dT = 0$		$dW_{max}$		$dW_{u,max}$



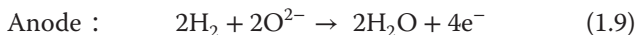
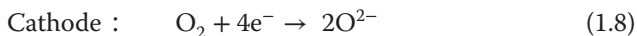
**Figure 1.3** Basic fuel cell arrangement. Source: Larminie *et al.* (2003) [21]. Reproduced with permission from John Wiley & Sons.

### 1.3.2 Solid Oxide Fuel Cells (SOFCs)

Solid oxide fuel cells (SOFCs), as the name implies, are completely solid-state entities that use ceramic electrolytes. The development of SOFCs can be traced back to the 1890s when Nernst discovered that stabilized zirconia ( $\text{ZrO}_2$ ) could conduct ions at certain temperatures, making zirconia a potentially useful electrolyte [21, 22]. Major manufacturers and their development focuses are listed in Table 1.3. A further investigation was carried out by Baur and Preis in 1943, showing that zirconia could serve as an oxygen-ion conducting electrolyte in fuel cells [22].

Today, this type of fuel cell is a high-temperature, solid-state electrochemical conversion device that produces electricity directly from electrochemical (oxidation) reactions. The cell operates at 600–1000°C where ionic conduction of oxygen ions takes place. Commonly, the anode is a Ni– $\text{ZrO}_2$  cermet and the cathode is Sr-doped  $\text{LaMnO}_3$ . Not using a liquid electrolyte avoids the attendant material corrosion and/or electrolyte management issues. The high temperature of the SOFC, nonetheless, poses stringent requirements on its materials, and hence the development of low-cost materials and the low-cost fabrication of ceramic structures that still fulfil the technical requirements are now the key technical challenges for the future utilization of SOFCs [17].

The basic operation of a SOFC is sketched in Figure 1.4. Oxygen ( $\text{O}_2$ ) is reduced at the cathode–electrolyte interface forming oxygen ions ( $\text{O}^-$ ) that are transported to the electrode through the electrolyte. Once at the interface between anode and electrolyte, these react with the hydrogen ions ( $\text{H}^+$ ) to form water ( $\text{H}_2\text{O}$ ) that is disposed of via the exhaust stream. Electrons are released at the anode and flow through the external load to the cathode, where they are used to reduce the oxygen molecules. This operating principle is summarized in Figure 1.4 and through the following reactions [23–25] (Eqs 1.8–1.10):

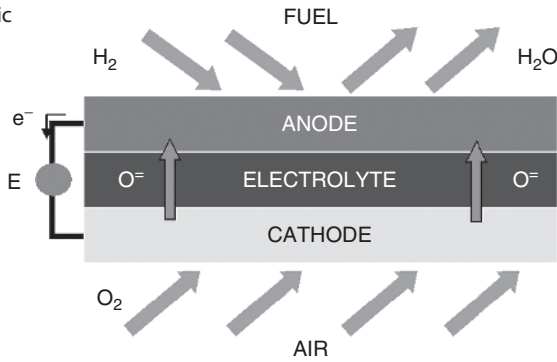


Owing to the overriding influence of the reaction shown in Equation 1.9, the reaction represented by Equation 1.10 is not

**Table 1.3** Major manufacturers of fuel cells.

<b>Manufacturer</b>	<b>Developments</b>
AFC Energy, Cranleigh, Surrey, United Kingdom (2006).	Alkaline fuel cells.
Apollo Energy Systems, Pompano Beach, Florida, USA (1966).	Develops, produces and markets fuel cell power plants, electrical propulsion systems, and alternative energy generation equipment.
Ballard Power Systems, Burnaby, British Columbia, Canada (1979).	Designs, develops, and manufactures zero-emission proton-exchange-membrane fuel cells. Ballard Power Systems, Inc. is a global leader in PEM (proton exchange membrane) fuel cell technology.
Doosan Fuel Cell America, Sunnyvale, California, USA (HQ) (2003).	A fuel cell manufacturer focusing on the stationary fuel cell and small business markets.
Intelligent Energy, Loughborough, United Kingdom (2001).	Specialists in the development of proton exchange membrane (PEM) fuel cells for application in the automotive, consumer electronics and stationary power markets.
UTC Power, South Windsor, Connecticut (1958).	Produces/develops fuel cells for numerous applications including for use in space and submarines.
SOFCpower SpA 2006, Trentino, Italy.	For stationary applications with electrical power requirements below 6 kW.
Mitsubishi Heavy Industries (MHI), established in 1914, Tokyo.	First domestic operation of a combined-cycle system combining SOFC and a micro gas turbine. Maximum power output of up 200 from 21 kW.
Rolls-Royce Fuel Cell Systems (RRFCS), 2002, Loughborough, UK.	Stationary power generation, applications range from 250 kW to >1 MW.
Hexis AG, 1997, Switzerland.	For stationary applications with electrical power requirements below 10 kW.
Siemens-Westinghouse, in SOFC/mGT business for more than 3 decades, USA.	For stationary applications with electrical power requirements above 100 kW.

**Figure 1.4** Basic SOFC operation.



further considered in the analysis regarding the electrochemical reaction set. Thus the overall electrochemical reaction (Eq. 1.11) is obtained by adding equations 1.8 and 1.9:



### 1.3.2.1 Electrolyte

Currently, the majority of SOFC developers use electrolytes made of zirconia stabilized by a small amount of yttria (3, 8, or 10%), namely YSZ [17]. When the temperature is raised to more than 800°C, such electrolytes become good conductors of oxygen ions and show minimal electrical conductivity (i.e. transportation of electrons) [17–22]. As a main shortcoming, the very high operating temperature of the cell makes material selection very difficult, although the high temperature also provides an exploitable characteristic for hybrid systems.

### 1.3.2.2 Anode

Present SOFCs use anodes made from zirconia cermet (a mixture of ceramic and metal) [17, 21]. The metal used is nickel, chosen primarily due to its high electrical conductivity and stability under chemically reducing conditions [17, 21]. Nickel is preferred to platinum, the metal of choice in low-temperature fuel cells, due to its much lower cost, with only a small trade-off in transport properties. The zirconia used is both to inhibit the sintering of the metal particles and to provide a thermal expansion ratio close to that of the electrolyte. A well-designed

anode should have high electrical conductivity to allow the flow of current, adequate ionic conductivity such that ions come into contact with the fuel flow, and high activity for the electrochemical and fuel conditioning<sup>1</sup> reactions [17]. At the same time, the anode should have a high porosity (20–40%) to allow mass transport of gases [17, 21].

### 1.3.2.3 Cathode

Like the anode, the structure of the cathode has to be porous to facilitate mass transport. Today, strontium-doped lanthanum manganite is widely used in cathodes. Alternative materials include P-type conducting perovskite structures that display mixed ionic and electrical conductivity; these are specifically worthy of consideration for lower temperature operation (about 650°C) as voltage loss becomes significant for the other standard materials [17, 21].

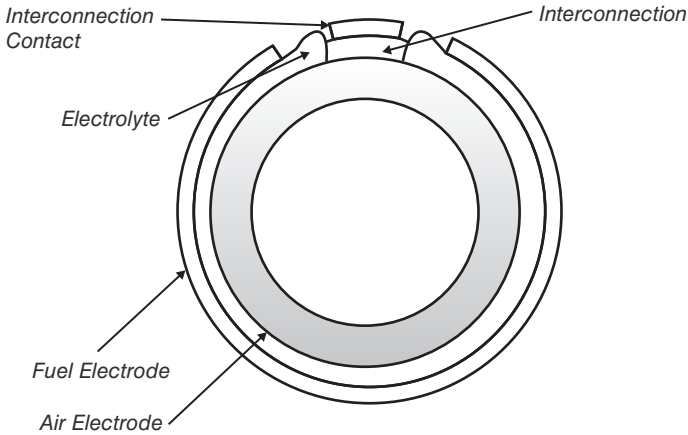
### 1.3.2.4 Interconnector

An interconnector exists to provide physical connection between neighbouring fuel cells so that a higher current can be produced by operating several fuel cells assembled in parallel. Under normal circumstances the only requirement for the interconnector material is that it has to have very high electrical conductivity. However, complications arise as SOFCs have a very high operating temperature. Problems such as different thermal expansion coefficients, cathode poisoning, and oxidation of metal become serious. For these reasons, at present, ceramic material (lanthanum chromite) is the favoured choice for tubular design SOFCs [17].

The components can be assembled together in various configurations (tubular, planar, etc.) each one of which exhibits advantages and disadvantages. As an example, Figure 1.5 illustrates a conventional cathode-supported tubular layout, which was dominant in SOFC technology in the late 1990s and early 2000s.

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<sup>1</sup> Fuel conditioning reactions refer to the reforming of natural gas and the water–gas shift, producing hydrogen from natural gas and carbon monoxide respectively. These two reactions produce the hydrogen that is eventually oxidized by the fuel cell.



**Figure 1.5** Tubular SOFC module configuration [17]. *Source:* US Department of Energy.

### 1.3.3 Fuel Cell Reactions

The operation of a fuel cell implies a number of electrochemical reactions employing hydrogen and oxygen as fuel and oxidant to generate power (even if some fuel cells are able to operate on different fuels, the main fuel for these reactors is still hydrogen). However, unlike oxygen, which can be easily obtained from air, hydrogen is not found naturally and it must be produced from other hydrogen-containing compounds, typically hydrocarbons and/or water. There are different ways to extract hydrogen from conventional hydrocarbon fuels, amongst which the normal industrial practice is to use fuel reforming [21, 26, 27]:

- 1) Water electrolysis.
- 2) Steam reforming.
- 3) Partial oxidation.
- 4) Auto thermal reforming.
- 5) Coal gasification.

### 1.3.4 Fuel Cell Performance

The performance of a fuel cell is often related to fuel cell voltage directly:

$$P = V \cdot I \quad (1.12)$$

$$P = V \cdot J \cdot A \quad (1.13)$$

where  $J$  refers to current density and  $A$  to fuel cell active area. Given that the latter parameter is determined during the design and manufacturing process, the voltage of a fuel cell during operation is affected by the variations in current density. Recalling the previous section on the thermodynamic principles of fuel cells, Gibbs free energy changes are used to define the work potential of the cell. In a fuel cell, the 'external work' involves driving the flow of electrons around an external circuit. Any work done by a change in volume between inlet and outlet is not harnessed by the fuel cell.

The reference point of zero energy in a fuel cell is normally defined as that of pure elements, in the normal state at standard temperature and pressure (25°C, 0.1 MPa) [21]. If this convention is adopted, then the term 'Gibbs free energy of formation',  $G_f$ , rather than the 'Gibbs free energy', is used (akin to the terms 'enthalpy of formation' rather than just 'enthalpy' [21]). For an ordinary hydrogen fuel cell operating at standard temperature and pressure (STP), this means that the Gibbs free energy of formation of the input is zero, which is a useful simplification [21]. This simplification (or standardization) is possible because it is the change in energy that is important, meaning that it is the change in this Gibbs free energy of formation,  $\Delta G_f$ , that gives the work potential of the fuel cell, that is, the difference between the Gibbs free energy of the products and the Gibbs free energy of the inputs or reactants [21]:

$$\Delta G_f = G_f \text{ of products} - G_f \text{ of reactants}$$

To make comparisons easier, it is convenient to consider these quantities in their molar-specific form. This is indicated by a line over the lower-case letter, for example,  $\bar{g}_{f,H_2O}$  is the molar-specific Gibbs free energy of formation for water [21].

The Gibbs potential is defined as:

$$G = H - TS \quad (1.14)$$

which is equivalent to:

$$\bar{g}_f = \bar{h}_f - T\bar{s} \quad (1.15)$$



For fuel cells, it is the change in Gibbs free energy that is responsible for the voltage induced. Therefore the following holds true:

$$\Delta \bar{g}_f = \Delta \bar{h}_f - T \Delta \bar{s} \quad (1.16)$$

Considering Equation 1.11, it can be observed that for every mole of hydrogen consumed ( $\text{H}_2$ ), one mole of oxygen ions is used ( $\text{O}^-$ ); one mole of water ( $\text{H}_2\text{O}$ ) and two moles of electrons ( $e^-$ ) are produced. It thus follows that the amount of electrons produced is then  $2 \cdot N$ ,  $N$  being the moles of hydrogen consumed; this is known as Faraday's law. Assuming one electron carries with it  $-e$  charge, the charge flow is  $-2 \cdot N \cdot (-e)$  which equals to  $-2F$ , where  $F$  is Faraday's constant which is defined as the electric charge of a mole of electrons.

Upon development of the electrochemical reactions, a voltage difference between electrodes is built up in the cell, which drives the flow of electrons from the cathode to the anode. On the assumption that there are no losses (the ideal case), this is called the Nernst potential and is denoted by  $E$ . The electrical work (joules) developed by an ideal fuel cell is then:

$$W_{el} = E \cdot I \cdot t = 2 \cdot F \cdot E \quad (1.17)$$

where  $I \cdot t$  is the flow of electrical charge passing through during a period of time  $t$ . This equation can now be linked to the change in Gibbs free energy through the information in Table 1.2:

$$\bar{g}_f = 2 \cdot F \cdot E \quad (1.18)$$

$$E = \frac{\bar{g}_f}{2 \cdot F} \quad (1.19)$$

This ideal potential of a cell operating at constant temperature ( $E$ ), which yields the maximum electrical work output that the cell can produce, is not attainable in practice. Indeed, the kinetics of the reactions, their activation energy, the limited mass diffusion rate of reactants (to get to the reaction sites) and products (to be evacuated from the reaction sites), and the limited electronic/ionic conductivity of the fuel cell constituents, introduce inefficiencies that manifest as voltage losses. These losses are explained in more detail later in this chapter. The impact of

pressure and temperature without these losses (i.e. on the ideal voltage of a fuel cell) are discussed in the next section.

### 1.3.5 Pressure and Concentration Effects

The foregoing discussion about the ideal cell potential  $E$  assumed the selection of a reference pressure and temperature (in this case  $25^\circ\text{C}$  and 1 atm), thus utilizing the standard cell potential  $E^0$ . However, in order to reduce the ionic resistivity of the electrolyte when integrated into hybrid systems, solid oxide fuel cells operate at a much higher temperature, and also typically operate at a higher pressure. This higher operating temperature and pressure means that the above expressions must be applied to Gibbs free energy changes at higher pressure and temperature or, more commonly, a correction must be applied to account for the effects of these two thermodynamic variables and of the concentration of reactants and products (whose proportion is not stoichiometric in a practical case). This correction is a mathematical expression of Le Chatelier's principle.

The correction for temperature can be introduced directly into the standard cell voltage  $E^0$ , which is evaluated at the operating temperature of the fuel cell. The corrections for pressure and composition are expressed as a complement to this. The resulting cell voltage is known as Nernst's potential and stands for the actual work potential of the fuel cell; that is, the voltage of a fuel cell operating at a given pressure, temperature and gas composition. A detailed derivation of the equation is not described here, but can be found in books and open literature [17, 21].

Considering the general reaction:



The activities of the reactants and products modify the Gibbs free energy change of the reaction. Using thermodynamic arguments [28], it can be shown that the general Nernst's equation related to the reaction can be expressed as:

$$\bar{g}_f = \bar{g}_f^0 + R \cdot T \ln \frac{P_J^j P_K^k}{P_M^m} \quad (1.21)$$

where  $\bar{g}_f^0$  is the change in molar Gibbs free energy of formation at standard pressure and the operating temperature of the fuel

cell. For the particular case of the hydrogen oxidation reaction, this equation has the following aspect:

$$\bar{g}_f = \bar{g}_f^0 + R \cdot T \ln \frac{P_{\text{H}_2} P_{\text{O}_2}^{1/2}}{P_{\text{H}_2\text{O}}} \quad (1.22)$$

The impact of the activities of reactants and products on the Gibbs free energy change is clear in Equation 1.22. The impact on Nernst's potential, and thus on electrical work, is analogous and can be easily assessed if Equation 1.21 is used, obtaining:

$$E = \frac{\bar{g}_f}{2 \cdot F} + \frac{R \cdot T}{2 \cdot F} \ln \frac{P_{\text{H}_2} P_{\text{O}_2}^{1/2}}{P_{\text{H}_2\text{O}}} = E^0 + \frac{R \cdot T}{2 \cdot F} \ln \frac{P_{\text{H}_2} P_{\text{O}_2}^{1/2}}{P_{\text{H}_2\text{O}}} \quad (1.23)$$

where the partial pressure of each species is calculated through their corresponding molar fraction according to Dalton's law. If  $\alpha$ ,  $\beta$  and  $\gamma$  are the molar fractions of hydrogen and steam in the anode and oxygen in the cathode, and  $P_{an}$  and  $P_{ca}$  are the absolute pressures of the anode and cathode respectively:

$$P_{\text{H}_2} = \alpha \cdot P_{an}; \quad P_{\text{H}_2\text{O}} = \beta \cdot P_{an}; \quad P_{\text{O}_2} = \gamma \cdot P_{ca} \quad (1.24)$$

These expressions can be introduced into Equation 1.23 to calculate Nernst's potential. With the assumption that the absolute pressures in the cathode and anode are the same (which is a reasonable assumption based on mechanical considerations), this yields to:

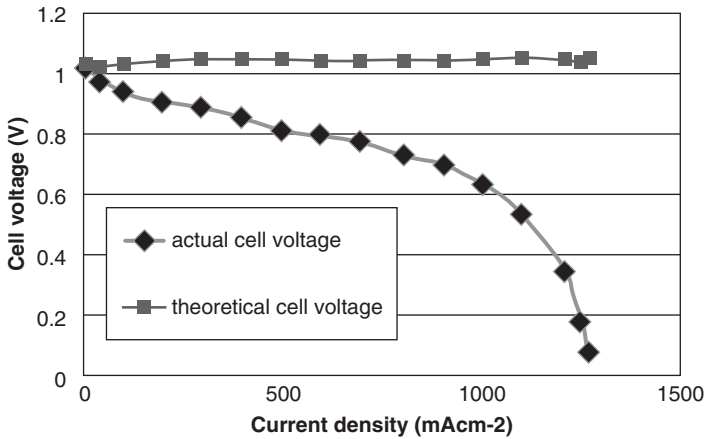
$$E = E^0 + \frac{R \cdot T}{2 \cdot F} \ln \left( \frac{\alpha \cdot \beta^{1/2}}{\gamma} P_{cell}^{1/2} \right) \quad (1.25)$$

$$E = E^0 + \frac{R \cdot T}{2 \cdot F} \ln \left( \frac{\alpha \cdot \beta^{1/2}}{\gamma} \right) + \frac{R \cdot T}{4 \cdot F} \ln(P_{cell}) \quad (1.26)$$

where the effects of temperature (through  $E^0$ ), composition and pressure on Nernst's potential can clearly be seen.

### 1.3.6 Irreversibilities in Fuel Cells

The performance of a SOFC operating at 800°C under atmospheric pressure is shown in Figure 1.6. The ideal Nernst's



**Figure 1.6** SOFC performance at 800°C under atmospheric pressure (graph generated using expression provided in [17]). *Source:* US Department of Energy.

potential is shown by the square symbols and associated line, whilst the diamond shapes represent the actual voltage differences between electrodes. It is observed that the latter is substantially lower than the theoretical limit, indicating that fundamental losses arise under normal operating conditions. As plotted in Figure 1.6, there is a rapid initial fall of cell voltage, followed by a slow and linear decrease. A final sudden decrease is also present at high current density.

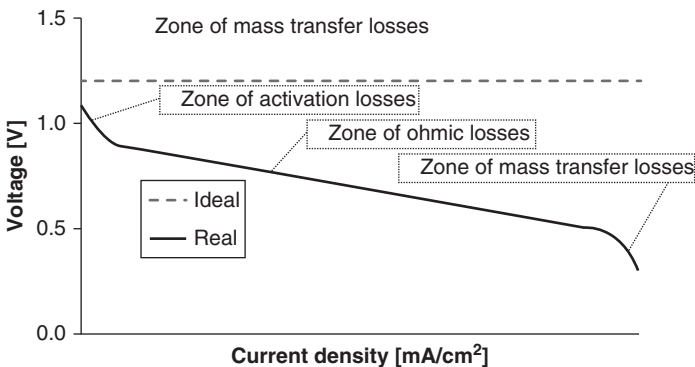
There are several reasons for this voltage drop with respect to the ideal value. The four most important are: activation losses; fuel crossover and internal currents; ohmic losses; and mass diffusion or concentration loss. The effects of the four types of losses are often easy to distinguish in a V–I diagram, especially for low-temperature fuel cells, as illustrated in Figure 1.7 [17]. A brief discussion of the relevant losses is given in the following paragraphs, while interested readers are directed to accessible government-contracted reports and other documents for more details on how these losses are described numerically [17, 21].

- *Activation-related losses:* these stem from the activation energy of the electrochemical reactions at the electrodes. These losses are primarily due to the sluggishness of

the reactions at hand, the electrocatalyst material and microstructure, reactant activities (and hence utilization), and are influenced very weakly by current density [17, 21].

- *Fuel crossover and internal currents*: these arise as fuel and electrons migrate through the electrolyte, due to the non-ideal nature of this element. For example, in a hydrogen fuel cell, electrons proceeding through the electrolyte instead of through the external circuit do not contribute to the production of electrical work.
- *Ohmic losses*: ohmic losses are brought about by ionic resistances in the electrolyte and electrodes, electronic resistance in the electrodes, current collectors and interconnects, and contact resistances. Ohmic losses are proportional to the current density, and depend upon the material selection, stack geometry and operating temperature [17, 21].
- *Mass-transport-related losses*: these are due to the limited mass diffusion rates of reactants and products at high current density conditions [17, 21].

An overview of the physics behind each loss, further to the descriptions above, is now provided. Reactions occurring on the electrodes are not instantaneous and require both a finite time to proceed and an initial threshold voltage to be met in order to overcome the activation energy of the reaction. Losses associated with these factors are known as activation



**Figure 1.7** Ideal and actual fuel cell voltage/current characteristics [17].  
Source: US Department of Energy.

losses and, for the cited reasons, the current density has little to no influence on the size of these losses. The electrolyte in fuel cells is designed to allow the flow of ions only [28, 29]. Nevertheless, in practice, due to the non-ideal nature of this element, fuel crossover and internal currents arise as fuel and electrons migrate through the electrolyte. For example, in a hydrogen fuel cell, electrons proceeding through the electrolyte instead of through the external circuit do not contribute to the production of electrical work. Moreover, according to Faraday's law, when a hydrogen molecule flows through the electrolyte, two moles of electrons are wasted corresponding to the anodic half-reaction (Equation 1.7) that fails to take place. Of all the losses, ohmic losses are the easiest to comprehend. All physical components of a fuel cell have finite conductivities, meaning each pose certain conductive resistances impeding the free flow of electrical charge carriers. Therefore, when ionic flow passes through the electrolyte or when electrons migrate past the electrodes and interconnectors, linear voltage drops occur according to Ohm's law.

As previously mentioned, fuel cells need oxygen and hydrogen as reactants to produce electrical current. The primary source of oxygen is air, resulting in the normal practice of adopting atmospheric air to provide the oxidant. This air stream is typically inducted to the cathode [28]. As previously discussed, hydrogen is generally produced through fuel reforming which, in the case of high-temperature fuel cells, is typically carried out in the anode (at least partially) in the presence of sufficient steam in order to prevent carbon deposition on the electrode surface [28]. As a result, the anodic flow will typically include hydrogen and additionally some carbon monoxide, carbon dioxide (coming from the reforming and water-gas shift reactions) and steam. Thus the flows at both electrodes are often not pure hydrogen or oxygen. The reactant gases have to diffuse through stagnant non-reacting gases and the electrode matrix structures to replenish the reactant gases being used on the electrode surface (the three-phase interface). This brings about regions of lower partial pressure due to the limited diffusion rate from the core flow to the reaction sites and, for the case of steam, vice versa. These lower partial pressures of reactants and higher partial pressure of products (where not adequately

evacuated from the reaction sites) cause a voltage drop as deduced from Equation 1.23. This is termed mass transport loss or concentration loss [17, 18].

### **1.3.7 Fuel Cell Applications**

Fuel cell technologies can be grouped into three primary areas: portable power generation, stationary power generation, and power for transportation. Further categorizations may be made according to fuel type and associated infrastructure. These categorizations are particularly important relating to the production, distribution, storage and dispensing of the fuels, crucial factors in implementing fuel cell technology. As a result of the inherent size flexibility of fuel cells, the technology may be used in applications with a broad range of power and plant size requirements. Fuel cells have application ranges from systems of a few watts to megawatts. Mobile fuel cell applications primarily include transportation systems and portable electronic equipment, while stationary applications primarily include combined heat and power systems for both residential and commercial needs [30, 31].

#### **1.3.7.1 Transportation Applications**

Nearly all leading car manufacturers have designed and developed at least one prototype vehicle using fuel cells [30, 31]. Manufacturers such as Toyota and Ford have chosen to feed the fuel cell with methanol, whilst others have used pure hydrogen – Opel using liquid hydrogen and General Motors storing hydrogen in hydride metals. There is a general short-term trend for auto manufacturers to use fuel cells using reformed methanol. However, as evidenced in the open literature, hydrogen still remains the fuel of choice in fuel cells for the majority of car manufacturers, mainly due to the inherent simplification of the fuel processing stage. The auto manufacturers' interest in fuel cells is not a new development. Since 1994, Daimler-Benz working in agreement with Ballard built a series of PEMFC powered cars, the first of which was fuelled with hydrogen, and in 1997 Daimler-Benz released a methanol-fuelled car with a 640 km range [30].

Furthermore, in 1993, Ballard Power Systems demonstrated a 10-metre light-duty transit bus with a 120 kW fuel cell system,

followed by a 200 kW, 12-metre heavy-duty transit bus in 1995. These buses use no traction batteries and operate using compressed hydrogen as the onboard fuel (which is a similar technology to that used in buses running on compressed natural gas stored over the roof in high pressure cylinders) [30].

#### **1.3.7.2 Portable Electronic Equipment**

In addition to large-scale power production, miniature fuel cells can be profitably/potentially employed as batteries to power consumer electronic products such as cellular telephones, portable computers, and video cameras [30, 31]. Small fuel cells could also be used to power telecommunications satellites, augmenting solar panel performance, whereas micro-machined fuel cells could provide power to computer chips. Finally, micro/nano fuel cells could safely produce power for biological applications, such as hearing aids and pacemakers [30]. Contrary to transportation applications where much development has been required to bring systems incorporating fuel cells to the level of maturity where they can provide a credible alternative to internal combustion engines in producing a mechanical output, in portable electronic equipment, fuel cells compete with devices such as batteries in directly producing an electrical output [30].

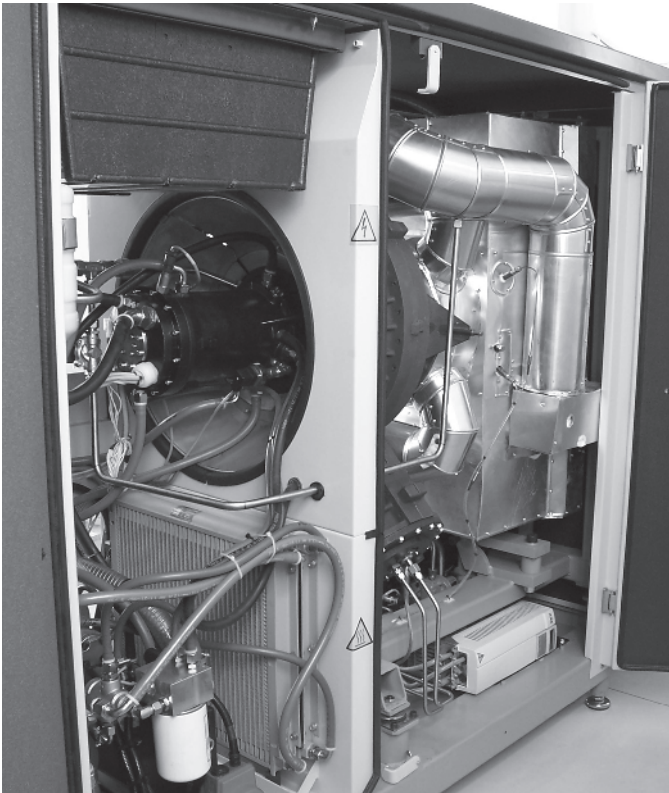
## **1.4 Gas Turbines**

Gas turbines (normally microturbines) are essential components to obtain high efficiency in hybrid systems. Their main components include an upstream compressor coupled to a downstream turbine, with a combustion chamber lying in-between. In small devices called microturbines, heat exchangers are also common in order to reduce fuel consumption. Finally, an electric generator converts the shaft work into electricity. A picture of the Turbec T100 microturbine (100 kW<sub>e</sub>) is shown in Figure 1.8.

### **1.4.1 Background of Gas Turbines**

The gas turbine was hailed as one of the greatest discoveries of the twentieth century and has, since the 1940s, been used for numerous applications from power generation to





**Figure 1.8** A Turbec T100 microturbine.

aircraft propulsion [32] due to its high reliability, efficiency, and cost-effectiveness. Further details related to gas turbines, specifically small-sized machines (called microturbines) based on the recuperated Brayton cycle and vitally important in hybrid system development, may be found in Chapter 3.

## **1.5 Coupling of Microturbines with Fuel Cells to Obtain ‘Hybrid Systems’**

The integration of a microturbine with a fuel cell to provide a power generation system with very high efficiency is generally

termed as 'hybrid'. Combining the two distinct systems increases the efficiency with respect to using a microturbine alone, from values between 25% and 30% to values close to 65% (low heating value (LHV) fuel to electricity). This increase in efficiency is due to the contribution of the fuel cell to power generation which is based on electrochemical energy conversion and thus not subjected to the limitations of the Carnot cycle. In a typical high-temperature hybrid system based on a SOFC and microturbine, such as those in [32–35], the air stream is first pressurized through a compressor and then supplied to the SOFC where fuel is added and the electrochemical conversion of chemical energy into electrical work takes place. The high-temperature exhaust stream from the fuel cell is then discharged into an expander which provides work to drive the compressor and electric generator. The operation of the cell at high pressure (corresponding to a higher voltage) and the additional work produced by the gas turbine are significant upgrades to a standard atmospheric pressure fuel cell. This twofold enhancement significantly raises system efficiency [32].

On the negative side, these integrated systems (fuel cells and micro gas turbines) have a number of critical design constraints/limitations that require serious operational and developmental consideration, both in stationary and transient operation, which will be explored in later chapters.

Examples of companies currently engaged in the development of hybrid system prototypes are Siemens-Westinghouse, Rolls-Royce Fuel Cell Systems and Mitsubishi Heavy Industries. Westinghouse has been engaged in SOFC development for more than three decades. Following a merger in 1998 the company later became Siemens-Westinghouse Power Corporation (SWPC), focusing on the research and development (R&D) of SOFC-mGT hybrid systems for the emerging distributed power market. This company developed a full scale 100 kW plant (without an integrated micro gas turbine) which operated for more than 15,000 hours. This plant met many of the testing targets, although the measured performance values were significantly lower than the potential targets of these plants [33]. Other improved plants over the years have been designed, developed and tested, as mentioned in the rest of the book.

**Table 1.4** Overview of some groups performing research in SOFC and SOFC-mGT hybrid systems [32, 36].

<b>Institution</b>	<b>Location</b>	<b>Main staff</b>	<b>Models</b>	<b>Current focuses</b>
NETL/D.O.E.	Morgantown, USA	Tucker, D. Shelton, M.	Transient model	Dynamics, control, start-up emulator
Siemens-Westinghouse	Erlangen, Germany	Hussmann, K. Ciesar, J.A.	Tubular SOFCs	Part-load, dynamics, control strategy, cycle layout and design
Rolls-Royce Fuel Cell Systems	Derby, UK	Agnew, G. Bozzolo, M.	IP-SOFC (RR) Hybrid systems	Part-load, dynamics, control strategy, cycle layout and design
German Aerospace Center (DLR), Institute of Combustion Technology	Stuttgart, Germany	Hohloch, M. Widenhorn, A. Lebküchner, D. Panne, T. Aigner, M.	Tubular SOFCs	Cycle layout and design emulators
Advanced Power and Energy Program (APEP) at the University of California, Irvine	California, Irvine, USA	Samuelsen, S. Brouwer, J. Rao, A.D. Yi, Y.	Tubular SOFCs	Part-load, dynamics, control strategy, cycle layout and design
TPG	Genova, Italy	Massardo, A.F. Costamagna, P. Magistri, L. Traverso, A. Ferrari, M.L.	Tubular SOFCs IP-SOFC (RR) Hybrid systems	Part-load, dynamics, control strategy, cycle layout and design

**Table 1.4** (Continued)

<b>Institution</b>	<b>Location</b>	<b>Main staff</b>	<b>Models</b>	<b>Current focuses</b>
Politecnico di Milano	Milano, Italy	Campanari, S. Iora, P.	Tubular SOFCs	Cycle design, part-load
VOK-LTH	Lund, Sweden	Assadi, M. Torisson, T. Selimovic, A. Kemm, M. Hildebrandt, A.	Planar SOFC, detailed compressor modelling	Dynamics, control, compressor surge modelling
SNU	Seoul, Korea	Song, T.W. Sohn, J.L.	Tubular SOFCs	Cycle layout and design
NTNU-EPT	Trondheim, Norway	Bolland, O. Thorud, B. Stiller, C.	Tubular SOFCs	Part-load, dynamics, control
NTNU-ITK	Trondheim, Norway	Imsland, L. Kandepu, R.	Lumped model	Dynamics, control
School of Automation, University of Electronic Science and Technology of China and Institute of Fuel Cell, Shanghai Jiao Tong University, Shanghai, China	China	Wua, X. Zhub, X.	Hybrid systems	Part-load, dynamics, control strategy, cycle layout and design

*Source:* Adapted from Damo (2016) and Stiller (2006).

### 1.5.1 Active Hybrid Systems Research Groups

Due to the importance of the related research activities in this field, Table 1.4 lists some research groups that are actively working in the field of hybrid system modelling and contributing to this field of knowledge in the form of publications based on experimental activities and/or theoretical development/analysis [32, 36].

## 1.6 Conclusions

Hybrid systems are expected to play a large role in the improvement of power generation systems. Manufacturers and researchers are devoting special efforts to developing this technology and bridging the gap to the market. However, at the moment these systems are not ready for commercialization due to both technical problems that are yet to be adequately solved, and the high cost of materials used to produce the fuel cells and related components. Hence, research and development activities including theoretical investigations and, primarily, experimental work are of critical importance.

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