Part I
Technology

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Technical Advancement of Fuel-Cell Research and Development

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1.1
Introduction

The world energy demand is growing at a rate of 1.8% per year. As a consequence of increasing industrialization, it has now shifted to today's developing countries. Since the higher demand is largely met with the fossil fuel reserves that are also responsible for emissions of greenhouse gases (GHGs) and other pollutants, emissions from developing countries may account for more than half of the global CO_2 emissions by 2030. The industrialized countries should therefore take the challenge to lead the way towards the development of new energy systems. This requires a comprehensive energy strategy that takes into account the entire cycle from development to supply, distribution, and storage in addition to conversion. It also includes considering the impact on the producers and users of energy systems. Short- and long-term goals to be addressed are greater energy efficiency and better integration of renewable energy sources. On this path characterized by technical developments, as an efficient and clean technology, fuel cells can make a substantial contribution. In the long term, alongside electricity, hydrogen will be a major energy vector.

A sustainable energy supply that is largely CO_2 free and based on electricity and hydrogen will be supplemented by fuel cells, which convert energy very efficiently. Since fuel-cell systems run very quietly and deliver high-quality electricity, they are particularly suitable for application in sensitive and sophisticated applications, such as in hospitals, IT centers, and vehicles. The efficiency of fuel cells, which rises with decreasing load, is nearly independent of system size and has proven to reduce energy consumption and regulated emissions significantly when used for vehicle propulsion. Even if conventional fuels such as diesel or natural gas are used, energy can be saved and emissions reduced in combination with reformers for mobile on-board power supply and decentralized energy supply. Fuel cells have the potential to convert hydrogen and other fuels into electricity very efficiently, producing negligible pollution. Furthermore, they are sufficiently flexible to be adapted to the different intermittent renewable energy sources that will enrich the

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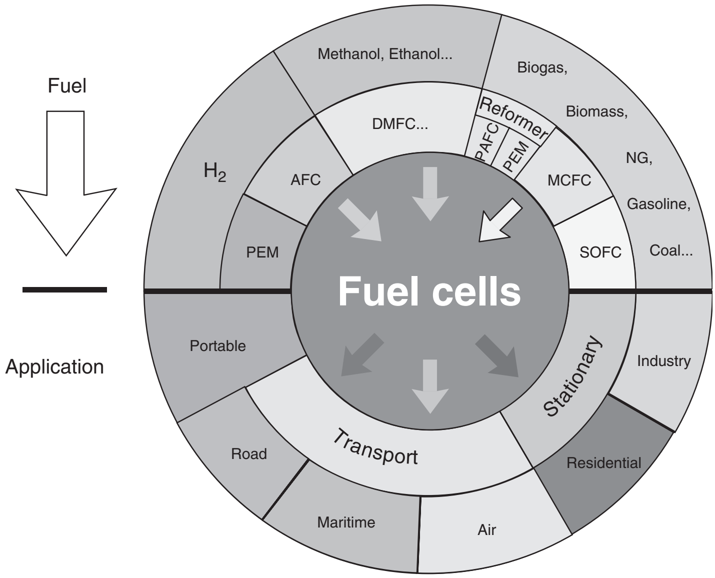

Figure 1.1 Fuel-cell technologies, possible fuels, and applications [1].

energy mix in the future. The numerous possible energy carriers, from solids (e.g., coal and biomass) and liquids (diesel, methanol, and ethanol) to gases (e.g., natural gas, biogas, and hydrogen) in combination with proven fuel-cell technologies shown in Figure 1.1 can be used in all those fields of application requiring a stable power supply. Fuel-cell systems conditioned in different ways satisfy power requirements from a few watts for portable 4C applications to the megawatt range for stationary applications such as decentralized combined heat and power (CHP) generation.

Global funding initiatives for research, development, and demonstration accompany the already great efforts of industry, and support fuel-cell technology with regard to the complex replacement processes required for capturing future markets.

1.2 Representative Research Findings for SOFCs

Two main concepts for solid oxide fuel cells (SOFCs) are currently under development: the tubular and the planar designs. In terms of long-term stability, the tubular concept has demonstrated the best results, while the planar design promises higher power densities.

1.2.1 Tubular Concepts

The standard tubular design is based on a porous cathode tube, of which a part is coated with a ceramic interconnect as a vertical stripe along the tube. The remaining surface of the cathode tube is coated with a thin electrolyte, where the overlapping with the interconnect is the critical part concerning gas tightness. The electrolyte is coated with the anode material. The Japanese company TOTO started to use this standard tubular design in 1989. TOTO invented cheap manufacturing technologies, called the TOTO wet process, based on slurry coating and sintering [2]. It uses tubes with a length of 0.66 m and an external diameter of 16.5 mm. Fuel gas is supplied to the outside of the cell while air is supplied to the inside via a thin ceramic tube, the so-called air supply tube. The cathode consists of lanthanum–strontium–manganese, the interconnect is made of lanthanum–calcium–chromate, the electrolyte of ScSZ, and the anode of Ni/YSZ. These cells can attain power densities of up to 330 mW cm^{-2} [3]. Twelve tubes are connected with nickel materials in a 2×6 arrangement (2 in parallel, 6 in series) to form bundles or stacks. The current path along the circumference of the tubes causes a high internal resistance, which limits the power density. As a result of high cathode polarization, an operating temperature of 900–1000 °C is necessary in order to achieve high power density (HPD).

The tubular concept of Siemens (derived from the activities of Westinghouse, already started in the 1970s) was based on a porous lanthanum–calcium–manganese cathode tube with a wall thickness of 2.2 mm and a length of 1.8 m, of which 1.5 m can be utilized electrically. A lanthanum–calcium–chromate interconnect, which serves to carry power away from the cathode, is deposited as a stripe on this tube by atmospheric plasma spraying (APS). A YSZ electrolyte layer is then sprayed on to the rest of the tube by means of APS and sintered until it is gas-tight. In a final step, the anode (Ni/YSZ) is also applied by means of APS [4]. The tubes are connected to form bundles using nickel felt. The operating temperature is in the range 950–1000 °C in order to achieve the required power density of ~200 mW cm^{-2}. In order to overcome the problem of high ohmic resistance of the tubular design, Siemens developed a modified concept using flattened tubes with internal ribs for reduced internal resistance (HPD tubes). A similar design, albeit anode supported, is being developed by the Japanese company Kyocera [5] and the Korean research institution KIER [6]. Siemens was also working on another design variant known as the Delta 9 design, which makes further increases in power density possible. Based on in-house analyses of the cost reduction potential of the tubular design and derived designs, Siemens abandoned this development work in late 2010 [7].

Another type of tubular cells uses the anode as the tube material. The US company Acumentrics develops anode-supported tubes with a length of 45 cm and an external diameter of 15 mm [8].

A different tubular design is being pursued in Japan by Mitsubishi Heavy Industries (MHI). The single cells are positioned on a central porous support tube and electrically connected in series using ceramic interconnect rings. This leads to an increased voltage at the terminals of the individual tubes. The fuel is fed into the inside of the tube and air is supplied to the exterior [2, 9]. The maximum tube length is 1.5 m with an external diameter of 28 mm. With these specifications, power densities of up to 325 mW cm^{-2} at 900 °C have been reported [10].

1.2.2
Planar Designs

Planar designs can be broken down into electrolyte-supported and electrode-supported designs. The former uses the electrolyte to stabilize the cell mechanically. The electrolyte is 100–200 μm thick for a cell area in the range 10×10 cm^2. Owing to the comparatively high ohmic resistance of the thick electrolyte, typical operating temperatures of this design are 850–1000 °C. For operation at very high temperatures, ceramic interconnects made of lanthanum–chromate are preferentially used. There is an obvious trend towards metallic interconnects, as these ceramic plates are restricted in size, require high sintering temperatures, have different thermal expansion behavior in oxidizing and reducing atmospheres, and have comparatively low electrical and thermal conductivities. The advantage of ceramic plates is the low level of corrosion and therefore low degradation of the contacts, which sustains the interest in this material. The metallic interconnects allow (and also demand) a reduction in operating temperature and make the manufacture of larger interconnect plates possible. The high thermal conductivity reduces the temperature gradients in the stack and allows greater temperature differences between the gas inlet and outlet, reducing the amount of air required for cooling. As the thermal expansion coefficient of conventional high-temperature alloys is much higher than that of zirconia, a special alloy referred to as CFY (chromium with 5% iron and 1% yttrium oxide) was jointly developed by the Austrian company Plansee and Siemens. This alloy is used by different companies throughout the world for their stacks, including Hexis (formerly Sulzer Hexis) and Fraunhofer IKTS in Dresden, Germany, and also Bloom Energy.

When Siemens discontinued its planar activities, Fraunhofer IKTS took over a large proportion of the existing know-how and has been systematically refining the technology. Cells are being developed in close cooperation with Kerafol, a company which has also been working closely together with H.C. Starck – another cell manufacturer in Germany – in the area of electrolyte–substrate cell production since 2009.

In the Hexis design, fuel is supplied to the center of the electrolyte-supported circular cell (diameter 120 mm), from where it flows to the outer rim of the cell. Here, the fuel that has not reacted within the cell is burned. Air is supplied from the outside and heats up as it flows towards the center of the cell. It then flows back outside the cell in parallel with the fuel. The stack is typically operated at 900 °C. Between 50 and 70 cells are stacked together, generating a power of 1.1 kW [11]. In order to reduce manufacturing costs, Hexis has since altered the two-layer interconnect design to a one-plate concept [12].

Similar designs are also used by the Japanese companies Kyocera, Mitsubishi Materials Corporation (MMC), Nippon Telegraph and Telephone. (NTT), and Toho Gas. Fraunhofer IKTS and Bloom Energy both use conventional cross-flow

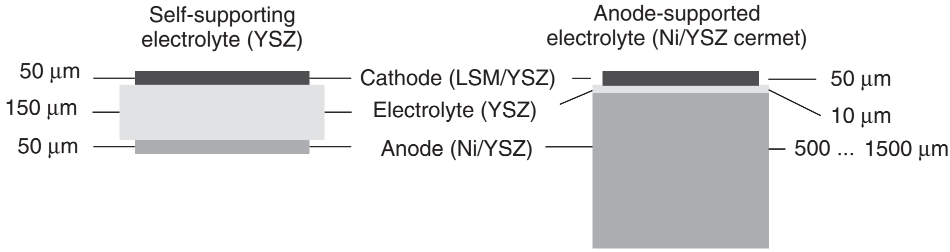

Figure 1.2 Anode substrate (right) in comparison with electrolyte substrate (left).

designs with electrolyte-supported cells soldered on to CFY interconnects. A joint development by MHI and Chubu Electric Power is the MOLB (mono-block layer built) design. Cells up to a size of 20 × 20 cm^2 are manufactured. They are based on a corrugated electrolyte layer. The electrolyte thus also contains the gas channels. This simplifies the design of the interconnect, allowing planar ceramic plates to be used. The largest stack of this type was built from 40 layers and delivered 2.5 kW at 1000 °C [13]. In 2005, MHI began testing cells measuring 40 × 40 cm^2 in 10-layer stacks as a basis for increased system output [14].

Since electrolyte resistance is the most significant obstacle to further decreasing the operating temperature, manufacturing thinner electrolytes constitutes a major challenge. This challenge can be overcome by shifting the function of mechanical stabilization from the electrolyte to one of the electrodes. For this concept, the anode tends to be preferred because it exhibits much better electrical conductivity. Therefore, no increase in ohmic resistance occurs when the electrode thickness is increased (see Figure 1.2). Nickel cermet also has good mechanical stability, which allows larger cells to be produced.

When Forschungszentrum Jülich began working on the development of this concept in 1993, it was one of the first institutions to do so. Since then, many developers throughout the world have come to regard this concept as the next generation of SOFCs. It allows the operating temperature to be reduced to between 650 and 800 °C while retaining and even surpassing the power density of electrolyte-supported cells operated at 900 °C. At the same time, this design allows cheaper ferritic chromium alloys to be used for the interconnects because their thermal expansion coefficient corresponds to that of the anode substrate.

At Forschungszentrum Jülich, anode substrates with a thickness of between 1 and 1.5 mm are manufactured by warm pressing. The electrolyte with a thickness of 5–10 µm is deposited on the substrate by means of vacuum slip casting. The stack design is based on a co-flow or counter-flow arrangement. The latter is favored for operation on natural gas with internal reforming. A 60-layer stack delivered 11.9 kW at a maximum temperature of 800 °C (average temperature in the stack ~700 °C) when operated on methane with internal reforming [15].

Similar concepts have been developed, for example, by Versa Power Systems (VPS) in Canada, Delphi and PNNL in the USA, and Topsøe Fuel Cells and Risø

National Laboratory in Denmark. In Germany, the companies H.C. Starck and CeramTec manufacture these anode-supported cells. Most of these institutions have also developed concepts using pure metal substrates instead of the anode cermet, to improve mechanical and redox stability.

A completely different design has been developed by Rolls Royce. Short electrode and electrolyte strips are applied to a porous, flat ceramic substrate. These single cells are connected electrically in series using ceramic interconnect strips, which leads to a high voltage output of one unit at a low current. Fuel gas is supplied to the inside of the supporting substrate and air to the outside. The operating temperature is about 950 °C [16]. Kyocera together with Tokyo Gas are developing a similar concept [17].

At DLR in Stuttgart, Germany, a concept in which all cell layers are produced by means of plasma spraying processes was developed in the mid-1990s. The cells are based on a metal substrate which promises to be more resistant to oxidation than the nickel-based anode substrate. Even though the power densities were increased in the last few years, they are still considerably below the values achieved for the anode substrates [18, 19].

1.2.3
Actors and Major Areas of Development

In the late 1990s, some of the most important developers in Europe, Daimler-Benz/Dornier and Siemens, discontinued their activities on planar SOFCs. After an interim phase, the number of companies and research facilities involved in SOFC development has increased again (Table 1.1). The planar technology is being developed further at research institutions such as Forschungszentrum Jülich, DLR in Stuttgart, and Fraunhofer IKTS in Dresden (all of them had already cooperated with Siemens and Dornier in individual fields in the 1990s) and at companies such as Staxera in Germany and Topsøe Fuel Cells in Denmark.

During the last two decades of the last century, Westinghouse (since 1998 Siemens) dominated developments in the USA. Since the Solid State Energy Conversion Alliance (SECA) program started, the situation has changed. Various activities in the field of planar SOFCs have been restarted or expanded, and some new consortia were founded. In its second phase, the SECA program is focusing on the development of power generation technology for a cost-effective, highly efficient central power station (>100 MW_{el}). The industry teams involved are Fuel Cell Energy (FCE) and VPS, UTC Power and Delphi, and Rolls-Royce, assisted by numerous research institutions [7]. A tremendous development took place at a new company, Bloom Energy, whose activities are partly based on those of Ion America, taken over by Bloom Energy. As of the end of 2011, Bloom Energy has sold more than 80 systems with a nominal power of 100 kW. They employ more people than all other developers in North America together.

During the 1990s in Japan, more than 10 companies were engaged in planar SOFC development. Because the goals of the NEDO "Sunshine" project could not

Table 1.1 The most important SOFC developments worldwide.

Continent	Facilities/ employees	Designs	Development focus[a]	
Europe	Industrial enterprises 17	Planar design	(1) Systems	12
		Anode substrate	Stacks	12
		Electrolyte substrate	Cells	11
	Research facilities 6	Metallic substrate	(2) Materials	9
		Porous ceramic substrate	(3) Fabrication	3
			Powders	2
	Employees 750–850	CGO electrolyte for 550 °C	System components	2
			Fuel processing	2
		Metallic interconnect	Interconnects	1
			Reformers	1
			Cell and stack testing	1
			Stack and system testing	1
			Modeling	1
North America	Industrial enterprises 12	Planar design	(1) Cells	16
		Tubular design	Stacks	14
		Microtubes	(2) Systems	9
	Research facilities 5	Anode substrate	Materials	6
		Electrolyte substrate	(3) Systems (low power)	3
		Metallic substrate	Modeling	2
	Employees >2000	Metallic interconnect	System testing	1
		Ceramic interconnect		
Asia and Australia	Industrial enterprises 14	Planar design	(1) Cells	17
		Tubular design	Stacks	16
		Microtubes	Systems	13
	Research facilities 4	Anode substrate	Materials	13
		Electrolyte substrate	(2) Systems (low power)	3
		Flat tubes	Systems (pressurized)	1
	Employees 600–750	Metallic interconnect		
		Ceramic interconnect		

[a] From most to least frequent.

be achieved completely, a reorientation took place and other companies started SOFC development. A demonstrative research project on small systems was started in 2007. By the end of 2011, more than 130 units in the range 0.7–8 kW had been installed based on different stack concepts, developed by TOTO, MHI, MMC, Kansai Electric Power Company (KEPCO), Kyocera, and Tokyo Gas [9].

1.2.4
State of Cell and Stack Developments

The field of cell and stack development comprises numerous activities. This makes it difficult to provide an overview, particularly one that is based on comparable operating conditions. There are three different types of cells:

- anode-supported cells at an operating temperature of 750 °C
- electrolyte-supported cells at 800–900 °C
- tubular cells at 900–1000 °C.

Table 1.2 lists the results achieved in terms of cell power density (at a cell voltage of 0.7 V), active cell area, degradation rates, duration of relevant long-term measurements carried out, and the power of the constructed stacks.

Although the different operating conditions and fuels used prevent direct comparisons, it is obvious that the highest energy densities are achieved with anode-supported cells, preferably with lanthanum strontium cobalt ferrite (LSCF) cathodes, although a number of tubular designs have clearly improved over the last few years. In addition to energy density, the manufacturable cell size is an important factor in characterizing the potential of the technology. In the meantime, the degradation values of planar cells are in the same range as those of the tubular cells produced by Siemens. At the same time, the demonstrated operating times have increased significantly (tubular 40 000 h, planar 26 000 h). Both properties are shown in Table 1.2.

With respect to the development status of system technology and long-term stability, the best results have been achieved with the tubular design by Siemens. However, Siemens has ceased work in this area. The majority of developers see a clear advantage in the cost reduction potential of planar technology. This is due on the one hand to more cost-effective manufacturing technologies and on the

Table 1.2 Results achieved for different SOFC concepts.

Parameter	Anode-supported cells, 750 °C	Electrolyte-supported cells, 800–900 °C	Tubular cells, 900–1000 °C
Power density at 0.7 V ($W\,cm^{-2}$)	0.46–2.0	0.03–0.63	0.11–0.53
Active cell area (cm^2)	20–960	80–840	30–990
Cell degradation rate (% per 1000 h)	1.4—0.2	1.0—0.5	2.0—0.1
Cell operating time (h)	≤26 000	≤10 000	≤4 000
Stack power (kW)	0.1–25	0.4–5.4	–
Ref.	[2, 8, 9, 15, 18–21, 23, 24, 26, 29–33, 36]	[2, 8, 9, 11–14, 20, 22, 25, 27, 34, 35]	[3–10, 16, 17, 20, 28]

other to the higher power density. In this context, there is a clear trend towards an anode-supported design using ferritic chromium steel as an interconnect material. In addition to a higher power density, this concept also allows the operating temperature to be reduced to below 800 °C.

1.3 Representative Research Findings for HT-PEFCs

One of the objectives of high-temperature polymer electrolyte fuel cell (HT-PEFC) development is to increase operating temperatures to between 150 and 180 °C. Higher temperatures make heat removal easier with a smaller cooling surface than in low-temperature polymer electrolyte fuel cells. In addition, the temperature level of the heat removed is higher and can therefore be easily utilized. Due to the higher operating temperature, HT-PEFCs also tolerate a higher proportion of carbon monoxide in the fuel. As a consequence, gas purification is simpler and therefore cheaper. As the membranes do not need to be wetted, costly water management is unnecessary.

A combination of phosphoric acid and polybenzimidazole (PBI) is currently the most interesting material for HT-PEFC membranes. PBI membranes doped with phosphoric acid can be manufactured in a synthesis process using different methods. The basic difference between them lies in whether doping with phosphoric acid is part of polycondensation, that is, whether it takes place *in situ*, or whether doping takes place by soaking the PBI foil in phosphoric acid, or whether it is affected via the gas diffusion layer (GDL) or the catalyst. The polycondensation method was developed and patented by BASF, which is currently the only company manufacturing membranes in this way.

1.3.1 Actors and Major Areas of Development

A number of companies and research institutions are responsible for advances in development. Industry contributions to R&D have been made by BASF, for example, which took over Pemeas in 2006. Pemeas was established by Celanese and a consortium of investors in 2004. Two years earlier, Celanese had begun its launch of a pilot production unit for high-temperature polymer membrane electrode assemblies membrane electrode assembly (MEAs). Another company contributing to R&D is Sartorius, which first became involved in the development of HT-PEFC MEAs and stacks of up to 2 kW in 2001. In 2009, Elcomax took over MEA activities from Sartorius and has marketed MEAs for HT-PEFCs since then. In addition, Fumatech has produced membranes based on AB-PBI (polybenzimidazole) since 2005. Danish Power Systems, a research-based development company which was founded in 1994, is distributing MEAs under the tradename Dapozol [38]. Recently, Advent Technologies started the production of MEAs. The membranes are not based on PBI but are also doped with phosphoric acid. Samsung Advanced Institutes of Technology published data on their own MEAs with excellent performance. An overview of actual HT-PEFC membranes can be found in the literature [39–41] Serenergy, a Danish company, is at present the only supplier of commercial HT-PEFC stacks in the kilowatt range [42]. Plug Power developed HT-PEFC stacks for the use in stationary applications and Volkswagen developed HT-PEFC stacks for automotive application, but stopped these activities recently.

In addition to industrial companies, several research and university institutes are working worldwide in the field of HT-PEFC. Forschungszentrum Jülich is developing HT-PEFC stacks in the power range up to 5 kW for on-board power supply running on diesel and kerosene. In addition, MEAs based on AB-PBI membranes provided by Fumatech are being developed. The Centre for Solar Energy and Hydrogen Research Baden-Württemberg, the Fuel Cell Research Center in Duisburg and the Fraunhofer Institute for Solar Energy Systems (FhG-ISE) also have activities in the field of HT-PEFCs. Their main focus is on stack and system development. Aalborg University in Denmark mainly investigates systems, stacks, and cells [43] whereas the Technical University of Denmark is well known for its research in the field of membranes and MEAs [44]. The key aspect of Spanish groups is on membrane and electrode development (e.g., [45, 46]) and the key aspect of the group at Newcastle University, UK, is on modeling, membrane, and electrode development [47]. Well-known groups performing membrane-related science can be found in the USA at the University of South Carolina and Case Western Reserve University [48, 49]. In recent years, increasing interest in HT-PEFCs can also be observed in China at the Dalian Institute of Chemical Physics [50]. In Korea, the Korea Institute of Science and Technology [51] and the Korean Institute of Energy Research [52] have reported relevant results in this field. Other academic and research institutions, for example, in Russia, are also active in R&D. An overview of the major findings in the HT-PEFC area can be found elsewhere [53–55].

1.3.2 Characteristic Data for Cells and Stacks

The power density of MEAs has reached a high level. BASF and Sartorius have the longest experience with their development, which is reflected in the high power densities and low degradation rates they achieve. MEAs based on the membrane materials produced by Fumatech, which embarked on the technology later, are developing rapidly. Their power densities are now on a par with those of Sartorius MEAs. Figure 1.3 gives an overview of the development of performance data.

The area-specific power densities of HT-PEFC stacks measured at 0.5 V, 160 °C, and with H_2 as a fuel increased from ~180 mW cm^{-2} in 2006 (Sartorius) and 2008 (Forschungszentrum Jülich) to 500 mW cm^{-2} in 2010 (Forschungszentrum Jülich and Fraunhofer ISE) [72–85].

1.4 Representative Research Findings for DMFCs

The development of direct methanol fuel cells (DMFCs) was reactivated all around the world around 1990 thanks to the use of membranes made of sulfonated

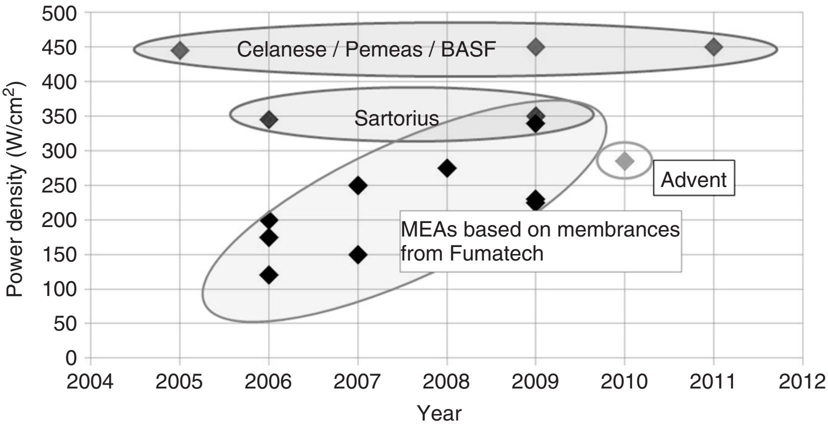

Figure 1.3 Power densities of MEAs manufactured by different developers [56–71] for comparable operating conditions. Temperature, 160 °C; fuel, pure hydrogen, pressure; 1 bar; cell voltage, 0.5 V.

fluoropolymers (Nafion) instead of electrolytes containing sulfuric acid. Development initially focused on mobile applications and then mainly on the area of ''portables'' as a possible replacement for batteries, since the increasing energy demand, in particular for modern cell phones (greater functionality, larger displays, etc.) also increases the discharge rate of batteries.

1.4.1 DMFCs for Portable Applications

More energy can be provided with the available volume and therefore longer lifetimes can be achieved for portable applications <50 W, since methanol or the DMFC system has a higher energy density than Li batteries. A significant share of global research and development work on DMFCs designed for portable applications is being carried out in China, South Korea, Japan, and Taiwan. This is illustrated by the fact that about two-thirds of publications on DMFCs are by Asian research organizations or companies [86]. Therefore, the first products focusing the power supply of small electronically devices (up to 5 W) were also developed in Asia. One example is the commercial available system from Toshiba for the charging of small electronic devices [87].

Nevertheless, the results obtained in Europe and North America are not insignificant. The Federal Ministry of Education and Research [Bundesministerium für Bildung und Forschung (BMBF)] intensively promoted this development in Germany with the ''Micro Fuel Cell'' lead innovation initiative in the last few years. The Fraunhofer Institute for Solar Energy Systems (FhG-ISE) in Freiburg has also been involved in the development of DMFCs since the mid-1990s, but only in the power range <50 W. In addition, it also develops direct ethanol fuel cells (DEFCs)

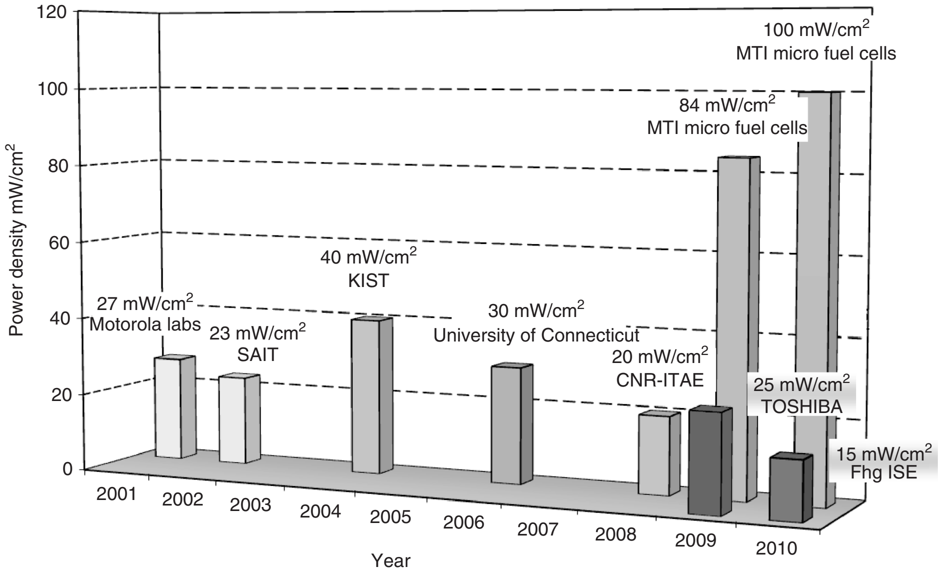

Figure 1.4 Development of the power density of passive DMFC systems at room temperature (20–30 °C).

in the power range from a few milliwatts to several tens of watts. In 2009, a planar micro fuel cell system produced by means of injection molding was developed with a consortium of small- and medium-sized businesses and in close cooperation with FWB Kunststofftechnik. Four injection-molded micro fuel cell modules were integrated into this 2 W DMFC system. The system supplies power for a locating system for container tracking for several weeks. In addition, a passively operated DMFC for an electrocardiograph (ECG) device was also set up [88].

In an international comparison, the US company MTI Micro Fuel Cells developed a passive system (see Figure 1.4) with a power density of 100 mW cm^{-2} [89]. However, no information is available on the catalyst loading of the MEA. Catalyst loadings in the range 6–16 mg cm^{-2} per cell can be found in the literature for passive DMFC MEAs.

1.4.2
DMFCs for Light Traction

Within the framework of a European project from 1996 to 1999, Siemens developed a DMFC stack with a single electrode area of 550 cm^2 together with IRD Fuel Cells (Denmark) and Johnson Matthey (UK) [90]. At a temperature of 110 °C, an oxygen pressure of 3 bar and a cell voltage of 500 mV, a power density of 200 mW cm^{-2} was achieved. Only 50 mW cm^{-2} was achieved at 500 mV in air operation at 80 °C and 1.5 bar. Unsupported Pt and PtRu catalysts with a loading of 1–4 mg cm^{-2} per electrode were used. Over the years, IRD Fuel Cells has improved their systems and they are now providing DMFC systems with 800 and 500 W power output. The current systems are operated with pure methanol or with a methanol–water mixture and with air [91].

In 2000, DaimlerChrysler presented the first go-cart powered by a DMFC system with a net power of 2 kW_{el}. The DMFC system, which was operated with pure oxygen on the cathode side, was developed in cooperation with Ballard Power Systems [92]. The 3 kW stack had an operating temperature of 100 °C.

The Center for Solar Energy and Hydrogen Research [Zentrum für Sonnenenergie- und Wasserstoff-Forschung (ZSW)] in Ulm, Germany, has developed and studied MEAs with nonfluorinated homopolymers together with the Institute of Chemical Process Engineering [Institut für Chemische Verfahrenstechnik (ICVT)] in Stuttgart. With these materials, power densities of up to 240 mW cm^{-2} were achieved in 2002, but with a high catalyst loading of >11 mg cm^{-2} per cell, a cell temperature of 110 °C, an anode pressure of 2.5 bar and a cathode pressure of 4 bar [93]. The first stack consisting of 12 cells was manufactured in 2004 with these MEAs using conventional Nafion 105 membranes. An electrical power of almost 120 W was achieved in air operation at an operating temperature of 70–90 °C, which corresponds to a power density of 55–60 mW cm^{-2} at a single cell voltage of 500 mV [94].

Forschungszentrum Jülich has been intensively involved in the development of DMFCs since 1996. As part of a dissertation on DMFC systems analysis, it was demonstrated that a pressure of more than 3 bar and an air:methanol ratio of 1.75 is required for an operating temperature of 110 °C [95]. Since this ratio is normally insufficient for the stable operation of a DMFC stack, higher air ratios require a higher cathode pressure, which has a negative impact on system efficiency. DMFC systems in the temperature range 80–100 °C are therefore not practical owing to the high losses for cathode air compression and the unfavorable heat management [96]. Even though a specific power density of 100 mW cm^{-2} at 500 mV was achieved at 110 °C after a short development time, further work concentrated on optimizing DMFC stacks under normal pressure. For example, a specific power density of 50 mW cm^{-2} in a compact 500 W DMFC stack under normal pressure at 70 °C was first achieved in 2002 [97]. A first 2 kW demonstration system was set up in 2003. Other systems followed in electric scooters in which the lead acid batteries were replaced with DMFC hybrid systems. In this way, the specific power density was increased to 90 mW cm^{-2} at 80 °C, and the catalyst loading was reduced from 6 to 4 mg cm^{-2} per cell [98].

As in Germany, DMFC development in Korea started in the mid-1990s with the construction of small stacks in the range of several watts. Subsequently the developed DMFC systems became more powerful and more efficient. In 2004, the Korea Institute of Energy Research (KIER) presented a 400 W stack with 42 cells and a catalyst loading of 10 mg cm^{-2}. In tests the stack was operated without additional heating and a power density of ~45 mW cm^{-2} at 500 mV was measured [99]. In 2009, an optimized DMFC system for a scooter consisting of two sub-stacks each with ~700 W peak power was realized. The maximum power density of the MEA in stack tests is above 90 mW cm^{-2} [100].

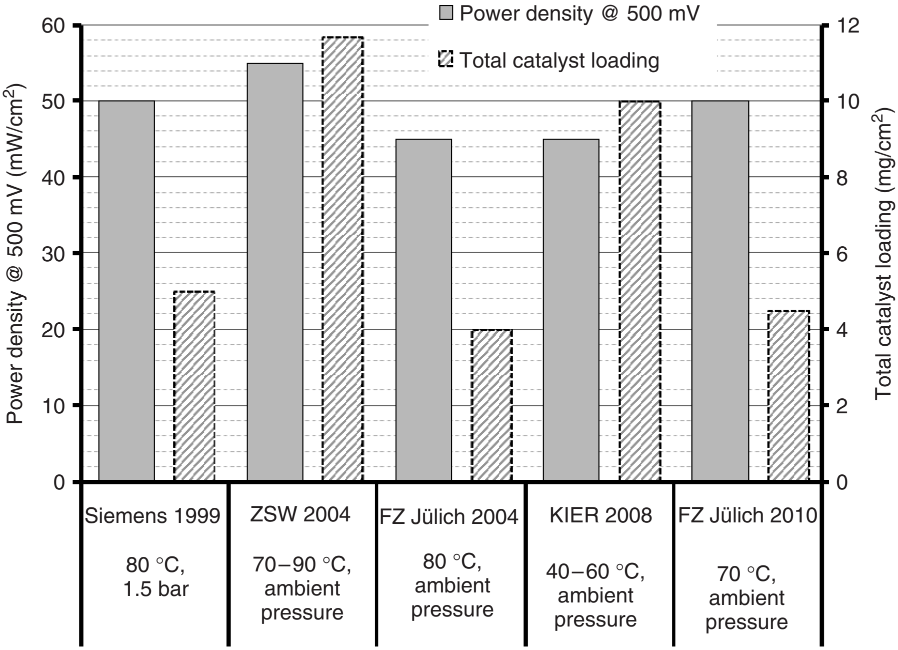

Figure 1.5 Specific power densities of membrane electrode assemblies in DMFC stacks in the power class >100 W.

Also in Japan, companies and research institutions developing DMFC systems. Yamaha Motor, for example, developed several DMFC systems for a two-wheel scooters. In 2003, the first scooter system with an output of 500 W was presented. In recent years, this system was improved and in 2007 a highly efficient 1 kW DMFC system reached a power density of 146 mW cm^{-2}. The fuel cell system achieves 30% system efficiency [100–102]. In the USA, Oorja Protonics has also developed DMFC systems with a power output of ~800 W for small forklifts. However, there is not much information available about the system setup and the system components [103]. Figure 1.5 provides an overview of the power densities of MEAs achieved at an average individual voltage of 500 mV and a temperature range of 70–80 °C.

The best fuel-cell stacks and systems today currently achieve lifetimes of at least 3000 h. SFC Energy, for example, guarantees an operating life of 3000 h within 36 months for its commercial DMFC systems. However, these systems only have a maximum power of 65 W, and the guaranteed 3000 h can also involve replacement of a stack [104]. The Danish company IRD Fuel Cell Technology markets an 800 W DMFC system which also has a lifetime of 3000 h [105].

In terms of reducing the degradation of the electrochemically active DMFC components, Forschungszentrum Jülich was able to increase the long-term stability from less than 50 h to more than 9000 h under real operating conditions from 2005 to 2011 by clarifying degradation mechanisms of MEAs in DMFC systems. For this

Table 1.3 Development of significant membrane modifications.

Approach	Impact	Ref.
Ionically or covalently bonded materials	Stabilized membrane structure	[109]
Sulfonated or nonsulfonated block copolymers	Limited swelling, methanol permeation, and long-term stability High proton conductivity	[110, 111]
Additives in Nafion and sPEEK	Improved conductivity/methanol permeability behavior	[112, 113]
Fully sulfonated polysulfones	Clearly suppressed methanol uptake	[114]
Barrier coatings/layers, for example, via plasma treatment	Reduced methanol permeation Increased membrane resistance	[115, 116]
Polymers with quaternary ammonium groups	Limited long-term stability (>60 °C), conductivity, and availability of ionomer solution	[117–119]

purpose, new corrosion-resistant and carbon-supported PtRu catalysts were used, in particular for the V3.3-2 DMFC system [106].

For both DMFC systems for light traction and for DMFC systems for portable applications, Nafion is still the standard membrane material. A general overview of the polymer electrolyte membrane materials, their modifications, and their function can be found in. [107] and with the focus on the DMFC operation in [108].

In the late 1990s and early 2000s, nonfluorinated homopolymers were studied as promising alternatives. In simplified terms, however, reduced methanol permeation and reduced conductivity are combined in these materials to achieve a DMFC performance comparable to that of Nafion-based MEAs, and the membranes had to be so thin that it was not possible to reduce substantially the absolute value for fuel loss by permeation. Table 1.3 provides an overview of the most significant membrane modifications.

1.5 Application and Demonstration in Transportation

1.5.1 Fuel Cells and Batteries for Propulsion

The major objectives of a reorientation in the energy supply sector, which is generally considered necessary, are the reduction of global and local impacts on the environment, the reduction of dependence on imported energy raw materials, and economic policy-related aspects. The German government has published numerous strategic documents on the role of mobility for reaching these objectives and has established programs for funding the associated energy conversion and storage technologies [120–124]. The technological reorientation within the transport sector focuses on the development of vehicle drives with batteries and fuel cells and also hybrid drives including plug-in hybrid electric vehicles (PHEVs). The goal of the German government calls for 1 million electric vehicles with batteries to be sold by 2020 and 6 million by 2030 [121]. If hydrogen is used in highly efficient fuel cells, renewable power that cannot be utilized due to grid stability can be stored temporarily in the form of hydrogen. Liquid fuels with a high energy density will still be required in the long term, predominantly for heavy trucks, aircraft, and ships. In the future, systems with fuel cells in the power range from ∼5 kW to over 1 MW could be used for the on-board power supply in such vehicles, providing a suitable fuel by reforming the fuel at hand. A renewable basis for liquid fuels is biomass, which can be converted into suitable fuels using biochemical or thermochemical processes. The following section deals with the assessment of the primary energy demand, GHG emissions and the cost of electric vehicle concepts with fuel cells and batteries.

Vehicle concepts whose drive structure and storage dimensions facilitate all-electric drive operation include the following:

- plug-in hybrids with a combustion engine or fuel cell system (PHEVs) with a range of up to 50 km in battery operation
- electric vehicles with fuel cell and battery (fuel-cell hybrid electric vehicles, FCHVs) with a range of over 400 km
- electric vehicles with a battery (battery electric vehicles, BEVs) with a range of up to 200 km.

PHEVs will not be considered in greater depth here, since their low battery capacity requires frequent operation of the combustion engine. The main factors for the comparability of these novel concepts to current vehicles are costs, but also the performance of the drive in terms of top speed and acceleration, and the range that can be achieved between refuelings or battery rechargings. Electric drives are considered to be easy to drive owing to the torque curve of the motor. No gear transmission is required at moderate top speeds, thus streamlining the system.

The fifth to sixth generation of some concept cars is already under development for FCHVs in hydrogen operation and is produced in processes nearing series production, with a total of nearly 800 units built since 1994. Today, only polymer electrolyte fuel cells (PEFCs) are used with operating temperatures between 80 and 95 °C. The preferred storage type is compressed gas storage at 700 bar.

Progress made in the development of fuel cell drives can be shown by referring to performance data for passenger cars (Figure 1.6). The figure shows that in an early phase of development, the stack performances were comparatively low, certainly due to considerably lower power densities or specific performances of fuel-cell systems. This allowed only a limited driving performance. The range was

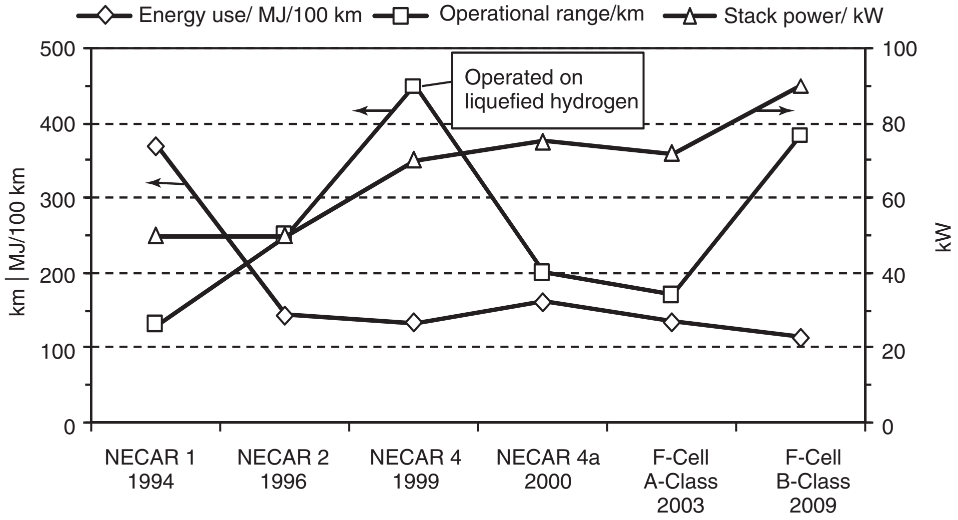

Figure 1.6 Development of range, stack performance, and energy demand of FCHVs.

also initially low when compressed hydrogen (CH2) was used, but was increased by improving the systems and applications of the 700 bar technology to ~400 km and by reducing the energy demand also shown in Figure 1.6, so that there are few restrictions today [125]. NECAR 4 with liquid hydrogen (LH2) already had a range of 450 km back in 1999 – in an unspecified driving profile – and therefore greater than values achieved with the 700 bar technology today. However, the LH2 technology is rarely used in passenger cars today. Other values that document the increase in range are available for the Toyota FCHV-adv, for example, which has a range of up to 830 km [126], whereas its predecessor's range was only 330 km.

Challenges for the further development of fuel-cell vehicles involve increasing power density, specific power and lifetime, improving storage technologies, and achieving competitive costs. Market success will be determined by the availability of the supply infrastructure. Progress in the development of fuel-cell systems can be measured using parameters such as specific power and power density, precious metal requirements and cold start properties. Louie [127] stated that current fuel-cell stacks successfully cold start at $-30\,^{\circ}$C and have a specific power of 1–1.5 $kW_{el}\,kg^{-1}$, corresponding to 2–2.25 $kW_{el}\,l^{-1}$.

Few data are available for the precious metal requirements, which are one of the most important cost drivers in fuel-cell technology. The progress achieved in the reduction of precious metal requirements for PEFCs in automotive applications is shown in Table 1.4.

It is difficult to document the progress made in the development of H_2 storage owing to insufficient data. Broad ranges with large time overlaps are usually stated for the important parameters – specific energy, energy density, and cost. Consistent data for specific lines of developments are not available. For Figure 1.7, the storage densities and specific energies according to the US Department of

Table 1.4 Precious metal requirements for PEFCs in automobile applications.

Company	Precious metal requirement per FCHV (g)	Year	Ref.
Daimler	30	2009	[128]
General Motors	112	2000	[129]
	80	2007	[130]
	30	2013	[130]

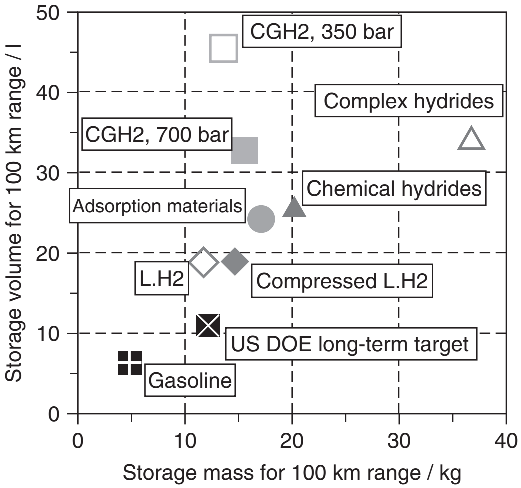

Figure 1.7 Mass and volume of different storage systems relative to a range of 100 km.

Energy (DOE) [131] for 2009 were therefore assessed using the specific energy requirements of FCHVs per 100 km in order to take into account the much more efficient utilization of energy in FCHVs in comparison with gasoline-powered passenger cars used as a reference here. The values in the figure represent the mass or volume of storage systems required for a range of 100 km, assuming that the FCHVs are designed for a range of 400 km. The data are taken from simulations based on the New European Driving Cycle [or Motor Vehicle Emissions Group (MVEG)] for determining the mass-dependent mechanical energy requirement at the wheels of small to mid-sized passenger cars [132, 133].

Some BEVs are produced in series today [134]. Lithium ion technology is used for this purpose, because it is best suited to requirements such as high specific energy and energy density, but also provides long lifetime and low self-discharge rates. Table 1.5 contains information on these batteries in comparison with lead

Table 1.5 Performance data for batteries.

Parameter	Lead acid	Ni–metal hydride	Li ions
Theoretic specific energy (Wh kg^{-1})	167	214	420
Effective specific energy (Wh kg^{-1})	35–49	45–75	65–150
Specific power (W kg^{-1})	227–310	250–1000	600–1500
Energy density (Wh l^{-1})	70–96	125–182	130–300
Power density (W l^{-1})	445–620	600–2800	1200–3000
Lifetime (years)	2–6	12	7–10
Lifetime (cycles × 1000)	0.1–0.3	2.5–300	2–300
Self-discharge (% per month)	2–3	20–30	2–10
Temperature range (°C)	−30 to 70	−10 to 60	−25 to 50

acid and nickel metal hydride batteries. On the cell level, a specific energy of up to 185 Wh_{el} kg^{-1} is achieved today [135]. In automotive applications, allowances must be made in this respect for spare capacity and system integration into an overall module including cooling, control electronics, and housing. For example, the figure for the Opel Ampera based on the data sheet in [136] is ~90 Wh_{el} kg^{-1}.

Grube and Stolten [132] reported detailed simulations based on the MVEG that were carried out for a comparison of BEVs with a range of 200 km and FCHVs with a range of 400 km. Table 1.6 gives an overview of primary energy demand, GHG emissions, and system costs, comparing vehicles with fuel cells and vehicles with batteries. It shows that, if the relevant cost targets can be met, the costs of the power supply system with fuel cells may be considerably lower at a range that is twice as high. With respect to the well-to-wheel (WtW) balance of primary energy input and GHG emissions, FCHVs are at a slight disadvantage if natural gas is used to produce hydrogen. These values can be further reduced by switching to primary energies for generating power and hydrogen with lower GHG emissions. The recharging of BEVs and refueling of FCHVs pose special challenges. In principle,

Table 1.6 Comparison of electric cars with batteries and with fuel cells [132].

Parameter	Electric car	
	BEV 200	FCHV 400
Cost of power supply system (€)	9733	7401
Stored energy (MJ)	102	396
Vehicle mass (kg)	1325	1313
Specific primary energy, well-to-wheel (MJ km^{-1})	1.61	1.65
Specific GHG emissions (g km^{-1})	88	97

refueling times, fueling procedures, and refueling intervals for fuel-cell vehicles are comparable today.

From the present point of view, grid services provided by BEVs are possible in principle; however, the influence of the depth of discharge and also charge and discharge capacities on battery lifetime will have to be considered. Long battery lifetimes can be achieved if the depth of discharge is in the range of a small percentage and if temperatures are considerably lower than 60 °C [137]. It is important for BEV balances to take into account the time of day at which batteries are typically recharged, since the energy must be fed in at the same time. In the short and medium term, hydrogen provision may benefit from the integration of residual hydrogen from industry, until new production capacities can be built in the long term, preferably on the basis of renewable energies and with the option of energy storage.

1.5.2 On-Board Power Supply with Fuel Cells

On-board power supply is required by almost all mobile applications. In the past few years, studies were conducted on on-board power supplied by auxiliary power units (APUs) for the transport of goods and passengers by sea and air. This includes aircraft, ships, passenger cars and, above all, trucks. Numerous US studies have considered the use of fuel cells in "line haul sleeper trucks" [138–141]. Targets for different APU applications are compiled in Table 1.7. The targets for applications in aircraft and ships are defined by considerably fewer values than for combined heat and power generation. The cost targets are most ambitious for passenger car applications. The power density target for aircraft applications is roughly the

Table 1.7 Targets for different APU applications and for stationary systems based on natural gas.

	Aircraft	Passenger car	Truck	CHP
Ref.	[142–144]	[145]	DOE targets 2015/2020 [146, 147]	
Power range (kW)	100–400	10	1–10	1–10
Efficiency (%)	40	<35	35/40	42.5/45
Specific cost	€1500 kW^{-1}	€40 kW^{-1}	\$600 kW^{-1}	\$450 kW^{-1}
Durability (h)	20 000/40 000	5000	15 000/20 000	40 000/60 000
Dynamic aging (% per 1000 h)	–	–	1.3/1	0.5/0.3
Power density ($W l^{-1}$)	750	333	35–40	–
Mass-specific power ($W kg^{-1}$)	0.5–1	250	40–45	–
System availability (%)	–	–	98/99	98/99
Cold starting	–	<1 s	10/5 min	30/20 min
Load cycle 10–90%	–	<1 s	3/2 min	3/2 min
Partial load	–	1:50	–	–

Table 1.8 Power densities achieved and targeted for APU systems.

Power density ($W_{el}\ l^{-1}$)	2000	2003	2005	2006	2008	2010	2011	2012	2015	2020
DOE 2006 targets	–	–	25	70	–	100	–	–	100	–
DOE target review	–	–	–	–	–	–	–	30	35	40
Delphi 3.5 kW_{el}	10	20	25	–	17	–	–	–	–	–
Cummins 1.5 kW_{el}	–	–	–	–	–	5	–	–	–	–
Webasto 1 kW_{el}	–	–	–	8	–	–	–	–	–	–
Truma 0.25 kW_{el}	–	–	–	–	–	–	2.7	–	–	–

same as for passenger cars, whereas the high long-term stability requirement corresponds more closely to the targets for stationary applications.

Table 1.8 shows the power densities achieved for different fuel-cell systems for on-board power supply in the power range from 250 W_{el} to 3.5 kW_{el}. The US company Delphi obviously made great progress in making systems more compact from 2000 to 2005. Based on these status data, the DOE established targets for the period up to 2020, first in 2006 and again in 2009. However, it became clear in 2008 that the power density decreased to 17 $W_{el}\ l^{-1}$ with increasing system autonomy and technical maturity. The power density to be achieved by 2020 is now 40 $W_{el}\ l^{-1}$. Many of the targets set by the DOE are tailored specifically to SOFCs for application in trucks. Other system providers have been and still are the companies Webasto (Germany) and Cummins (USA). A 250 W_{el} device for on-board power supply in campers manufactured by Truma (Germany) is also shown. The device works with liquefied petroleum gas (LPG) as an energy carrier; a micro steam reformer from IMM (Germany) produces a hydrogen-rich gas.

How can the power density of a system for on-board power supply be significantly increased? Figure 1.8 shows the power density of catalysts for partial oxidation, autothermal reforming, and steam reforming. Depending on the process and the fuel used, the power density is between 10 and 50 $kW_{el}\ l^{-1}$. Mixing zones and heat exchangers are part of the design, resulting in power densities of 1–5 $kW_{el}\ l^{-1}$ for reformers as a core component for fuel production. PEFC stacks for the automobile sector achieve up to 1.5 $kW_{el}\ l^{-1}$. SOFC stacks manufactured by Delphi (USA) have a residual value of at least 720 $W_{el}\ l^{-1}$. Less compact stacks have a power density in the same range as HT-PEFC stacks of between 50 and 200 $W_{el}\ l^{-1}$ today. If the different components for fuel production are interconnected as a package, 120 $W_{el}\ l^{-1}$ can be achieved with current designs. For example, if the best SOFC stack is combined with a compact partial oxidation reactor (3.3 $kW_{el}\ l^{-1}$), an HPD of 600 $W_{el}\ l^{-1}$ could be achieved at best. However, only 17 $W_{el}\ l^{-1}$ can be obtained today since further system components such as heat exchangers, pumps, blowers, and compressors and also suitable electronics are required. Making these systems more compact will require intensive efforts regarding component development and innovative system design.

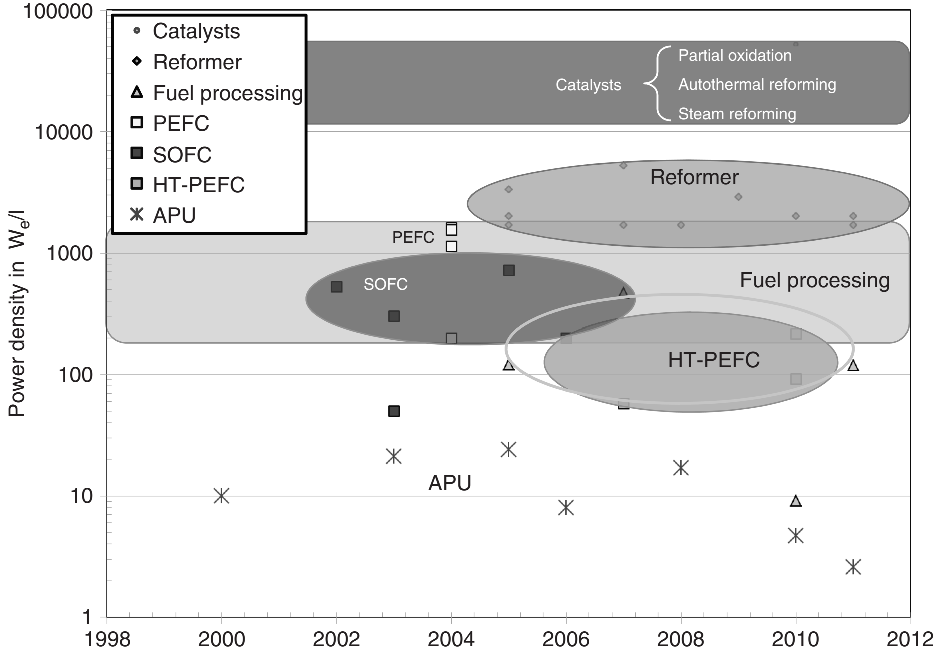

Figure 1.8 Power densities achieved for catalysts, reformers, fuel processing systems, fuel-cell stacks, and APUs.

1.6 Fuel Cells for Stationary Applications

1.6.1 Stationary Applications in Building Technology

In 1996, Vaillant started its 13 year development work on fuel-cell heaters based on natural gas as an energy carrier. It took 6.5 years to develop a prototype. Another 6.5 years were planned until the product was launched on the market. Fifteen second-generation demonstration systems were installed in 2002. The reliability of the systems tripled. From 2004 to 2006, the focus was on field testing. Fifty-six third-generation demonstration systems were tested in Germany, The Netherlands, Spain, and Portugal. The volume of the natural gas reformer was cut by half, the electrical efficiency was increased from 18 to 30%, and the stacks achieved lifetimes of up to 6000 h. It was found that for the third generation, the minimum power could be reduced from 2.5 to 1 kW. A lifetime of up to 12 000 h seemed possible. There were, however, problems with the reliability of the electronics, with controls, and with conventional system components. Work on conventional PEFCs ceased at the end of 2005. In the end, the cost objectives could not be achieved with Nafion membranes and the relatively complex process of fuel production. Work on HT-PEFCs followed, and also a cooperation with Staxera and Fraunhofer IKTS for the development of an SOFC building energy system which started in 2006. Field tests were planned starting in 2011 [148, 149].

In 1997, the first devices for building energy supply based on a PEFC prototype manufactured by Dais Analytic Corporation (DAC) in Boston were installed at HGC Hamburg Gas Consult. Nine ''alpha units'' were installed and tested until 1999. European Fuel Cell GmbH (EFC), a 1999 spin-off, started to build an improved ''beta unit'' in 2001. This company was then taken over in 2002 by the Baxi Group (the third-largest heating technology group in Europe with Brötje as one of its well-known brands). A preproduction series was planned starting in 2009. When the field tests with a ''gamma'' version are successfully completed, 40 000 devices of the marketable aggregate are to be produced per year in the period from 2010 to 2013 [150–152].

The company inhouse engineering GmbH was founded in 2005 as a spin-off from Schalt- und Regeltechnik GmbH. The company is developing 5 kW fuel-cell systems based on PEFCs and natural gas reforming. Field tests were planned to start in 2009. HT-PEFCs are being considered in a development project. The development partners are TU Bergakademie Freiberg (responsible for the reformer), DBI Freiberg (burners, cooling device), and the University of Magdeburg (control system). The prototype with four cells had an electric power of 150 W in 2009. A 30-cell system with 1.5 kW was subsequently to be developed for operation with a reformate. When the project ended in August 2009, the subsystem for fuel production in particular had been successfully developed. No information was made available on the overall system [153].

All the R&D work presented so far uses natural gas as an energy carrier. In the MÖWE collaborative project, the partners, that is, S&R Schalt- und Regeltechnik GmbH, Öl-Wärme-Institut GmbH (OWI), Behr GmbH, and Umicore AG, studied the development of a steam reformer based on light heating oil for decentralized hydrogen production and system testing using a stationary PEFC system. Recent research findings with diesel steam reformers show that degradation already becomes apparent after a few operating hours [154]. Regenerating the catalyst by oxidation of the carbonaceous deposits is possible [155]. However, there is still a considerable need for development.

Since 2006, a relatively large number of companies have constructed complete SOFC systems. Most of these systems are initial laboratory test systems. However, they still highlight the impressive progress made during the last few years. Hexis has the most experience with small building energy systems and has made great progress in terms of reliability and long-term stability with Galileo, a system that it has developed further [12]. Ceramic Fuel Cell Limited (CFCL) has already demonstrated a net electrical efficiency of 60% with a 1.5 kW system [156]. SOFCs received an extra boost by the work of Bloom Energy, which operated behind closed doors for several years and has been selling 100 kW systems since 2009.

Current work on stationary applications of fuel cells for building technology are being carried out within the German Callux demonstration project by the heater manufacturers Baxi Innotech, Hexis, inhouse engineering GmbH, and Vaillant, together with the utility companies E.ON Ruhrgas, MVV, EnBW, EWW, and VNG, and with the research institution ZSW. Up to 800 field test systems are to be installed by 2012, preferably in private households; the 100th device was built and installed in November 2010. The systems are based on SOFCs and PEFCs, can be fueled with natural gas or bio natural gas, and have an electrical power of 1 kW_{el} and a thermal power of 2 kW_{th} in the combined heat and power system.

1.6.2 Stationary Industrial Applications

Based on Ballard's development in the 1990s of PEFC stacks for mobile applications, stationary applications were also developed. The first 210 kW combined heat and power system was tested by Ballard in Burnaby (Canada). The system was developed together with Alstom. A field test program with improved 250 kW systems (BEWAG, EBM-Schweiz, etc.) was launched in 2000 [157]. In the test period from 2001 to 2004, seven 250 kW systems were delivered worldwide. Component reliability was one of the main problems. In addition, the low electrical efficiency of 35% was a great disadvantage in competition with conventional technology. The longest operating time was 6500 h.

Development work at MTU resulted in a 250 kW system with molten carbonate fuel cells (MCFCs) operated at Ruhrgas in Dorsten in 1997. The system had a short lifetime, but provided valuable information for improvements. In total, 35 HotModul systems had been delivered (together with FCE) by the end of 2006. Several systems had a lifetime of >30 000 h until the stack had to be replaced. The overall electrical efficiency is between 45 and 46% at 50–100% of the rated power [158].

Based on the good results achieved with eight SOFC systems of the 25 kW class (cell tubes with a length of 50 cm) up to 1995 (accumulated operating time 35 000 h; more than 100 thermo cycles; degradation ~0.2% per 1000 h), Siemens started to develop a system in the 100 kW class with cell tubes 1.5 m long. The first 100 kW SOFC system was put into operation in Arnhem (The Netherlands) in 1997. Extensive maintenance was required after 3500 operating hours, since several tubes were damaged. Subsequently, the system was operated for more than 20 000 h with negligible degradation and an overall electrical efficiency of 46% [159]. In 2000, a hybrid system operated under pressure (coupled with a gas turbine) was put into operation in the USA. Despite numerous breakdowns, this system achieved ~2000 operating hours and an overall electrical efficiency of 52% [160].

1.7 Special Markets for Fuel Cells

In the past few years, a number of applications for fuel cells have emerged that may enter the market early [161, 162]. This is not a matter of niche applications, but as these markets are less price sensitive they are well suited to act as door openers for the broad application of fuel cells. These include:

- on-board power supply for campers and sailing yachts
- electric vehicles such as e-bikes, scooters, and vehicles for tourists
- battery replacement for emission-free drives in electric warehouse trucks
- uninterruptable power supply (UPS) and emergency power supply, for example, for telecommunications, hospitals, control centers, and IT centers
- micro applications (4C applications).

The power required for these applications ranges from 100 mW to ~50 kW. Many applications use methanol in DMFCs, methanol or LPG in connection with a reformer in PEFCs, or hydrogen in PEFCs, as energy carriers. Table 1.9 shows the most important developments for this segment in the last 20 years with a special focus on the German situation.

1.8 Marketable Development Results

1.8.1 Submarine

Siemens tested on-board power supply for submarines with a 100 kW alkaline fuel cell on the U1 submarine of the German Navy in 1988. The 6 month test runs were very successful and constituted the basis of the German Navy's decision to equip the new 212 generation of submarines with fuel cells. However, the more recent PEFC development was used. The fuel cell developed by Siemens is based on a Nafion membrane with a high platinum loading and a metallic bipolar plate with an elastomer seal, all developed specifically for the utilization of H_2/O_2.

Nine modules of the 34 kW type were connected in a 300 kW system and used in the German Navy submarines. The further development led to 120 kW modules that are currently being used in the class 214 submarines intended for export [178]. Initial tests were carried out in submarines in 2003 [179]. These fuel-cell systems have been available as a commercial product for this particular application for a few years.

1.8.2 DMFC Battery Chargers

Toshiba launched its DMFC-driven Dynario battery charger system in 2009. Owing to the use of concentrated methanol solution, the system has a size of only $150 \times 21 \times 74.5\ mm^2$ and a low weight of 280 g. The system has a hybrid structure, which uses a lithium ion battery charged by the DMFC to store electricity. The Dynario system and its fuel cartridge fully comply with the International Electrotechnical Commission's safety standards; it was sold in Japan, together with a special fuel cartridge for simple and fast refueling, in a limited edition of 3000 units [87]. With a DMFC device that is called Mobion, the US company MTI has developed a product for a similar market to Toshiba's Dynario systems. The strategy

Table 1.9 Development status of fuel-cell systems for special markets.

Market	Developer	Country	Fuel cell	Fuel	Max. continuous system power	Ref.
On-board power supply	SFC Energy	Germany	DMFC	CH_3OH	<250 W_{el}	[163]
	Truma	Germany	HT-PEFC	LPG	210–250 W_{el}	–
	EnyMotion	Germany	PEFC	LPG	250 W_{el}	[163]
Electric vehicles	Clean Mobile/SFC Energy	Germany	DMFC	CH_3OH	250–500 W_{el}	[163]
	Masterflex	Germany	PEFC	H_2	200 W_{el}	[163]
Material handling	Siemens	Germany	PEFC	H_2	10 kW_{el}	[164]
	Still/Linde/Proton Motor	Germany	PEFC	H_2	18 kW_{el}	
	Still/Hydrogenics	Germany/ Canada	PEFC	H_2	10 kW_{el}	[165]
	Still/Hoppecke/ Linde/Nuvera	Germany/ Italy	PEFC	H_2	5 kW_{el}	–
	Still/Hoppecke/ Linde/Hydrogenics	Canada/ Germany	PEFC	H_2	12 kW_{el}	[165]
	Gruma/Linde/ Hydrogenics	Germany/ Canada	PEFC	H_2	–	[166]
	Forschungszentrum Jülich	Germany	DMFC	CH_3OH	1.3 kW_{el}	[167]
	Nuvera	Italy	PEFC	H_2	10 kW_{el}	[168–170]
	Proton Motor	Germany	PEFC	H_2	2, 4, or 8 kW_{el}	[171]
	Plug Power/Ballard	USA/Canada	PEFC	H_2	1.8 or 10 kW_{el}	[172–174]
	Oorja Protonics	USA	DMFC	CH_3OH	0.8 kW_{el}	[103]
UPS	Future E/Ballard Power Systems	Germany/ Canada	PEFC	H_2	0.5, 1, or 2 kW_{el}	[163]
	IdaTech	USA	PEFC	CH_3OH	2.5 or 5 kW_{el}	[175]
	IdaTech	USA	PEFC	H_2	2.5 or 5 kW_{el}	[176]
	Dantherm/Ballard	Denmark/ Canada	PEFC	H_2	1.6 or 5 kW_{el}	[177]
4C applications	FhG-ISE	Germany	DMFC	CH_3OH	–	–
	Friwo/Solvicor/ FhG-ISE	Germany	DMFC	CH_3OH	~100 mW_{el}	[163]

of the Mobion DMFC is the use of highly concentrated methanol in a simple system where the water management is done within the MEA by back-diffusion of water from the cathode to the anode [180]. At various exhibitions, Samsung SDI has shown their DMFC systems that can be used for charging mobile phone batteries or for powering laptops. In 2009 they presented a DMFC system with a power output of 25 W that was developed for military application [181, 182].

Commercial DMFC systems are currently being sold by SFC Energy in Brunnthal-Nord (Germany). It has sold more than 20 000 DMFC systems in the power range up to 250 W from when it was founded in 2000 until 2011 [183]. These fuel cells were developed as battery chargers for a voltage range from 12 to 24 V specifically for the reliable provision of power for mobile and portable applications in the power range up to 250 W. They are mainly used in the recreational sector, that is, for supplying power to electricity consumers in mobile homes, caravans, and sailboats. A lead acid battery is usually charged by a DMFC with a continuous output of 25–90 W, which means that the systems have a charging capacity of 600–2200 Wh per day. Other areas of application are transportation and safety engineering and also environmental sensor technology.

A frequently cited disadvantage of methanol is its toxicity. What is often forgotten, however, is that methanol is much less toxic than the conventional energy carrier gasoline, which owing to its benzene content is, moreover, also carcinogenic and teratogenic. Methanol, in contrast, although acutely toxic, is neither teratogenic nor carcinogenic and is much more readily biodegradable than gasoline or diesel. It is therefore not harmful to handle methanol if hermetically sealed containers are used that permit safe handling, as in the case of SFC's fuel cartridges. These cartridges, for example, have the seal of approval from the German Technical Control Board (TÜV-GS Siegel) and are licensed for transportation purposes on-board land vehicles, ships, and aircraft.

1.8.3 Uninterruptable Power Supply/Backup Power

In addition to the portable battery charger systems, the uninterruptable power supply of critical systems for telecommunication infrastructure and hospitals is an interesting market for fuel-cell systems [184]. The US company Hydrogenics supplies the HyPM XR system that is available with a power output of 4, 8, and 12 kW. These systems can be scaled up to a power output of 100 kW and they are easy to integrate into an electrical cabinet [185]. The companies Dantherm and Ballard have a cooperation where Ballard is the stack supplier and Dantherm the system integrator. The Dantherm power systems have a power output of 1.6 or 5 kW [177, 186–188]. The US company IdaTech offers systems that are constructed for outdoor use. These ElectraGen systems are available for operation with hydrogen or for use with a methanol–water mixture in combination with a reformer. The reformer can provide gaseous hydrogen from the supplied liquid mixture of methanol and water; this makes storing hydrogen cylinders on-site unnecessary and reduces the overall costs of the fuel supply. The systems are available with a power output of 2.5 or 5 kW [175, 176].

German system integration companies that already have products on the market are Rittal, P21, b + w Electronics and FutureE Fuel Cell Solutions. They all use PEFC systems, with either pure hydrogen or methanol as an energy carrier. The methanol-based systems are fuel-cell backup power systems from IdaTech. All other system providers use fuel-cell systems based on pure hydrogen. DMFC systems manufactured by SFC Energy (see Section 1.8.2) are also used for emergency power supply in the low-power range.

Among the system integration providers mentioned above, only P21 produces its own fuel cell stacks. Rittal uses stacks from Ballard for high-power UPS systems (RiCell Flex fuel-cell system up to 20 kW) and stacks from Schunk for less powerful systems (RiCell 300, 600, 900 W). A strategic partnership established between Rittal and IdaTech in 2003 for the integration of fuel-cell systems for emergency power systems has since been terminated. Rittal now purchases stacks from Ballard via FutureE Fuel Cell Solutions and from Schunk, who developed their own stacks. FutureE Fuel Cell Solutions is a start-up company founded in 2006 which sold its first fuel-cell emergency power systems of the Jupiter family in 2008. These are modular hydrogen systems with a maximum power of 40 kW.

1.8.4 Light Traction

Owing to financial support from the government, fuel-cell users in the USA are entitled to a federal tax credit up to \$3000 kW^{-1}, so the market for light traction applications especially in the USA is booming [189]. Different companies have developed fuel-cell systems for material handling applications and numerous such systems are now running in different distribution centers around the USA. The companies Plug Power, Nuvera, Hydrogenics, and Oorja Protonics are the main actors in the market. In the year 2011 alone for Plug Power it was possible to double the number of systems sold annually to 1456 modules up to October [190].

Whereas Plug Power, Nuvera, and Hydrogenics using pressurized hydrogen as fuel (350 bar) the Oorja Protonic systems use methanol. The Oorja DMFC system has a lower power output than the hydrogen systems and it does not replace the complete battery, but in combination with a conventional battery it is used as a range extender.

In the light traction sector, the only marketable products in Germany are also manufactured by SFC Energy. They are based on the DMFC EFOY Pro Series. These devices are referred to as ‘‘range extenders,’’ internal chargers for the batteries of electric scooters or small electric cars with a maximum electric power of 90 W, which make the range of these vehicles greater.

1.9 Conclusion

Intensive global research and development efforts together with groundbreaking demonstration activities have conclusively demonstrated that in selected fields of application, fuel cells have a high potential for effectively addressing the challenge of reducing CO_2 emissions and conserving resources by replacing conventional energy technology in the future. Combined with conventional energy carriers, fuel cells will then contribute substantially to sustainability in the energy supply sector.

Laboratory and field tests have demonstrated the suitability of SOFCs, particularly with anode-supported cells, for applications in the field of decentralized combined heat and power generation and large on-board power supply systems. These cells have a power density of up to $2\,W\,cm^{-2}$ at 0.7 V, a size of up to $960\,cm^2$, a stack power of up to 25 kW, and technically relevant aging rates of 0.2% per 1000 h. However, coupling SOFCs with a combined cycle process for highly efficient power generation with systems efficiencies of more than 80% will require considerable efforts to improve SOFC reliability and complex innovations in terms of systems and plant technology. HT-PEFCs with PBI membranes doped with phosphoric acid have been developed for about 15 years and are therefore still relatively new. Nevertheless, there is conclusive evidence that they are suitable for electrochemically converting fuels containing CO and that fuels available on the market can therefore be used, for example, diesel and kerosene. Reducing the cell overpotential on the cathode side is considered to be a means for greatly enhancing the electrochemical performance of the cells. Today, power densities of $0.5\,W\,cm^{-2}$ can be achieved in HT-PEFC stacks with an operating temperature of 160 °C and a cell voltage of 0.5 V. DMFCs are of great interest for all those applications that are powered by batteries today and struggle with limitations in terms of functionality and operating life. In spite of high catalyst loadings of $\sim$4 $mg\,cm^{-2}$ per cell, the power densities of DMFCs are hardly above $50\,mW\,cm^{-2}$, depending on the pressure (<4 bar), the operating temperature (70–100 °C) and the air:methanol ratio. This is essentially a consequence of methanol permeation to the cathode, which promising R&D approaches such as the production of barrier layers by means of plasma treatment intend to address.

The transportation sector aims to use fuel cells to increase the efficiency of passenger vehicle and bus drivetrains and also for on-board power supply used in heavy goods vehicles. Hybridization with a suitable battery dimensioned for the intended purpose is crucial for the performance and efficiency of the overall system. Mid-sized cars field tested today have a specific energy requirement of slightly over 100 MJ per 100 km. With an electric stack power of 90 kW and a tank system for gaseous hydrogen at 700 bar, vehicles have a range of almost 400 km. Cold-start ability at up to −30 °C, a noble metal loading of 30 g per vehicle, and a stack power density of $\sim 2\,kW_{el}\,l^{-1}$ – values achieved during development – show that current passenger cars with a fuel-cell drive are now technically mature enough to be launched on the market as soon as ongoing field tests are completed. On-board power supply (APUs) for aircraft, ships, railroads, and trucks is targeted at compact, efficient, and low-emission energy systems that use the fuel available on-board, for example, kerosene, diesel, or LPG. For power generation by means of fuel cells, this requires an adapted fuel production system and also further system components. Current APUs with an average power density of $\sim 17\,W_{el}\,l^{-1}$ are 2–44 times below the estimated target values.

The strategy for stationary applications of fuel cells for decentralized energy supply in the power range from 1 to 5 kW_{el} for private households and from

100 to X000 kW_{el} for industrial and local facilities has changed numerous times. Development is currently focused on small- and medium-sized SOFC systems, which are undergoing intensive field testing all over the world. The benchmark for the electrical efficiency of a 1.5 kW_{el} system produced by CFCL is 60%.

Fuel-cell applications in the special markets segment, for example, on-board power supply, lightweight electric vehicles, emergency power supply, and very small electronic systems, are also undergoing in-depth field testing. The fuel-cell types used are DMFCs, HT-PEFCs, and PEFCs with upstream LPG reforming and PEFCs with hydrogen. The electrical system powers are between 100 mW_{el} for 4C applications and 18 kW_{el} for forklifts. Special fuel-cell systems for the power supply of submarines with PEFCs, for small consumers in the recreational sector with DMFCs, uninterruptible power supply with PEFCs, and DMFCs, and also for driving microcars with DMFCs, have achieved a position in the market.

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