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Fuel cell electric vehicles and hydrogen infrastructure: status 2012

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Abstract: Automotive fuel cell systems have made huge progress over the last few years; recent developments concerning hydrogen vehicle technology and infrastructure are presented.

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OPINION

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1 Introduction

Today, on a global scale, approximately one billion vehicles are on the roads. There are recent forecasts that this number could rise by the year 2020 to about 1.5 billion vehicles. That significant growth is mainly caused by the ongoing impressive economic expansion and industrial development of China and India. Although in these two countries the car ownership ratio is still quite low, both markets became recently very important for the global automotive industry. For many car manufacturers, China is already today their single biggest market.

More than 95% of the fuel used for propulsion purposes is produced from fossil sources of energy. Even the implementation of the most advanced conventional powertrain options hence cannot prevent that this will inevitably lead to a higher total crude oil demand by the transportation sector and consequently to higher global CO_2 emissions. Due to the fact that such a 50% increase in demand for oil and CO_2 production would be completely unacceptable for financial, ecological and energy security reasons, every automotive technology strategy needs to include a replacement of fossil fuels as energy carriers.[1,2]

At many car manufacturers, the development of battery electric and fuel cell electric vehicles, respectively, powertrain concepts has a very long tradition. The modern history dates back to the 1960s and 1970s,[3,4] *e.g.*, on the hydrogen side, the world's first fuel cell electric vehicle, the GM Electrovan of 1966, was developed by General Motors based on alkaline fuel cells and fuel storage in the form of cryogenic hydrogen and oxygen.[1,4a,5] But since these technology paths were not mature enough for a commercial application at that time, they fell into oblivion.

During the 1990s, both the battery electric and the fuel cell electric vehicle programs were revived and led among other things to the development of the GM EV1,[3] the world's first modern-era mass-produced EV, and to Daimler's NECAR1 fuel cell vehicle. Today, many automotive companies, such as GM, Honda, Hyundai and Toyota, pursue large-scale hydrogen programs. Over the last decade, BMW, Nissan, Volkswagen as well as several small and medium-sized companies also presented hydrogen concept cars to the public.

A brief well-to-wheels comparison of different powertrain options is given in Fig. 1a. Electrification pathways provide the possibility to drive the greenhouse gas (GHG) emissions of the automotive passenger transportation sector toward zero. The reductions in GHG emissions shown here for the pure battery-electric vehicle (BEV) may be exaggerated since its limited driving range prevents it from being used for all trips. Trips longer than the EV driving range (Fig. 1a assumes 100 miles or 160 km) would have to be taken with another car or even transport mode, and would likely increase the overall GHG footprint of the journey. The range-extender electric vehicle does not have this limitation. Here a variant with a 40 mile or 64 km EV range is shown (see ref. 2 or 3 for an E-REV definition). The same lack of range restriction is valid for the fuel cell electric vehicle technology (FCEV). Of course it has to be stated that this only holds true as long as hydrogen refueling is widely available. Hydrogen infrastructure aspects will be discussed in detail in Section 3 of this publication. The symbols in Fig. 1a also explain the respective energy mix for the European Union (EU), the United States of America (US) and China. For the assessment, the EU numbers were chosen as standard values. With the gasoline and diesel options, transportation is tied exclusively to oil and subject to oil price volatility. With the addition of compressed natural gas (CNG) or biofuel vehicles, transportation can diversify from its dependence on fossil oil. The various electrification options, E-REV, BEV, and FCEV, provide an opportunity for an even further increased diversification from oil.

This paper will focus merely on fuel cell electric vehicles and the required technologies as well as infrastructure efforts. Ref. 2 is thus recommended to those readers who are interested in a brief summary on sustainable transportation and extended-range electric vehicles. Details of the extended-range EV powertrain and strategic aspects of vehicle electrification in general are explained in ref. 3 and 4. In particular, ref. 4*c* provides a comprehensive summary and comparison of studies relating to the various types of alternative propulsion. Ref. 4*d* describes an interesting automotive application of electrochemistry in the field of catalytic converters and emissions control where energy storage or propulsion is not directly concerned.

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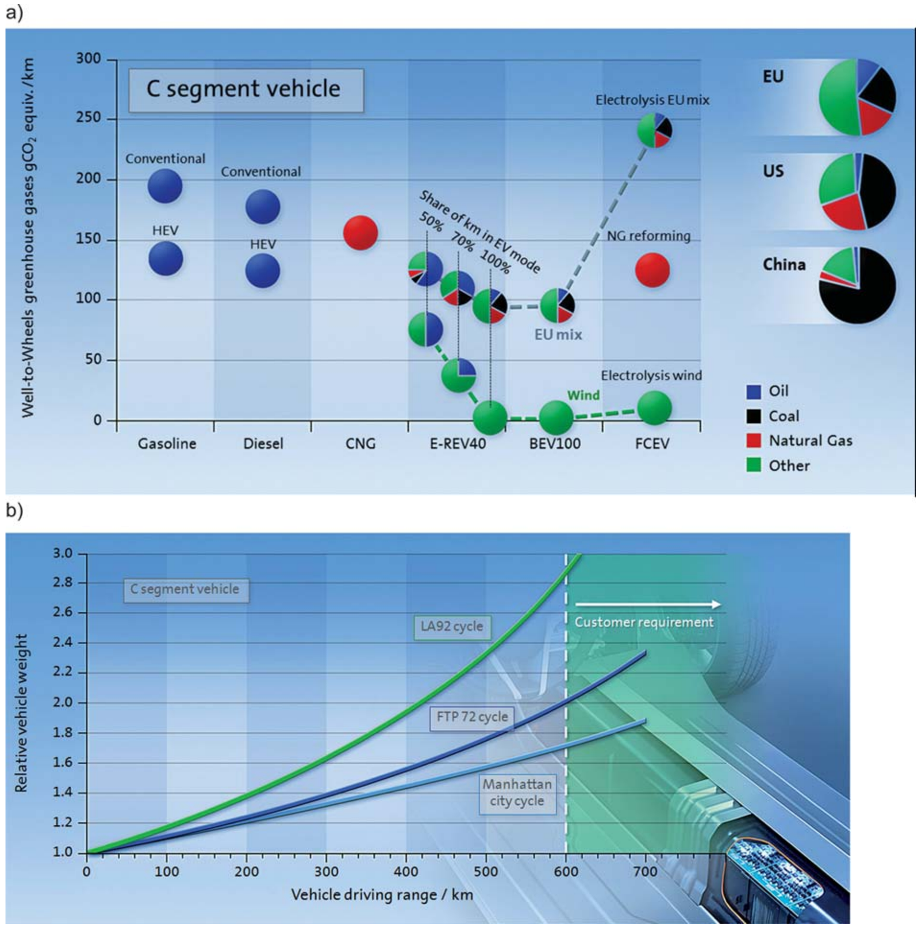

Fig. 1 (a) Well-to-wheels greenhouse gas emissions for various propulsion types and fuel sources; sources: well-to-tank path – JRC/EUCAR/CONCAWE study, tank-to-wheels path based on GM analyses for a compact car (C segment vehicle), U.S. on-road[4a,b] and (b) vehicle curb weight sensitivity to the battery-electric vehicle driving range depending on various driving cycles, considering a compact car (C segment vehicle).[4a]

2 Fuel cell electric vehicles and hydrogen technologies

2.1 Hydrogen as an energy carrier and electric powertrain concepts

The evaluation and down-selection of the right alternative propulsion technologies to replace fossil-fuel based powertrains is a matter of high societal and technological importance. Some voices state that battery electric vehicles could be a bridging strategy towards the final implementation of automotive hydrogen technology. By contrast, often within the alternative propulsion community, the impression is raised that an exclusive decision between either the case of hydrogen fuel cell technology (FCEV) or that of pure battery electric vehicles (BEV) has to be made.

From the authors' point of view, due to the extremely different energy densities of the utilized energy carriers, the need for an exclusive decision is definitely a misconception. There are optimal fields of application for each technology (*e.g.*, see Fig. 2 of ref. 2). A typical vehicle range of 500 km can be realized by using current gasoline or diesel engine technology without any problems. Today, modern diesel vehicles often even exceed the 1000 km range mark. Considering the diesel case, the tank system installed for an operating range of 500 km would only weigh *circa* 43 kg and a storage volume of less than 50 l would be needed. If a similar vehicle range is targeted for a pure battery electric vehicle, even if utilizing advanced Li-ion technology, the weight of the complete energy storage system would be close to 850 kg. Depending on the driving cycle, that battery mass would contribute to a very significant part of the total vehicle weight (see Fig. 1 and ref. 4a). The shown LA92 (total distance: 15.8 km, average speed: 39.6 km h^{-1}) and the FTP72 (urban route of 12 km with frequent stops; average speed: 31.5 km h^{-1}) cycles are typical test procedures used to evaluate the fuel economy of passenger vehicles in the USA. The Manhattan city cycle (maximum speed 40 km h^{-1}, average speed: 11 km h^{-1}) is usually applied to assess the efficiency of buses and is just provided for comparison to illustrate the effect of low average vehicle speeds. In contrast to any diesel or gasoline vehicle, which can be re-fueled within minutes, the re-charging of such a 100 kW h battery takes hours (even when applying a 50 kW DC fast charger → 2 hours), not to mention the required time when using conventional electrical outlets at home.

On the other hand, for a zero-emission vehicle powered by hydrogen, one has to install a tank system weighing *circa* 125 kg (based on 70 MPa compressed gaseous hydrogen technology). Such a tank system would contain 5 kg of hydrogen (or 200 kW h of stored chemical energy) and could be re-fueled within 3 to 5 minutes using existing technology. This important feature has been proven in large-scale vehicle demonstration programs by several car manufacturers, for example within the framework of the German "Clean Energy Partnership" (see also Section 3). Industry estimates show that the 70 MPa tank technology, considering high-volume production, could be produced for approx. US$ 3000.[2] For comparison: a 100 kW h Li-ion battery system providing the same 500 km zero-emission range would reach a production cost of US$ 30 000 to 50 000 (ref. 2) and would impact, as mentioned before, the total vehicle weight dramatically (see Fig. 1b and ref. 4*a*).

Due to this framework dictated by the energy density, the pure battery electric vehicle is most appropriate and could become commercially viable for urban mobility applications, using dedicated sub-compact car architectures (*e.g.* even unconventional two-seaters like the Renault Twizy), targeting reliable real-world ranges of up to 150 km. Many car manufacturers, mainly the Renault–Nissan alliance, Mitsubishi, Smart, Chevrolet and BMW have already started or are currently working on the market introduction of large-volume battery electric vehicles. Also advanced technology activities such as the GM EN-V two-wheeler concept for advanced urban mobility, as well as the smart-grid capable Opel Meriva MeRegioMobil BEV, establishing a bi-directional power flow between the electric vehicle and the grid, are ongoing.[3,4,6] E-REV vehicles (extended-range EV[3]), such as the Chevrolet Volt, the Opel Ampera or the Fisker Karma, are on the other hand appropriate vehicles for drivers who sometimes—but not as standard driving pattern—need longer ranges. In addition these customers have to be willing to accept a small internal combustion engine in order to ensure the range beyond the initial range of pure EV operation. Also plug-in hybrid electric vehicles (like the Toyota Prius, Ford C-Max Energi or the Volvo V60 PHEVs) address a similar segment of the car market.

By contrast, fuel cell electric vehicles (FCEVs) always operate as zero-emission vehicles and can be re-fueled quickly. In addition to vehicle ranges of 500 to 600 km at full performance, they offer also the possibility to realize family-sized cars, either as popular van-like vehicles in Europe (HydroGen3 → Opel Zafira) or as SUVs (HydroGen4 → Chevrolet Equinox).

2.2 Technology status of the HydroGen4

The current generation of automotive fuel cell technology has been tested and demonstrated on a larger scale throughout the world since about 2005. Over recent years, it could prove its viability and demonstrated very high average efficiencies and vehicle ranges. Details on efficiencies and vehicle usage are discussed in particular in Sections 2.4 and 3.2.1.

The detailed specifications of GM's and Opel's current 4th generation FCEVs are given in Table 1 and the main system components are shown in Fig. 2a. This cross-over vehicle is based on a standard Chevrolet Equinox and the structural modifications required to install and house the fuel cell powertrain, as well as the hydrogen fuel storage, are shown in Fig. 2b. Most of the modifications relate to the integration of the compressed gas vessels since, as shown in Fig. 2b, significantly more space is needed to provide the comparable range of an equivalent gasoline or diesel vehicle.

2.3 Hydrogen storage

As the first major sub-technology, hydrogen storage shall be discussed now. But since this topic is evaluated by the authors extensively elsewhere (*inter alia* in the first paper of this series[1]), the fuel storage part is kept deliberately brief. After extensive testing and in-house developments of the potential, finally a decision was made for 70 MPa compressed hydrogen gas (CGH2) as being the right solution for the market introduction of hydrogen technology. This fact is also representative of a wider automotive industry consensus. For those readers who are interested in further

Table 1 Technical specifications of the GM HydroGen4 (ref. 2)

GM HydroGen4	5-Door, crossover vehicle, front-wheel drive, based on the Chevrolet Equinox
Dimensions	
Length	4796 mm
Width	1814 mm
Height	1760 mm
Wheelbase	2858 mm
Trunk space	906 Liter
Weight	2010 kg
Payload	340 kg
Hydrogen storage system	
Type	3 type IV CGH2 vessels
Operating pressure	70 MPa
Capacity	4.2 kg
Fuel cell system	
Type	Proton Exchange Membrane (PEM)
Cells	440
Power	93 kW
Battery system	
Type	Nickel-metal hydride (NiMH)
Power	35 kW
Energy content	1.8 kW h
Electric propulsion system	
Type	3-Phase, synchronous motor
Continuous power	73 kW
Maximal power	94 kW
Maximal torque	320 Nm
Performance	
Top speed	160 km h^{-1}
Acceleration (0–100 km h^{-1})	<12 s
Range	320 km (standard fleet); 400 km (enhanced technology)
Operating temperature	−25 °C to +45 °C, vehicle can be parked at ambient temperature <0 °C (without external heating)

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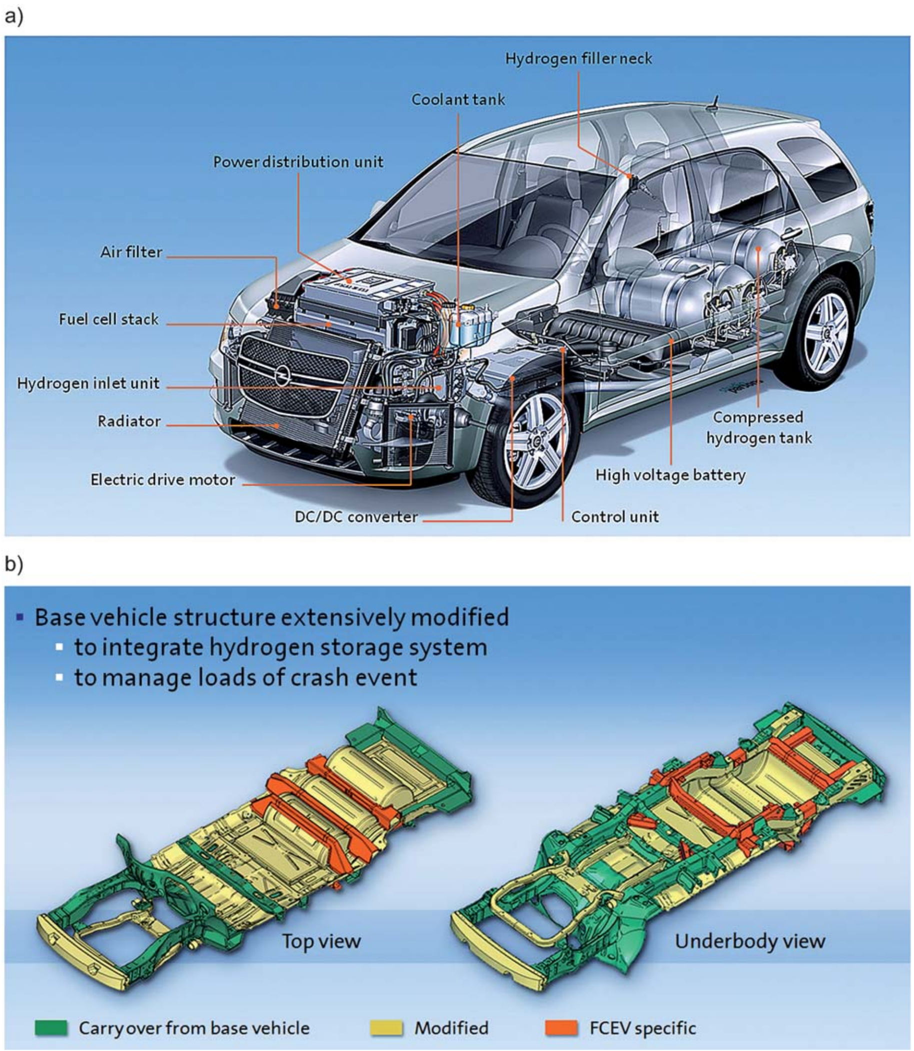

Fig. 2 (a) X-Ray drawing of the GM HydroGen4 and (b) structural modifications compared to a standard Chevrolet Equinox.

details (also including the technology status of solid state hydrogen absorbers), ref. 1 and 7 are highly recommended.

Currently, two types of CGH2 vessels are under discussion. They are the so-called type III and type IV tanks. A type III vessel consists of an inner metallic liner and sheets of carbon fiber composites. The liner prevents hydrogen permeation and the composite is required for the necessary mechanical strength and stability. In the case of a type IV vessel, the metallic liner is replaced by a plastic one, typically high-density poly-ethylene (HD-PET). GM and Opel opted to use type IV vessels since they provide major advantages:

- 20% lower weight with identical volumetric storage density

- higher potential regarding long-term fatigue and durability (virtually no liner cracking)

- lower-cost carbon fibers with a lower modulus of elasticity can be used.

As depicted in Fig. 3, material cost dominates, and even at a production level of only 10 000 tank systems per year, type IV tanks are by a factor of nearly 2.4 more cost efficient, compared to type III technology. This feature is also projected to be maintained when future developments and increasing production volumes are considered. Industry estimates show that using type IV tank technology and considering high-volume production, a 5 kg automotive H_2 system could be produced for US$ 3000.[2]

A good review on carbon-composite storage vessel technology, as well as on the production and validation processes, is to be found in ref. 8.

As shown in Table 1, the GM HydroGen4 utilizes three vessels containing a total amount of 4.2 kg of hydrogen. Since the overall range of a vehicle is not only dependent on the tank size but also on the fuel consumption and hence efficiency of the powertrain system, these numbers will now be assessed in the following part.

2.4 Automotive fuel cell system

2.4.1 Fuel cell stack fundamentals. The fuel cell stack represents the core component of the fuel cell power system. Today, membrane electrode assemblies (MEAs) consisting of a proton-conducting polymer membrane (proton exchange membrane, PEM) based on poly-perfluorosulfonic acid (PFSA) and carbon-supported platinum nanoparticles as catalysts (Pt/C) are state-of-the-art for automotive applications. The relatively high proton conductivity of PFSA

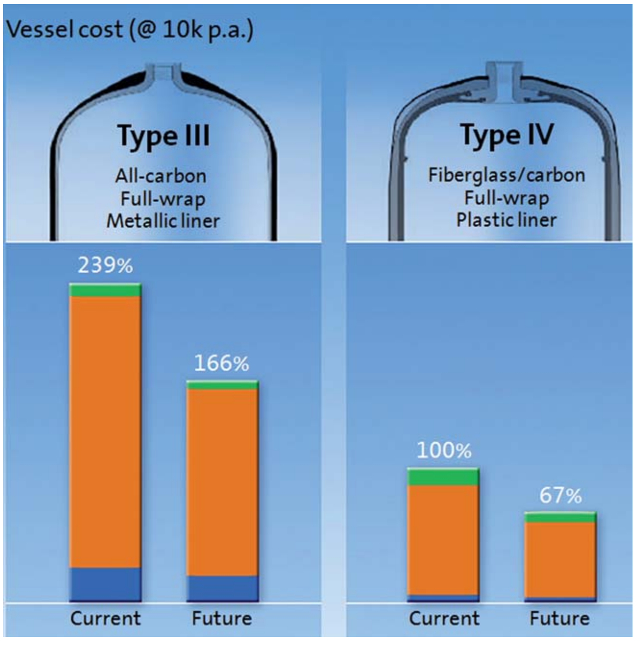

Fig. 3 Cost distribution for type III and type IV vessels; orange: carbon fiber composite; blue: liner; green: other.

membranes under automotive operating conditions (temperatures typically between 60 and 80 °C) in combination with the high activity of the Pt-catalyst enables fuel cell stacks to fulfil automotive performance targets. A schematic illustration of such a real-world cell built up of the MEA, diffusion media and bipolar plates, is shown in Fig. 8 of ref. 2.

The fuel cell is a direct converter of chemical into electrical energy: in contrast to a battery, the fuel is not contained at the electrode, but supplied to the electrode from a separate storage sub-system. As long as fuel and oxidant are supplied to the fuel cell at sufficient quantities, the generation of electrical energy is ensured. The challenge consists in evenly supplying all single cells of the stack with fuel and removing the reaction products (*i.e.* the product water) properly.

A major advantage of the fuel cell is its inherent high conversion efficiency, as that value is obviously independent of a combustion process and therefore also of the Carnot cycle limitation. However, a popular misconception exists (even among the educated public) that culminates in claiming that fuel cell efficiencies could reach maximum efficiency values of up to 100%, since there is no combustion process taking place.

The Carnot cycle efficiency represents the theoretical upper limit of a thermodynamic machine operating between a high temperature, typically achieved by burning fuel in the combustion chamber, and a low temperature which is somewhere between exhaust gas and feed gas temperature. In practice, the higher temperature is limited by material properties and heat losses. The lower process temperature usually cannot be decreased below the ambient temperature values (assuming air or water cooling). Consequently, efficiencies greater than about 55% are not possible, even in an almost perfect-world scenario (*e.g.*, the operating conditions of a modern large-scale thermal power station). For internal combustion engines, the maximum combustion chamber temperatures are in the order of 2900 °C and the exhaust gases are shoved out at about 1100 °C. For real-world applications, the theoretical efficiency is further reduced by additional technical losses to values below 40%. These extra losses are attributed to the system's inherent properties (such as heat losses, friction, and gas exchange losses) as well as to losses caused by auxiliary drives or components (such as oil and water pumps).

In the case of an automotive fuel cell, two gases – hydrogen and oxygen – are converted into water, and the difference in their chemical potentials is transformed into electrical work. Since a fuel cell does not *a priori* represent a thermal engine, no temperature difference between two reservoirs is needed for energy generation. Relevant are the internal energies of the chemical compounds involved and only the usable free energy is converted into electrical work. However, thermodynamic losses are always present due to the entropy release.

Under standard conditions, for a H_2/O_2 fuel cell, the reaction enthalpy and the Gibbs free energy show the following values:

- Reaction enthalpy ("max. heat"):
$$\Delta H = -286 \text{ kJ mol}^{-1} \text{ H}_2;$$
$$\Delta H/2F = -1.48 \text{ V} = -U_{th}$$

- Gibbs free energy ("max. work"):
$$\Delta G = -237 \text{ kJ mol}^{-1} \text{ H}_2;$$
$$\Delta G/2F = -1.23 \text{ V} = -U_{rev}$$

In the case of hydrogen as fuel, a certain amount of entropy is released in the form of heat during the formation of water molecules. The water molecule represents a state of higher order, and its entropy is reduced by heat emission. Theoretically, a fuel cell operating under standard conditions (T = 25 °C) achieves an ideal efficiency of $\Delta G/\Delta H = U_{rev}/U_{th}$ which equates to 83% (considering liquid water to be the waste product of the H_2/O_2 reaction). In a real-world electrochemical cell the maximum usable cell voltage U_{rev} is equivalent to the difference of the two standard potentials.

2.4.2 Real-world fuel cell system technology. A real-world fuel cell system (FCS) (see Fig. 4) consists – beside the electrochemical cells – of components which pre-condition and deliver the hydrogen and the ambient air to the fuel cell, *e.g.* the air compressor. The FCS, the power electronics and the electric traction system together constitute the fuel cell power cube which generates mechanical torque.

Since the introduction of the HydroGen series of vehicles by GM and Opel, the combined fuel cell and electric traction systems were packaged in a way so that they fit into the same volume as an internal combustion engine (ICE) module. Such a highly integrated power cube allows a simple and cost-efficient vehicle assembly in existing manufacturing facilities. Hence, this approach is a likely

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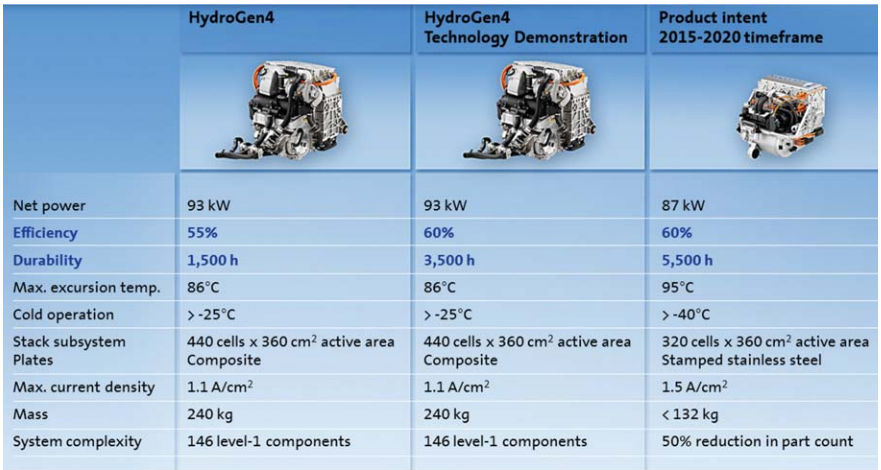

	HydroGen4	HydroGen4 Technology Demonstration	Product intent 2015-2020 timeframe
Net power	93 kW	93 kW	87 kW
Efficiency	55%	60%	60%
Durability	1,500 h	3,500 h	5,500 h
Max. excursion temp.	86°C	86°C	95°C
Cold operation	> -25°C	> -25°C	> -40°C
Stack subsystem Plates	440 cells x 360 cm^2 active area Composite	440 cells x 360 cm^2 active area Composite	320 cells x 360 cm^2 active area Stamped stainless steel
Max. current density	1.1 A/cm^2	1.1 A/cm^2	1.5 A/cm^2
Mass	240 kg	240 kg	< 132 kg
System complexity	146 level-1 components	146 level-1 components	50% reduction in part count

Fig. 4 GM fuel cell power system design evolution and main requirements; from the HydroGen4 (introduced in 2007) to the product intent system of the 2015–2020 timeframe.

technology scenario for the introduction of FCEV volume production on the basis of existing vehicles' platforms. There is, however, no principal technical restriction that would rule out a different configuration of the fuel cell powertrain components on board of the vehicle (*e.g.* see Fig. 9a of ref. 2). The scalability of fuel cell technology also enables the adaptation to different vehicle sizes. For example, the propulsion system that was originally developed for the GM HydroGen3 van eventually was adapted to a small vehicle, the Suzuki MR Wagon FCEV, using a shorter fuel cell stack with a reduced cell count (*e.g.* see Fig. 10 of ref. 1).

Worldwide, already more than 10 000 customers drove the 119 HydroGen4 vehicles used in four countries (30 of these are operated within the "Clean Energy Partnership" in Germany). The vehicles went through a total road performance of over 4 million km (status as of mid-2012). For detailed numbers see Section 3.2.1.

Within the framework of "Project Driveway", the fuel cell powertrain of the HydroGen4 fleet has been very successful in demonstrating performance, customer usage and satisfaction. Nevertheless, the product intent fuel cell system (2015–2020 timeframe) has even higher requirements (see Fig. 4 and 5) in terms of efficiency, durability and cost. High powertrain efficiency is needed to enable an acceptable vehicle range with a modestly sized hydrogen tank system and low well-to-wheels CO_2 emissions. The durability and cost targets are driven by the end customers' expectations of what is acceptable for a passenger car. These expectations are related to what the driver is familiar with from the conventional gasoline or diesel technology.

In particular, the cost needs to decrease considerably until commercial introduction by the 2015–2020 timeframe. The target value, considering large-scale production, is as low as about one quarter of the cost (see Fig. 5a) for the HydroGen4 design of 2007. Besides economies of scale (manufacturing, parts sourcing, *etc.*), the amount of expensive materials used in the stack is of great importance: the platinum content of 80 grams per hydrogen fuel cell system, for example, is expected to decline by 2015 to about 30 grams. Moreover, in laboratory experiments, it could be proven that in the long run (*i.e.*, the 2020–2025 timeframe) it is possible to reduce the platinum content to less than 10 grams. This figure corresponds to the amount of Pt used in current automotive three-way catalytic converters. A roadmap to achieve such low values is shown in Fig. 5b: the application of various types of platinum-alloy technology based on nano-particles plays an important role here.

Other key factors are an improved system integration, the introduction of proven best-practice manufacturing methods adopted from conventional large volume production processes, a technology transfer from conventional powertrain development programs, as well as synergies with other GM hybrid and electric vehicle programs. Doing so, by 2015, it is possible to realize a system (see Fig. 4) that is only half the weight and requires just half the volume of today's HydroGen4 system while using just 50% of the current number of level-1 components.

The major change between the HydroGen4 fuel cell stack of the 2007 vehicle generation and the product intent system for the 2015–2020 timeframe is the highly integrated stack design and the application of dedicated and specifically developed automotive components from established suppliers. Typical automotive design cycles take 4 to 5 years. *Id est*, the powertrain technology selected for a mass production vehicle needs to be stable, mature and reliable five years before the first cars are handed over to end customers. Therefore, the above-mentioned product intent system is already today well-defined and only existing, as well as proven, technology is used. System testing on powertrain level is ongoing.

The cell count will be reduced from 440 cells down to 320 cells with an active area of 360 cm^2 resulting in a higher required maximum current density of 1.5 A cm^{-2} and excursion temperature of up to 95 °C.

Furthermore, the bipolar plate material will be changed from carbon composite to stamped stainless steel. The thermal mass of the stack will be decreased and the repeat distance between individual cells reduced, facilitating an improved freeze start-up capability and packaging. Enabling operation down to −40 °C is required to be competitive to internal combustion engines.

In order to demonstrate the maturity and viability of the required technology, GM initiated the "Technology-Demonstration-Fleet" (or briefly "TechDemo")

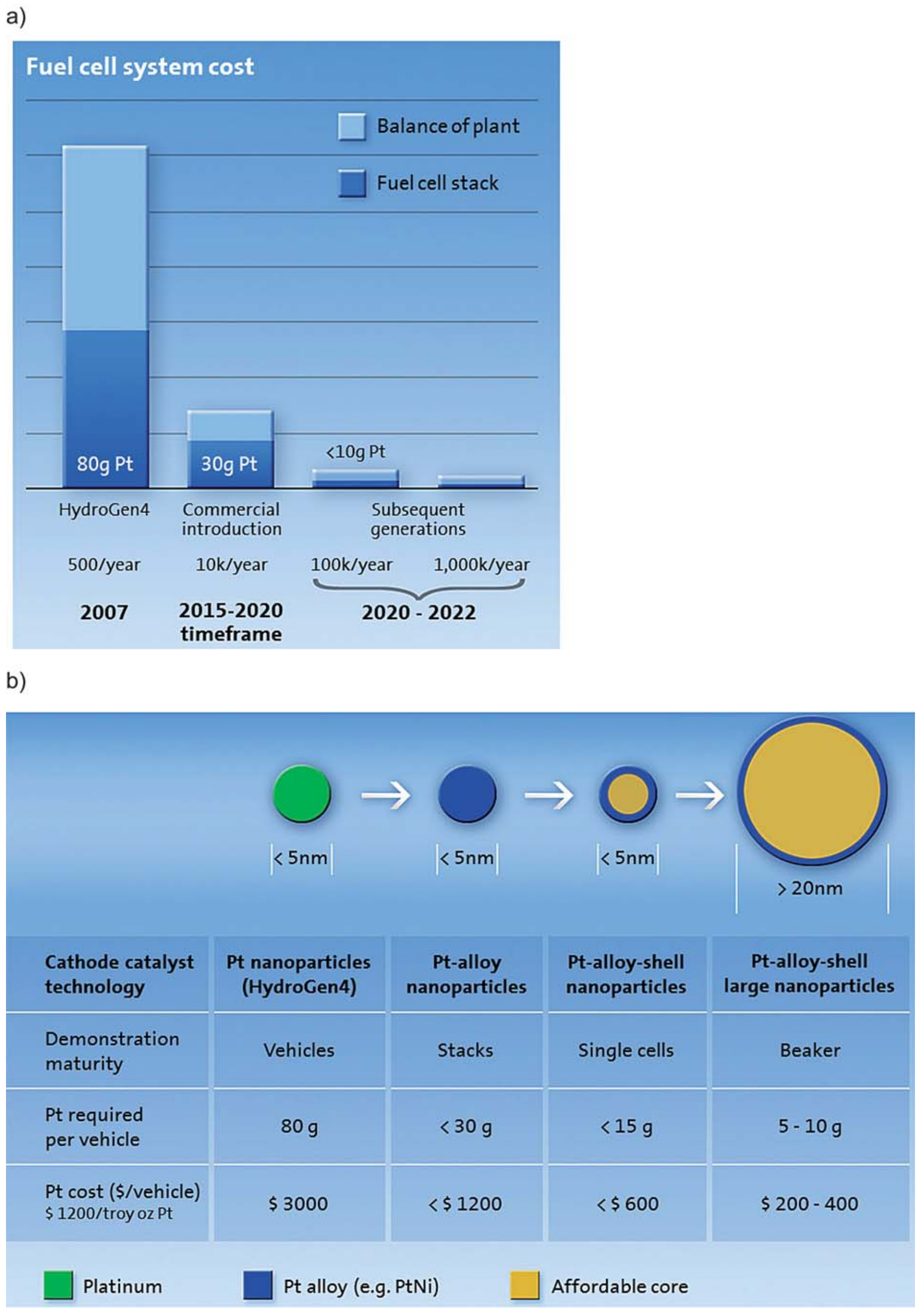

Fig. 5 (a) Fuel cell system cost, fuel cell stack (dark blue) and balance-of-plant components (light blue) and (b) long-term platinum reduction roadmap until 2025.

project. This engineering fleet based on the HydroGen4 vehicles focuses on two main initiatives: (1) to significantly increase the overall efficiency in the drive cycles as defined by the US Environmental Protection Agency EPA and (2) to demonstrate 3500 h stack life in the vehicle to close the gap towards auto competitiveness which requires 5500 h of operation during a vehicle lifetime.

In addition, this development step is also used to validate new control functions and optimize the performance of the fuel cell system (FCS) itself. The relative humidity (RH) sensor that is used in the cathode subsystem of the conventional HydroGen4-systems is removed. The humidity control is now implemented as a model-based approach that allows the elimination of the fault-prone, physical sensor. Furthermore, the calibration of the FCS start-up sequence is extended in such a way that the current ambient temperature (as well as the length of system off-time) is considered for starts below freezing level. As a result, if the system is not completely frozen, the start-up time of the FCS could be reduced from a maximum of 30 s down to 7 s. Also, the occurrence of so called "fail-safe starts" is reduced. In contrast to the standard HydroGen4-vehicles where a component failure leads to a shut-down of the whole system, the "TechDemo" system considers additional information. If the hydrogen and oxygen concentrations are known, the

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system is in a well-defined condition, and the subsequent start-up can be performed according to the standard procedure. By contrast, if the gas concentration is unknown, the system conducts a reliable "fail-safe start" which takes more time.

The first major goal of the "TechDemo" initiative is to increase overall efficiency. In addition to the aforementioned advantages of fuel cells originating from basic thermodynamic considerations, the high efficiency of a fuel cell powertrain is in particular caused by the fact that a fuel cell reaches its highest efficiency at part load where most driving takes place. On the other hand, under full load conditions and at high speeds, fuel cells are just as efficient as internal combustion engines. Also at very low power output, the fuel cell system efficiency decreases sharply since many balance-of-plant components have to be running even at idle power. All these effects can clearly be seen in the tank-to-wheels efficiency of the GM HydroGen4 standard fleet version² given in Fig. 6.

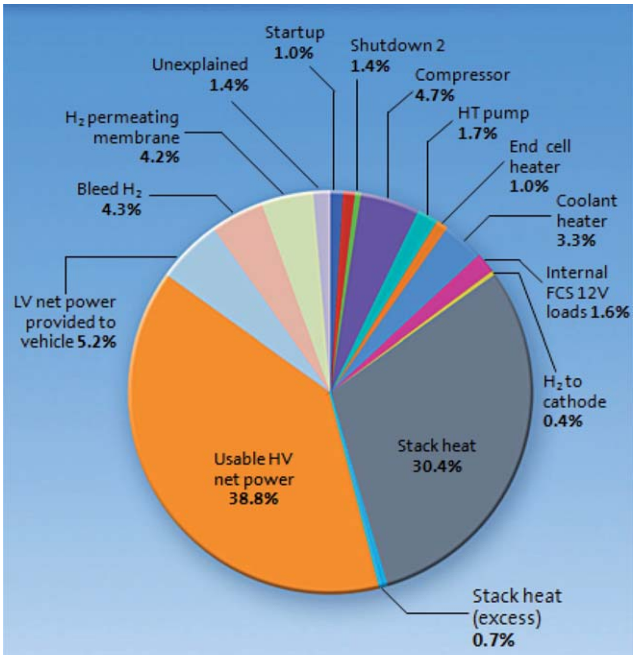

Fig. 7 Energy flow analysis of a HydroGen4 fuel cell system.

The standard fleet vehicles proved to be more efficient than the comparable conventional Chevrolet Equinox vehicle using a gasoline engine by a factor of about 2 (EPA composite cycle 4.6 L per 100 km of gasoline equivalent in comparison with 9.6 L per 100 km of gasoline). As mentioned before, the advantage compared to the internal combustion engine (ICE) is maximal between 20 and 70 km h^{-1}.

A detailed analysis of the different energy flows was executed (see Fig. 7) for the FCS in the HydroGen4 vehicles. The most energy-consuming activities, sequences or components were identified, analyzed and systematically improved.

The thermal energy for heating the coolant is significantly reduced by decreasing the minimum coolant inlet temperature while ensuring that the interior comfort is not affected. End-cell heaters, located between the endplate and the first active cell of the stack, which secure a stable operation of these cells during warm-up, are completely shut off after the system reaches its operating temperature.

In the cathode subsystem, the minimum compressor speed is reduced and the air volume procedure during the shut-down sequence is now implemented as a function of the ambient temperature. As a result, all product water is removed for temperatures below the freezing level. At higher ambient temperatures, some product water remains in the system and is blown out during the subsequent start-up event. All these measures result in a total fuel efficiency gain of 20% quantified *via* city and highway drive cycles defined by the United States Environmental Protection Agency EPA compared to the standard HydroGen4 fleet. This improvement corresponds to a range extension by 80 km to about 400 km in the combined EPA cycle on 4.2 kg hydrogen.

Fig. 8 shows the efficiency gain as a function of the FCS net power. Whereas the "TechDemo" stack with its modified membrane shows a virtually identical polarization curve, the FCS efficiency is improved substantially. Especially in the net power range between 5 and 15 kW, which is dominant in typical driving patterns, the efficiency is significantly enhanced to values up to 60%.

The second major goal of the "TechDemo" initiative is to increase the fuel cell stack lifetime in a vehicle. Fig. 9 shows the mileages and operating lifetimes of the different vehicle generations. The HydroGen3 propulsion system showed a B50 value of only 250 hours (*i.e.*, 50% of

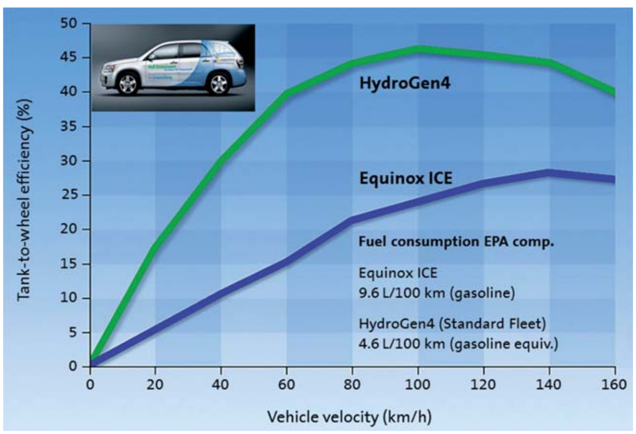

Fig. 6 GM HydroGen4 standard fleet version (green curve) and Chevrolet Equinox gasoline ICE (blue curve) tank-to-wheels efficiency over vehicle velocity.

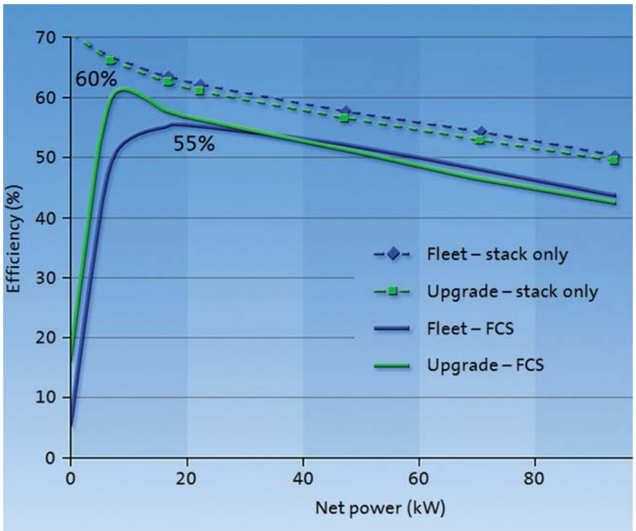

Fig. 8 Fuel cell stack efficiency (dotted lines) and fuel cell system efficiency (straight line) over net power; HydroGen4 standard fleet version: blue curves; "TechDemo" system: green curves.

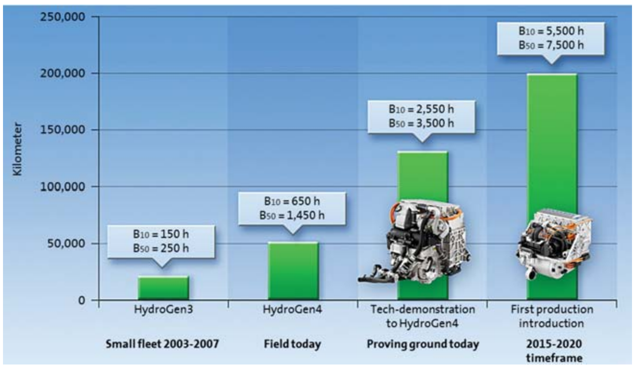

Fig. 9 Stack-in-vehicle durability of various generations of GM fuel cell systems.

the sample reached a lifetime greater than 250 hours) while the B10 value was 150 hours (10% of the sample was broken in less than 150 hours). The mean lifetimes and mileages made a big step forward with the current standard fleet HydroGen4 (1450 hours). Another significant improvement is projected for the 2015–2020 timeframe powertrain design. This system will show a mean lifetime of 7500 hours and a total mileage of 200 000 km.

Already 10 years ago, stack operating hours of several thousand hours were demonstrated in a lab environment where steady-state, well-humidified and continuous operating conditions are dominant. However, operating modes in a vehicle on the road are completely different, for example transients and startup–shutdown sequences are in fact prevalent. In the case of earlier vehicles, such as the HydroGen3 (see ref. 1) of 2002, processes that impact stack durability were insufficiently understood and therefore the fuel cell system control strategy did not consider these damaging events or conditions.

The "TechDemo" initiative takes a close look at the main stack failure modes in mobile applications namely mechanical and chemical membrane degradation, voltage loss during operation, as well as corrosion of the catalyst support material. Furthermore, the fundamental mechanisms for all failure modes are investigated based on system data analysis and tests on a material level. Based on the identified root cause, applicable counter measures were developed on a FCS level, implemented in the system control software, and validated on a vehicle level.

(a) Membrane degradation. The FCS in a vehicle on the road is operated dynamically which leads to variable humidification levels of the polymer electrolyte membrane in the fuel cell stack. Field data analysis revealed occasional RH-transients as low as 10% RH and up to >100% RH (i.e., liquid water). Fig. 10 shows for the example of Nafion-112 the volume expansion in % as a function of the relative humidity level.

A change in humidity from dry to 60% leads to a membrane volume expansion caused by swelling of about 15%. If liquid water is produced, the expansion is even greater than two thirds of the original thickness. Since the membrane is at least partially constrained in the fuel cell by the electrode, the gas diffusion layer and the bipolar plates, the frequent expansion and contraction creates substantial mechanical stress. These processes can initiate and propagate microscopic cracks that result in the crossover of the reactant gas leading to individual cells dropping out, causing a complete shut-down of the fuel cell system.

Start-up, idle operation, but also technical issues concerning the sensor, can create system conditions in which the membrane can dry out or is exposed to high voltages. These conditions promote the chemical degradation in which radical species as byproducts or side reactions of the electrochemical reactions are formed and directly attack the polymer membrane causing polymer decomposition. This continuous thinning of the membrane increases again the gas crossover and can cause a complete shut-down as described above. Ref. 9 gives an in-depth look at this issue. The product intent FCS will use a newly developed composite membrane in combination with a more humid stack operating strategy in order to overcome the mechanical and chemical degradation of the polymer membrane.

(b) Voltage degradation. Over time, stack voltage loss rises continuously due to increasing ohmic resistance, for example by corrosion of the bipolar plates or increasing contact resistance values between individual components. In addition, mechanisms at the catalyst level contribute to irreversible voltage losses. Thermodynamically driven platinum particle growth reduces the electrochemically active catalyst area and hence increases the cell overpotential leading to a lower stack voltage at a given current density. On the one hand, Pt dissolution at temperatures relevant to PEFCs (80 °C) is

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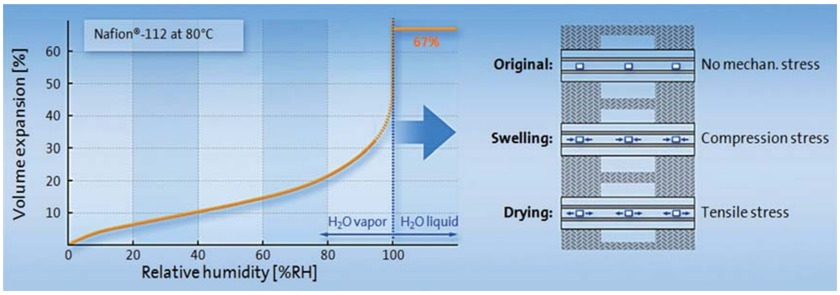

Fig. 10 Volume expansion of Nafion-112 at 80 °C as a function of relative humidity (RH).

accelerated at high steady-state potential (0.9 to 1.1 V *vs.* RHE) or fuel cell idle operation. On the other hand, this process occurs if the platinum catalyst is cycled between its metal- and oxide-phase or the cell potential in a real-world fuel cell changes from more than 900 mV to less than 800 mV or *vice versa*. These voltage transients enable the accelerated formation of oxidized platinum species. The real coarsening of Pt particles occurs *via* two different processes (see Fig. 11): a nanometer scale Ostwald-ripening process where smaller Pt particles dissolve in the ionomer phase and redeposit on larger Pt particles and a micrometer-scale diffusion process, where dissolved Pt species diffuse in the ionomer phase and subsequently precipitate in the ionomer phase of the electrode or the membrane *via* reduction of Pt ions with cross-over hydrogen from the anode (see ref. 10 for details).

Obviously, neither dynamic nor idle operation can be avoided in a vehicle on the road. In fact, these operating conditions are dominant in mobile applications. Therefore, the FCS stand-by mode and the so-called voltage-suppression-functionality are implemented on the system level to mitigate dwell times at high cell voltages and excessive voltage cycling between 800 mV and 900 mV. The FCS transitions into a stand-by mode (*id est*, the system is turned off) if certain conditions are given: *e.g.* when the vehicle speed or acceleration request is below a specific threshold. In these cases the electrical energy is provided by the high voltage battery. The voltage-suppression mode is active when the FCS is operated in idle- or low-load-operation. Thereby, electrical components like endcell- or coolant-heater are engaged in order to pull the cell voltage below the critical value of 875 mV. Both functions reduce the number of harmful voltage cycles and the time periods at high potential, which minimizes the loss of the electrochemical active platinum area and hence significantly reduces the voltage loss of the fuel cell stack.

Carbon corrosion of the catalyst support material is another mechanism which leads to irreversible voltage losses in the fuel cell. Hydrogen/air-fronts in the anode-compartment during startup/shutdown cycles especially after long off-times can lead to short-term potential excursions of the cathode electrode to 1.2–1.5 V.[11] At these high potentials, corrosion rates of standard carbon-supports are very high. Thus, free floating Pt-particles emerge and coalesce to larger particles, consequently reducing the electrochemically active platinum surface and leading to an additional voltage loss. Mobile applications require an estimated 35 000 startup/shutdown cycles over the life of a vehicle including extended parking time without having any significant voltage loss. Therefore, the objective must be to eliminate the root cause; in this case the high, corrosive potential or the hydrogen/air-fronts in the anode-compartment. The "TechDemo" fuel cell system accomplishes this by introducing two functions: the so-called oxygen-depletion-function is in operation during the shut-down. It actively reduces oxygen in the cathode path by turning on electrical consumers like the coolant-heater and operates the stack at an oxygen stoichiometry of one. This minimizes the amount of oxygen diffusing from the cathode to the anode and avoids accumulation of large amounts of oxygen in the anode-compartment.

The so-called "hydrogen-in-park"-function is engaged during long vehicle off-times. Small amounts of hydrogen are injected into the anode to chemically react with potentially diffused oxygen in order to prevent the build-up of a hydrogen/air front.

Fig. 12 shows the effect of an oxygen-depletion shutdown and the anode concentration after a shutdown event.

As long as hydrogen is present at the anode, the oxygen concentration equals zero. Only when the hydrogen is completely consumed, oxygen diffuses from the cathode and slowly accumulates at the anode. Now, the oxygen cannot react chemically any more with the formerly present hydrogen. Comparing the green curves one can clearly see that the dedicated O_2-depletion shut-down process postpones the point in time when oxygen can start to accumulate. The anode hydrogen blanket persists for four additional hours (9 h *vs.* 5 h after shut-down). That point in time, when the blanket has completely disappeared, is ideal for the first hydrogen injection.

Oxygen-depletion and hydrogen-injection as part of the FCS control strategy ensure that no hydrogen/air-fronts are built and the subsequent startup of the vehicle does not damage the catalyst support material.

Catalyst contamination is another root cause for voltage losses for fuel cells in mobile applications. Before the hydrogen and oxygen gases reach the location where the actual fuel cell reaction takes place, they come into contact with several materials used in components of the anode- and cathode-subsystem. Even though all materials are well-chosen, sometimes there is a trade-off between cost efficiency and functionality. For example, in order to achieve cost targets,

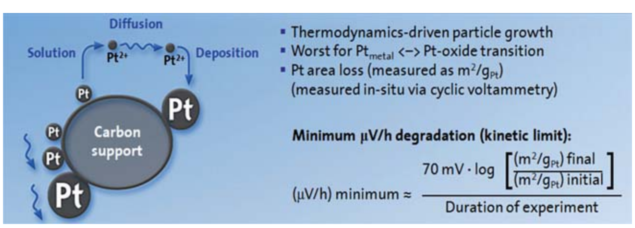

Fig. 11 Coarsening of Pt particles *via* two different processes due to voltage cycling.

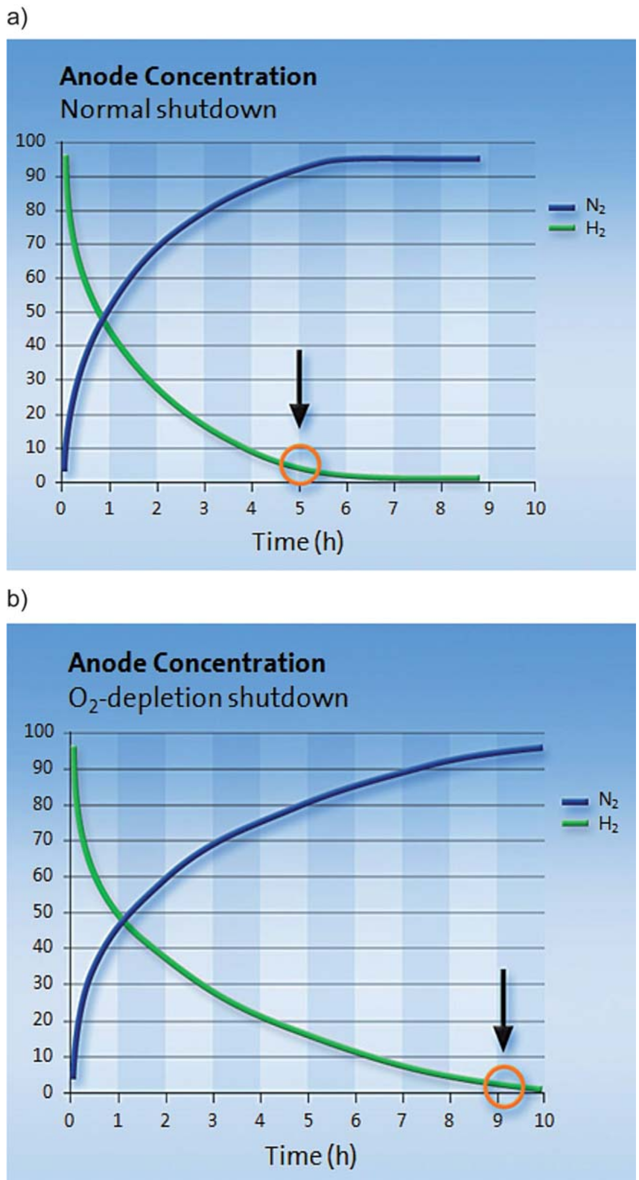

Fig. 12 Anode concentration (N_2, H_2) after shutdown, (a) without and (b) with O_2-depletion function turned on.

materials were chosen which enable contaminants to arrive at the catalyst layer, to adsorb on the surface, to decrease the electrochemical active surface and hence to reduce the overall electrode performance. Fig. 13 shows the impact of contaminants on the fuel cell polarization curve using the example of ethylene glycol (an automotive coolant).[12]

For the first test, 50 ppm glycol are injected on the anode and for a consecutive test on the cathode of a 50 cm^2 lab cell. The contamination leads in both cases to a significantly lower voltage–current curve (red curves). At the cathode, the polarization curve even collapses completely, *id est*, no current can be drawn beyond 0.4 A cm^{-2}. However, these lab tests also show that the anode, as well as the cathode, can be completely recovered from ethylene glycol poisoning by cyclic voltammetry (CV). After the second sweep the performance is fully recovered (light blue curves).

In order to remove contaminants from the electrodes in a vehicle environment the standard shutdown procedure is modified. The calibration is set in a way that the anode, as well as the cathode catalyst, is exposed to high and low potentials. All relevant contaminants are either oxidized or reduced and eventually flushed out. Fig. 14 shows the effect of the implemented automated stack recovery procedure.

Voltage loss (solid orange line) is observed over time in a durability test for the average cell voltage of a complete FCS in a lab-environment. However, the stack voltage recovery procedure (blue arrows) yields a steep increase of the cell voltage. After each recovery step the stack was removed from the FCS, mounted on a test stand and underwent a polarization curve (dotted orange line) to verify the recovery. Now, one can clearly observe two rates of voltage run degradation. The irreversible portion is mainly caused by unavoidable large voltage cycles (*e.g.* harsh acceleration), whereas the reversible part of the total voltage loss can be recovered by the implemented procedure.

GM's "TechDemo" initiative is an important engineering step towards the volume production of fuel cell vehicles in the 2015-2020 timeframe which demonstrates a B50 stack lifetime in a vehicle of 3500 h. Therefore it was absolutely essential to get an understanding of the interaction between vehicle operation and stack operating conditions. To do so, the dynamic operation, start- and shutdown-events, as well as processes during long off-times were investigated. Hurtful events were identified and correlated with the stack failure modes. Eventually, appropriate countermeasures, initially developed in a lab environment, were implemented successfully in the vehicle operating strategy. In particular, the above-described standby-mode, the voltage-suppression- and oxygen-depletion-function, as well as hydrogen injection during long off-times and the automated stack recovery procedure were implemented to significantly improve the stack durability in vehicle operation.

3 Infrastructure build-up and commercialization efforts

3.1 Hydrogen and fluctuating renewable energy

3.1.1 Automotive boundary conditions. For battery technology and especially electric grid stability reasons, charging times of at least one to several hours or even longer periods and the utilization of a sophisticated smart charging communication protocol are required to avoid extremely high power

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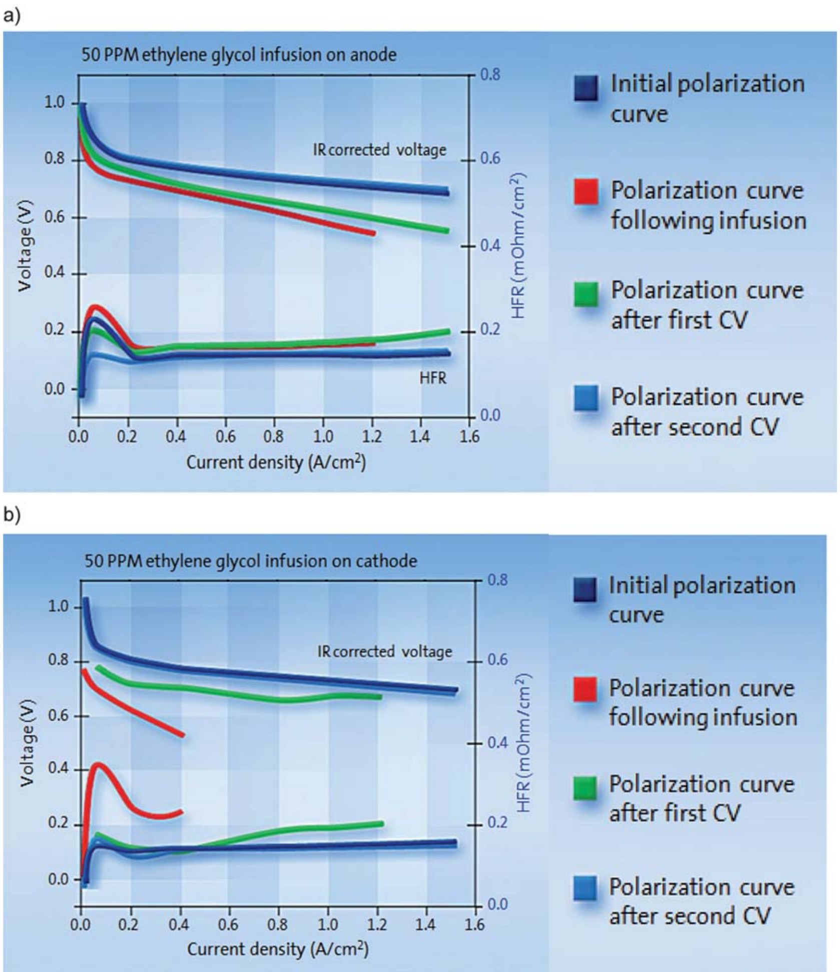

Fig. 13 Impact of 50 ppm ethylene glycol on (a) anode and (b) cathode of a 50 cm^2 lab cell on voltage and membrane resistance (HFR: High Frequency Resistance).

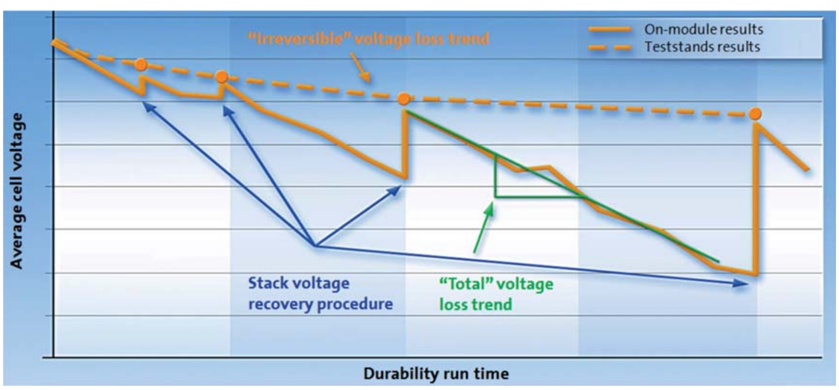

Fig. 14 Effect of the automated stack recovery procedure on the cell potential over time.

loads. Such technologies and protocols are tested with the Opel Meriva BEV in the MeRegioMobil and iZEUS projects.[3,4,6] In cooperation with two utilities and a major software company, MeRegioMobil and the succeeding iZEUS project have set up a complete smart home infrastructure at the Karlsruhe Institute of Technology and operate smart-grid-capable EVs from Daimler and Opel. Within the project framework, three Opel Meriva BEVs were built. These cars are capable of establishing a bi-directional power flow[6] between the grid and the vehicle for the first time in the world. This technology enables smart-charging and even the usage of the vehicle battery as a buffer for electric energy controlled by the grid operator or the utility.

Uncoordinated charging of one million EVs (from a total German car parc of 40 million vehicles) at a single point in time in the early morning or the evening would create a power demand of 3.5 GW when using standard German home sockets (at maximum 3.5 kW) and infrastructure installations. Obviously, even this modest fleet would cause a very substantial challenge when the charging takes place without coordination by the grid operator, *e.g.* as mentioned above through sophisticated smart-grid technology.[6] To put this into perspective, one single large-scale base load power station generates typically only about 1 GW of electric power. In the year 2010, all German nuclear power stations combined provided *circa* 20 GW to the grid, producing about 23% of the German electricity demand. Another challenge emerges also for the public infrastructure: a standard public charging point becomes blocked for hours by just one customer. Thus, for EVs, there exists a strong inter-dependency between two for conventional ICE vehicles distinct activities, namely "parking" and "re-fueling". A typical EV for urban applications should have a range of at least 100 km and consumes about 20 kW h per 100 km under real-world conditions including charging losses. An average daily driving profile could require a vehicle range of 40 km, *i.e.*, 8 kW h of electric energy need to be re-charged per day. Considering a standard German outlet with a power level of about 3.5 kW, that procedure would take 2 to 3 hours. The mentioned inter-dependency is not the case for fuel cell electric vehicles (FCEVs) where a typical re-fueling process takes only three to five minutes. Therefore it is essential to consider not only pure battery electric vehicles but also FCEVs for a future sustainable transportation system,[2] depending on driving profile and application. Additionally, hydrogen offers a different and very important advantage. A typical 70 MPa compressed gaseous hydrogen vessel has a system energy density of about 1600 W h kg^{-1} compared to values of significantly less than 200 W h kg^{-1} of usable energy for even the most advanced automotive battery packs (see Table 1 of ref. 1 and ref. 7 on hydrogen storage densities). Because of this comparatively high energy density due to its nature of being a chemical energy carrier (such as gasoline and diesel), hydrogen, methane or other "designer energy carriers" could serve as the ideal partner for the intermediate storage of fluctuating, renewable energies. In doing so, excess amounts of sustainable energy sources such as solar and wind power can be made available not only for stationary but also for automotive applications if fuel cell technology and infrastructure would be available at reasonable cost in the 2015–2020 timeframe.

3.1.2 Available options. As a case study, the largest German electric grid, the so-called "TenneT control area" (formerly known as "E.ON control area") is examined. This grid reaches from the North Sea through central parts of Germany to the Alps. In October 2008, the power fed into the grid by wind turbines fluctuated significantly (see Fig. 15). *E.g.* on October 5 and 6, 2008, within a few hours, the feed-in dropped from about 8000 MW to virtually zero. Unfortunately, such weather conditions occur on a regular basis.

By contrast, an excess amount of available wind power caused at several points since 2009 extreme effects on the energy markets, such as significantly negative prices of down to minus 25 ct kW^{-1} h^{-1} for electric energy at the European Energy Exchange (EEX).

In the future, this kind of challenges will gain in importance since the share of fluctuating renewable energy is expected to rise substantially. According to the German wind energy industry association, the nationwide total installed wind power capacity has risen in 2010 to 27 215 MW, a 5.6% increase over the respective 2009 value.[4,13] In the first 9 months of 2011, wind power contributed already on average about 8% (solar power accounts for 3%) of the German gross electricity production.[14]

But not only these short-term fluctuations need to be covered. Wind power shows also significant seasonal dependencies regarding the electric energy generation. During the winter half-year in Germany, typically 3.5 TW h of wind energy were generated per month (based on 2003-2009 statistical data), while during summer this number drops on average to values below 2 TW h.[4,14] Even more dramatic seasonal dependencies are observed for solar energy (May 2011: 2.5 TW h, January 2011: 0.3 TW h).[14] Furthermore, the amount of solar power fed into the German grid increased significantly in the first half of 2012. As peak value, more than 20 000 MW were produced around noon on sunny days in May 2012.

Solving these issues will become even more important and urgent when the planned off-shore wind farms (*e.g.*, in the North Sea) will gradually come online later this decade. And these developments are of particular importance after the Fukushima accident in 2011. As a reaction, the German government decided to phase out the nuclear power stations providing a major share of the nation's baseload. For compensation, the government intends to increase the share of fluctuating renewable energy under the "Energiekonzept" plan to 35% in 2020 and 80% in 2050.[14] Unfortunately, those technologies feature low annual utilization numbers (see Fig. 15). An additional challenge for the grid stability emerges from the fact that, typically, wind power installations are located in the sparsely populated coastal areas close to the North and Baltic Seas. By contrast, solar power is mainly fed into the grid in Germany's south and also the population and industry centers are to be found mainly in the western state of North Rhine-Westphalia and the southern states of Hesse, Baden-Württemberg and Bavaria. Both facts are leading to a very significant energy transport challenge. During the 2011/2012 winter, the current German grid infrastructure was already operated extremely close to its limits. The

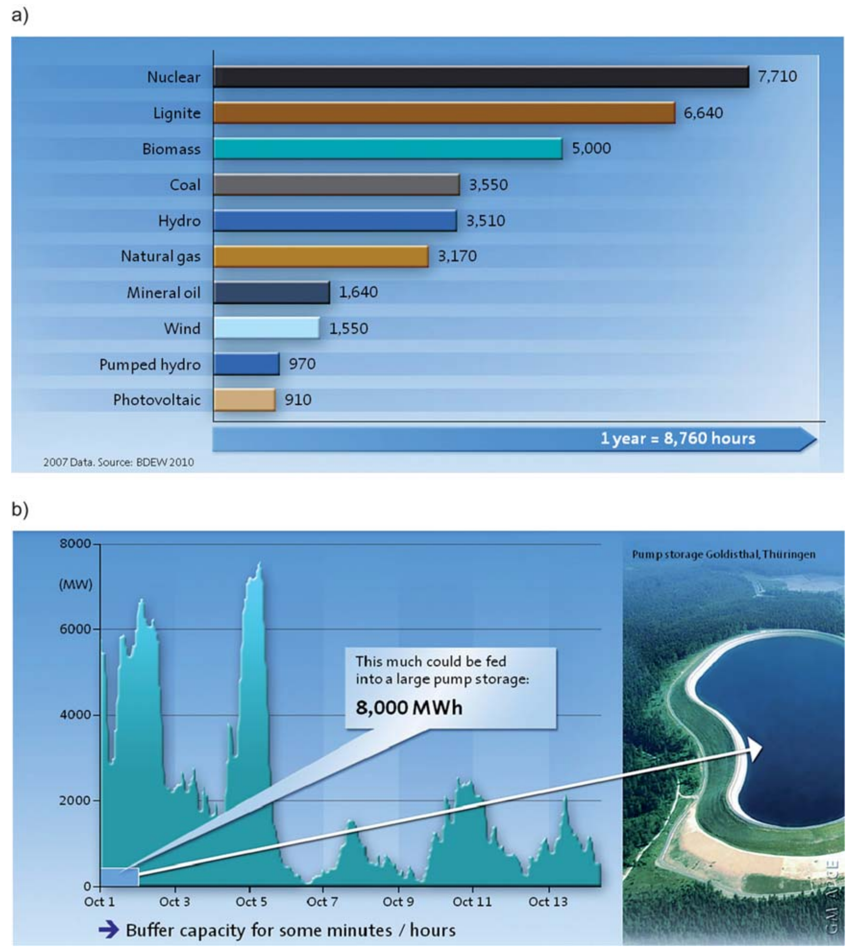

Fig. 15 (a) Typical annual utilization of power station capacity in Germany, 2007;[14] and (b) fluctuating wind energy in October 2008 in the grid operated by TenneT compared to biggest German pumped storage hydro power station Goldisthal (a Vattenfall installation in the state of Thuringia[2,15]).

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stability could only be maintained by using an "emergency back-up" concept. Temporarily, old (*i.e.*, low-efficiency) conventional power stations had to be re-started, significant additional energy imports at very short notice, and further extraordinary measures and interventions by the grid operators, as well as the government regulatory authority, were required.

A "buffer" to handle these issues and store excess energy to cope with the supply fluctuations would be extremely helpful to energy producers and grid operators. Such technologies would also enable an option to provide energy back to the grid during high market demand phases. The only commercially viable option for this kind of intermediate storage facilities is currently realized as pumped storage hydro power stations. Goldisthal,[15] the largest facility in Germany, offers a maximum storage capacity of 8000 MW h. Another proposed technology for this application is compressed air reservoirs. Salt caverns in the Northern German plains could be utilized. These caverns are typically used to store natural gas. Considering a cavern with an operating volume of two million m^3, about 4000 MW h of electric energy could be stored. At the moment, there are only experimental – and no commercial – facilities in operation since this storage dimension is even lower than what is achievable with the current standard technology, namely pumped storage hydro power stations.

In case hydrogen would replace compressed air as a storage medium[2,16] up to 600 000 MW h of energy, corresponding to about 3.6 million FCEV tankfuls, could be stored in an identical salt cavern. Unlike conventional pumped storage hydro technology, hydrogen is able to offer far more than just an energy buffer for a few minutes or hours. The excess wind energy of several days could be absorbed in such a large-scale installation.[16] Eventually, there are two paths available for the usage of the stored gas. It could be either converted back into electrical energy or could serve as a renewable fuel for fuel cell electric vehicles.

Also often proposed is the concept of battery electric vehicles that could be used as energy store or controllable load while parking. But due to the comparatively low energy density of automotive Li-ion batteries, even large fleets of smart-grid capable EVs are not able to provide a viable order of magnitude of energy storage capacity. For durability and customer ease-of-use considerations, 10% of the total nominal energy content should not be exceeded. If 8000 MW h of electric energy needs to be stored and 5 kW h of the usable energy content of an EV battery could be externally controlled by the utility, already 1.6 million EVs would be needed. Considering a smaller battery pack typical for urban vehicle applications, the required fleet size even needs to be significantly larger. If only a value of 2 kW h could be used, that number would increase to about 4 million EVs. And these 1.6 and 4 million EVs, respectively, would just replace one established pumped storage hydro power station like Goldisthal. Also, other large-scale stationary batteries – Na–Sulfur chemistries are commercially most widespread – cannot provide energy storage dimensions comparable to a hydrogen-based system[2,4a,16] since the H_2 system energy density is still far superior as explained in Section 3.1.1., see also ref. 1 and 7.

Since the set-up of a viable and sufficiently dense hydrogen infrastructure in the short term is considered a significant challenge by virtually all major stakeholders, concepts have been presented over the recent years to utilize synthetic natural gas (SNG) as chemical energy carrier (in particular by Specht and Sterner[17a]). Also under this concept, as a first step, fluctuating renewable energy is used for the electrolysis of water. The produced hydrogen and CO_2 (taken from a CO_2 producing industrial or biogas facility) react *via* the well-known Sabatier process to form methane (in this case also known as synthetic natural gas):

$$4H_2 + CO_2 \rightarrow CH_4 + 2H_2O(g)$$

Typically, Ni-based catalysts are used and according to Specht, Sterner *et al.*, the reaction takes place in a fixed bed reactor at temperatures in between 250 and 500 °C at a pressure of *circa* 0.8 MPa. The produced process gas is not pure methane: it consists roughly of 87% CH_4, 6% CO_2 and 7% H_2. This SNG product could be fed in the existing natural gas pipeline and storage network in many industrialized countries. Doing so, additional energy storage capacities of the TW h dimensions could be reached.

It is also interesting to note, when discussing the Specht/Sterner proposal, that the current standard and most cost-effective hydrogen production process is based on natural gas steam reforming.[7] Also automotive fuel processors are producing hydrogen-rich reformate from hydrocarbons, either for propulsion or exhaust-gas after-treatment purposes.[7,17b]

3.1.3 Efficiency evaluation. Unfortunately, although the Sabatier reaction is highly exothermal, the conversion step from hydrogen to methane is not for free. The Sabatier chemical equation *inter alia* directly states – based on mass flows – that 8 kg of hydrogen (corresponding to an FCEV range of 800 km) would be needed to produce 16 kg of methane (equivalent to an ICE range of 350 km). Furthermore, also the energy usage has to be assessed, see Table 2 for detailed numbers considering various paths.

The basic idea behind the SNG concept to use the conversion of electric energy into chemical energy carriers and the utilization of the existing natural gas infrastructure for the integration of renewable energy into the energy system of an industrialized country is definitely worth considering. But by evaluating Table 2, it becomes clear that the gas storage should be carried out at the process step in the process chain which provides the greatest advantage in terms of conversion efficiency: the SNG process builds on top of the hydrogen production step and leads to a considerably higher technology complexity and higher investment cost. Energy-wise, 100 GW h of electrical energy translates on average *via* the Sabatier process to 58 GW h of chemical energy in form of SNG compared to 71 GW h in form of hydrogen. Obviously, the storage and direct use of renewable hydrogen thus have to be preferred from an energetic point of view,[4a] especially since it is also possible to feed hydrogen directly into the existing natural gas grid up to a concentration of 5%.

Unfortunately, the SNG picture gets even less attractive when vehicle applications are considered. Due to the comparatively low efficiency of the internal combustion engine (also valid for CNG engines) and the occurring well-to-wheels losses, only about 10% of the primary wind energy would be available at the wheels of a natural gas vehicle.[4a,4b] If the process would be stopped at the hydrogen stage and a fuel cell electric vehicle (FCEV

Table 2 Large-scale energy storage: efficiency of various conversion paths.[17a] SNG: Synthetic Natural Gas

Path	Efficiency	Boundary condition
Electricity-to-gas		
Electricity → hydrogen	57–73%	Compression up to 8 MPa (pipeline pressure)
Electricity → methane (SNG)	50–64%	Compression up to 8 MPa (pipeline pressure)
Electricity → hydrogen	64–77%	Without compression
Electricity → methane (SNG)	51–65%	Without compression
Electricity-to-gas-to-electricity		
Electricity → hydrogen → electricity	34–44%	8 MPa compression, 60% re-conversion efficiency
Electricity → SNG → electricity	30–38%	8 MPa compression, 60% re-conversion efficiency

efficiency about 2.5 times greater than for ICE vehicles[2,18a]) would be considered, about ∼30% (ref. 4*a* and 4*b*) of the primary energy could be used for transportation purposes at the wheels. In addition the direct usage of the hydrogen has obviously also the advantage of being locally emission-free. In contrast, in the case of a SNG-powered CNG vehicle, NOx and CO_2 would be locally emitted.

It is therefore highly favourable, especially for the transportation application, to keep the number of the energy conversion steps as low as possible.[4] This goal can be realized by using batteries as electric and hydrogen as chemical energy carrier, and their utilization in various types of electric vehicle concepts.

Ref. 4*b* is the industry's gold standard regarding well-to-wheels CO_2 emissions and energy consumption. At this point it has definitely to be concluded that for any kind of alternative powertrain the source of energy and the related indirect emissions are of extreme importance. These issues are, *e.g.*, discussed for electric vehicles in ref. 17*c*. For a detailed discussion of the advantages and disadvantages of various alternative powertrains both on a technology and also on a total-cost-of-ownership level, ref. 4*c* and 25 are recommended. An interesting study on which future lithium electrode chemistry (*i.e.*, through their materials properties and improvement potential) will potentially enable a successful widespread market introduction for electric vehicles is to be found as ref. 18*b*.

3.2 Commercialization efforts

3.2.1 Demonstration projects. The roll-out of hydrogen fuel cell vehicles will not only change technology and value chains in the automotive sector, but also business models for the involved energy and gas companies. Hydrogen producers (*e.g.* Air Liquide, Air Products, Praxair and Linde) will become an important stakeholder in the automotive energy chain and the control over automotive energy carriers could at least partially transition away from oil-producing companies like BP, Exxon, Shell and Total. Also the generation of renewable energy and green hydrogen by electric utilities or completely new companies will play an important role. A whole new kind of decentralized energy companies could emerge by transformation of existing traditional companies or as start-ups. For automotive companies, at first glance, the business model will stay the same, as the manufacturer and vendor of passenger vehicles. But some automotive companies could intend to become energy providers themselves and extend their business model towards full-service energy and mobility solutions.

Large-scale demonstration projects are an intermediate step to test the elements of the new energy chain on a manageable level before up-scaling to an area-wide commercial deployment starts. Demonstration projects are thus being performed on all links of the value chain, *i.e.* production, storage, distribution and retail of hydrogen, as well as vehicle development, manufacturing and operation.[1]

For production and storage of hydrogen, several pioneer infrastructure projects in Germany are currently testing the conversion of wind energy into hydrogen. The efforts by German wind farm operator Enertrag with projects in Mecklenburg-Vorpommern and at the new Berlin-Brandenburg International Airport are a notable example. Additionally, in December 2011, the consortium "performing energy – Bündnis für Wind-Wasserstoff" was created to commercialize the wind-hydrogen technology. This organization consists of several academic partners, three German states but also major industrial companies such as Siemens, Linde, Enertrag, Total, and Vattenfall. The consortium intends to establish also a strong link to the transportation sector by providing an interface to the German "Clean Energy Partnership" hydrogen vehicle demonstration program.

Currently, vehicle manufacturers are in a pre-commercial phase of technology demonstration[18a] – a phase which does not exist in the engineering development process of conventional cars and powertrains. One prominent example is GM's "Project Driveway", for which the above-described HydroGen4 variant of the Chevrolet Equinox was developed and type-approved according to U.S. Federal Motor Vehicle Safety Standards (FMVSS). Out of about 170 vehicles that were produced in 2007 and 2008, 119 were handed over to private and commercial customers. As of mid-2012, that fleet has now accumulated over 4 million km of real-world experience, with three cars counting each well over 110 000 km. One vehicle already reached a stack operating time of more than 1600 hours. This project on public roads included over 10 000 drivers (non-GM persons), and more than 24 000 re-fuelings, dispensing about 71 500 kg of hydrogen. As of mid-2012, "Project Driveway" comprises already four full winters in the US (New York, Michigan, California) and Germany: more than 18 500 successful freeze starts due to cold weather conditions had to be performed by the fleet. The GM and Opel fleet operation included participation in national demonstration projects in the US, Germany, Korea and Japan, and as such was partly funded by national authorities.[19] Originally, the HydroGen4 powertrain was just designed for an operating lifetime of 2.5 years or 50 000 km. When comparing these numbers to the project results, it becomes clear that the corresponding expectations were exceeded significantly.

Germany's large-scale demonstration project for hydrogen as automotive fuel is organized by the "Clean Energy Partnership" (CEP), a public–private partnership

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which comprises 15 companies from the automotive and energy sector, as well as the oil and gas industry. Starting from Berlin as the first "Leuchtturm" or "bellwether" demonstration with four refueling stations, the infrastructure roll-out to other regions is executed over the course of the current phase III as well (see Fig. 16). Results for the velocity distribution for the Berlin HydroGen4 sub-fleet, underlining the urban driving conditions in large metropolitan areas, are given in Table 3. The results emphasize that full-load driving conditions, where FCEV technology would lose its efficiency advantage, are rather rare, even in the demanding German automotive environment. At least this holds true as long as large urban agglomerations like Berlin, Frankfurt, the Rhein-Ruhr area and Stuttgart are predominantly considered.

The aim of Germany's "National Innovation Program Hydrogen and Fuel Cell Technology" (NIP) is to prove the everyday suitability of hydrogen for transportation, and within the program duration, up to 50 stations can be expected to be built by 2015. On June 20, 2012, on the occasion of a dedicated Berlin government press conference, this strategic plan was personally confirmed by the German minister of transport, Peter Ramsauer, accompanied by high-ranking representatives of Daimler, Linde, Air Products, Air Liquide and Total.

Table 3 Velocity distribution of the HydroGen4 fleet for the Berlin sub-project of the German Clean Energy Partnership (CEP)

Velocity interval	Cumulative percentage
0–20 km h^{-1}	53%
21–40 km h^{-1}	17%
41–60 km h^{-1}	20%
61–80 km h^{-1}	4.5%
81–100 km h^{-1}	2.5%
101–120 km h^{-1}	1%
>120 km h^{-1}	0.5%

3.2.2 Hydrogen infrastructure and vehicle total-cost-of-ownership considerations. During the transition from technology demonstration projects to the phase where real customers are required to buy hydrogen vehicles, the obstacles to overcome are twofold:

- the cars are more costly than conventional cars

- the refueling infrastructure is less dense.

The higher vehicle cost might partly be compensated by accompanying measures, as are discussed for electric vehicles, including monetary as well as non-monetary incentives.

Several studies have been conducted on hydrogen supply for fuel cell vehicles (see ref. 20–25). Most studies[20,23,25] agree that fuel cost will be competitive with gasoline or diesel. The operation of a full-blown hydrogen retail business can be similar to today's gasoline or diesel retail system, and most likely hydrogen will be sold alongside conventional fuels, *i.e.* only a few dedicated hydrogen filling stations might emerge.

However, the studies also agree that under-utilization is the main obstacle for the commercial viability of a hydrogen infrastructure in the early phase. The biggest economic challenge is the capital cost and eventually the business model of the retail fuelling stations – they have to be set up for vehicle refueling as their single purpose, and will require a significant investment into refueling hardware. By contrast, other parts of the infrastructure, *i.e.* hydrogen production and distribution, are existing already today for serving other hydrogen applications, and, consequently, their extension to the car refueling supply business is profitable from the very beginning.[20] During the transition period, a substantial challenge will be the matching of scale and timing of retail infrastructure investment with actual hydrogen demand. A balance needs to be achieved between the minimum investment to meet initial demand *versus* the value of abundant fueling station availability in support of FCEV sales growth.

Fig. 16 CEP regions in current phase III (2011–2013), with more than 10 fueling stations and over 100 fuel cell vehicles.

Fig. 17 is given for general illustration of the so-called retail station dilemma, the graph is based on modeling by Shell and GM for the Los Angeles area.[20] The average utilization of the station is of high importance for the actual hydrogen cost. Exact numbers are strongly dependent on the site-specific setup, such as capacity of the station, footprint of the hydrogen installation, *i.e.* number of pumps and hydrogen storage technology, and whether the hydrogen is produced on-site.

Reasonable cost for hydrogen as automotive fuel can only be achieved with highly utilized stations, and thus small

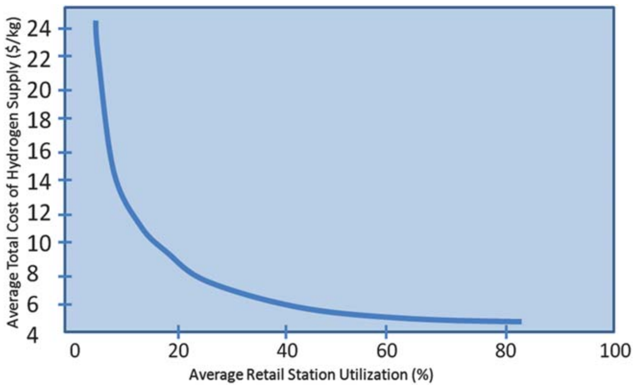

Fig. 17 Effect of station utilization on hydrogen cost (illustrative, based on modeling by Shell and GM for the Los Angeles area).[20]

stations which serve only a few cars (200 or even lower) are needed to minimize the effect of under-utilization in the beginning. Competitive fuel cost, however, can only be achieved with larger stations, which eventually replace the small stations which have been set up during the infrastructure build-up phase. A careful planning and coordination of a roll-out is vital, as even initially a minimum coverage has to be ensured and thus more refueling stations are needed than could be justified by vehicle numbers and market forces.

A full economic discussion of these points is out of the scope of this paper, but beyond the brief introduction in ref. 20, ref. 25 is highly recommended to those readers interested in detailed fuel cost projections and total-cost-of-ownership assessments. Also the comparative study of Contestabile *et al.* provides an interesting insight.[4c]

Planning of fuelling stations needs to go along with forecast of vehicle numbers and regions of their deployment. Geographic concentration and coordinated vehicle/infrastructure rollout will be part of the solution, and the respective governments will play a crucial role. In Germany, the H2Mobility coalition is a joint industry and government driven approach to achieve this alignment.[18a]

For planning a network of fuelling stations, studies on customer needs have been carried out. Results suggest that a surprisingly low number of stations are necessary for an initial roll-out. In an early phase, a few "lighthouse" urban regions with a high population, and therefore vehicle density,(see Fig. 17 and 18) need to be served with a sufficiently dense network. Outside of these regions only a comparatively low number of refueling stations along the major motorway network is required to ensure cross-country mobility between the large urban agglomerations. With this approach, considering Germany again, as few as only 140–220 refueling stations would be sufficient to roll out the first 100 000 fuel cell cars.[23] Complete area coverage could be achieved with as few as 1000 hydrogen stations, which would be able to serve the first million FCEVs. A similar study on the European level is given in ref. 24.

Attractive are the areas of high population and vehicle density, in order to get sufficient utilization of the fueling stations as early as possible. Fig. 18 shows how the car parc is distributed over Europe. A study that is discussing and assessing a staggered roll-out for Europe is recommended as ref. 25 and was financed by the H2mobility consortium comprising car, energy, and engineering companies.[18a] As a first step, H2Mobility focuses on Germany to set up the required fuel stations by a collaboration or "joint entity" of those industrial players and government institutions interested in a successful hydrogen infrastructure.[18a] Sister initiatives in other European countries are currently under discussion, or are already operational such as in the UK.

A major outcome of all studies mentioned above was that for a major European country such as Germany, France or the UK about 200 initial stations are needed and that the required overall early investment is significant but limited to about 200 to 300 million Euros per country over the course of several years. Since these stations do not provide a return on investment due to the so-called "retail station dilemma", government support for the companies setting up such a structure is needed.

For the end customer, according to ref. 25, the total-cost-of-ownership values (TCO) during the early stages (2010–2015) will be quite high compared to conventional ICE technology: 0.9€ per km compared to 0.2€ per km. This gap is mainly caused by initially high hydrogen prices (compare station utilization discussion above) and high vehicle cost for advanced propulsion technology at comparatively low production volumes. When switching to large and fully utilized stations and the implementation of real high-volume vehicle production, this picture changes substantially. The cost impact of automotive mass manufacturing is, *e.g.*, explained in Section 5 and Fig. 11 of ref. 1.

Based on the industry survey of ref. 25, this transition is expected to take place by 2020 to 2025: TCO values of 0.3 to 0.25€ per km would be reached. The TCO of conventional ICE vehicles is projected to increase through higher fuel prices and the cost of advanced powertrain and exhaust gas after-treatment technologies in order to comply with future stricter vehicle emission and fuel economy rules, respectively, CO_2 requirements. Therefore, fuel cell technology will by then be fully competitive. But such a change will not happen overnight. By contrast, it rather will be a continuous process over several years. Since for the end customer, during the early phases, the TCO values are not yet competitive, time-limited government support for vehicle purchases and non-monetary incentives for the car owner are essential to reach smoothly the required economies of scale regarding vehicle manufacturing.

4 Conclusions

Virtually all car manufacturers intend to reduce the usage of fossil fuels like gasoline and diesel as energy sources for automotive applications, especially by a continuously increasing electrification of the powertrain. Unfortunately, the current and future automotive battery energy density provides limitations for the development of pure battery electrical vehicles as soon as reliable vehicle ranges significantly greater than 150 km are required.

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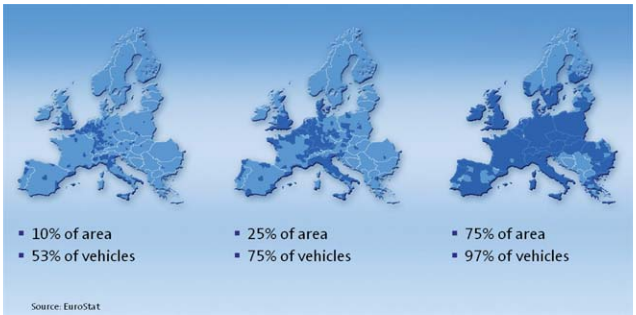

Fig. 18 Hydrogen infrastructure build-up and geographic concentration of the car parc in Europe (Eurostat[25]).

Therefore, GM and Opel pursue the concept of the extended-range electric vehicle (E-REV) and the fuel cell electric vehicle (FCEV). Extended-range (E-REV) and battery-electric vehicles (BEVs) both provide opportunities for load leveling through smart charging. This makes them a complementary technology to solar and wind power generation. In the longer term, however, load leveling and large-scale energy storage as chemical energy in the form of hydrogen offers a by far greater potential. At the same time, when used for propulsion, hydrogen enables the usage of an electrical drivetrain in combination with the high energy density of a chemical energy carrier. Today, fuel cell electric propulsion systems are applicable to all vehicle classes, but their continuous power and their hydrogen storage requirements have to be balanced against the vehicle package and the requirements of the thermal system (*e.g.* radiator size). Nevertheless, nearly all automotive companies agree that the FCEV is the only advanced propulsion option that provides long-range zero-emission driving combined with reasonably short refueling times of 3 to 5 minutes. The major technological challenges on the vehicle side have been overcome since the publication of the 2007 review on the status of fuel cell vehicles.[1] GM's "Project Driveway" and the "Technology-Demonstration-Fleet" proved that automotive fuel cell technology has reached a technology maturity, durability, and cost level which allows it to enter the early commercialization phase as defined in ref. 1. 70 MPa compressed gaseous hydrogen is the storage technology of choice and promoted by virtually all manufacturers of fuel cell vehicles. Only the necessary build-up of a fuel infrastructure still remains a significant challenge in all relevant automotive markets. Establishing a fueling network for fuel cell electric vehicles requires a joint approach by all the major stakeholders (*inter alia*, auto industry, energy companies and governments) and this process has to be accomplished in parallel to the vehicle rollout during the early commercialization phase (2015–2020 time frame). On the other hand, large infrastructure and energy supply investment are required for all future fuel options, including maintaining a "business-as-usual" approach based on gasoline and diesel fuel. The initial set-up of a hydrogen infrastructure for Germany would consist of about 200 stations and could be accomplished by a significant but definitely manageable investment of 200 to 300 million Euros over several years. Complete area coverage of a country like Germany could be reached with about 1000 stations. Ultimately, the degree of electrification and the displacement of fossil energy carriers are a function of energy prices, technology progress (regarding conventional, as well as alternative technologies), infrastructure availability, the regulatory framework, vehicle performance, and, finally, the vehicles' total cost of ownership for the end customer.

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