The role of fuel cells as key technology in the transition to clean energy for transportation

Today’s fossil fuel-based transportation contributes a considerable share to the overall emission of greenhouse gases. Fuel cells have emerged as energy-efficient and clean alternative to internal combustion engines. But although the dawn of the hydrogen age has been proposed for years, the search for cost-competitive fuel cell systems that compare favourably against state-of-the-art technology concerning performance and costs continues.

Current fuel cell technology is slowly approaching a stage of development where a wider introduction into vehicle power systems may be envisaged for the near future. The major issue that has now to be addressed is to develop different pathways for integrating fuel cells in on-board power systems. The choice of fuel has become one of the key questions, determining not only on-site emissions but also compatibility with state-of-the-art technology and fuel distribution systems. Whereas hydrogen is commonly seen as the fuel of choice with respect to fuel cell performance and zero emissions, other fuel options suggest themselves, especially during the transition from fossil fuels to clean energy sources. Different promising fuel options such as natural gas and biogas, and the liquid alcohols methanol and ethanol or exotic fuels like ammonia are reviewed and their potential utilization in fuel cells are investigated with respect to compatibility with on-board fuel cell systems, fuel production and distribution infrastructure, and emissions.

In this paper, a critical review of the possibilities and limitations of current and near-future fuel cell technology for transportation is given. A special emphasis is put on comparing different strategies for a wider introduction of this emerging technology.

Le rôle des piles à combustible en tant que technologie-clé pour la transition vers une énergie propre pour le transport

Le transport actuellement basé sur les combustibles fossiles contribue en grande partie à l'émission globale de gaz à effet de serre. Les piles à combustible, propres et disposant d'un haut rendement, se sont révélées comme une alternative au moteur à combustion interne. Cependant, bien que le début de l'ère de l'hydrogène soit annoncé depuis des années, on continue de rechercher des systèmes à pile à combustible compétitifs dans le domaine de la performance et des coûts par rapport aux technologies de pointe.

La technologie des piles à combustible s'approche graduellement d'un stade de développement qui permet d'envisager dans un proche avenir une introduction avancée dans les groupes électrogènes embarqués. Il s'agit maintenant de développer différentes perspectives pour intégrer les piles à combustibles dans les groupes électrogènes embarqués. Le choix du combustible déterminant non seulement les émissions sur place mais aussi la compatibilité avec les technologies de pointe et les systèmes distributeurs de combustible présente là une des question-clé. Alors que l'hydrogène est souvent considéré comme le combustible à choisir compte tenu de la performance de la pile et de l'absence d'émissions, d'autres combustibles s'imposent, surtout pendant la phase transitoire des combustibles fossiles vers des sources d'énergie propre. Différents combustibles prometteurs comme le gaz naturel et le biogaz, les alcools liquides méthanol et éthanol et d'autres combustibles moins conventionnels comme l'ammoniac et leur application potentielle dans les piles à combustibles sont étudiés en tenant compte de la compatibilité avec les systèmes à pile à combustible embarqués, de l'infrastructure de la production et de la distribution du combustible et des émissions.

Ce rapport présente des considérations critiques sur les possibilités et les limites, actuelles et futures, de la technologie des piles à combustible pour le transport. L'accent est mis sur la comparaison des différentes stratégies pour l'introduction avancée de cette technologie émergeante.

THE ROLE OF FUEL CELLS AS KEY TECHNOLOGY IN THE TRANSITION TO CLEAN ENERGY FOR TRANSPORTATION

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1 Introduction

The transportation sector is responsible for a significant share of the overall emissions of greenhouse gases and particles produced by the utilization of fossil fuels. The issue of transportation will become even more important in the future, due to the globalisation of economies and an accelerated development of developing countries. Hence, the only feasible way of reducing emissions produced by the transportation sector will be to introduce new technological approaches primarily with respect to the fuels utilized in vehicles, as well as the traction systems applied.

Fuel cells are commonly considered as very promising option in the transportation sector. Fuel cells offer a number of advantages compared to internal combustion engines that make them a prime candidate for utilization in transportation applications. Although the advantages of introducing fuel cells into vehicles are commonly undoubted, there are still a number of issues that have to be addressed before considering a wider application of fuel cell technology. One of the most important questions is the choice of fuel. Hydrogen is the fuel of choice with a fuel cell, but the lack of a ready hydrogen infrastructure as well as the question which primary energy source is to be utilized for producing the hydrogen, prevent a wider application of hydrogen in the near future. Other fuels are suggested, both as transition from a fossil based economy into a hydrogen based economy, and for ultimate use. Utilization of fossil fuels will have to be reduced within the near future due to the shortage of the remaining reserves, and due to the emissions produced.

Fuel cells will aid in reducing and ultimately moving away from the utilization of fossil fuels in transportation applications. This paper will first provide a brief overview of the current situation concerning current and projected utilization of fossil fuels, describing the reserves as well as the impact on the environment imposed by utilization. Then, fuel technology is investigated with respect to state-of-the-art performance, current areas of R&D and potential applications. In this, the most promising cell chemistries for mobile applications are investigated with a special emphasis on the PEM fuel cell, being the primary candidate for mobile applications. Finally, different fuel options are discussed and their possibilities and limitations in fuel cell based applications investigated.

2 Global emission scenarios / Scénarios des émissions globales

Approximately 85% of the world energy demand is derived from fossil fuels [1]. In the year 2001, some 362 EJ of fossil fuels were consumed (150 EJ crude oil, 78 EJ natural gas, 134 EJ coal). These figures have to be compared against safe resources of 5,987 EJ of crude oil, 4,922 EJ of natural gas and 28,852 EJ of coal. Additional resources - which will have to be proven in the future - are estimated as large as 3,709 EJ of oil, 6,280 EJ of natural gas and 195,000 EJ of coal. Thus, the reserves of fossil fuels will be consumed by 2041 for oil, 2063 for natural gas, and 2217 for coal, assuming that the aforementioned safe resources will be consumed at the current rates [2,3].

Utilization of fossil fuels has put a strong burden on our environment. Some 60 million tons of CO_2 (equivalent to 16 million tons of carbon) are released into the atmosphere each day [4,5]. The actual impact caused by these emissions is heavily discussed among experts. It is a fact that carbon dioxide that has been stored in oil, natural gas, and coal for millions of years is now released back into the atmosphere. An impact of this continuous release of additional carbon dioxide into the atmosphere on the climate seem very probable, although the actual rise in temperatures that can be traced back to human exploitation of fossil sources cannot be calculated accurately. This is due to the fact that future emissions are yet unknown, and the sensitivity of the climate with respect to variations in the atmospheric carbon dioxide content will have to be determined on a larger scale [6,7].

The Intergovernmental Panel on Climate Change (IPCC) has elaborated four so-called SRES scenarios (Second Report on Emission Scenarios). Each scenario calculates with a different economical, social and technological development of our society, causing a different attitude towards the utilization of resources and a different attitude towards environmental awareness. The results calculated with these scenarios are nevertheless very alarming. A strong increase in the concentration of carbon dioxide is predicted (150-280%), and even the scenario assuming the least impact for the world climate predicts a further rise in temperatures as high as 1.4℃.

The Austrian house of parliaments has decided to accept the Kyoto-protocol in 2002. Austria has thus committed to reduce greenhouse gas emissions by 13% compared to the emission of 1990 and to support renewable energy sources (wind energy, photovoltaics, biogas, small-scale hydro power) in order to achieve this ambitious goal [8].

3 Fuel cells / Piles à combustible

Heat engines utilized in mobile (e.g. internal combustion engines in cars) and stationary (e.g. gas turbine in power plants) applications convert the chemical energy supplied by the fuel into heat, and subsequently convert a fraction of this heat into mechanical energy utilizing pistons and crankshaft or turbines, for instance. This mechanical energy can then either be used directly for traction applications in a vehicle, or is converted into electricity utilizing a third conversion step.

The conversion step from chemical energy into heat is avoided in a fuel cell. Hence, the fuel is not burned but rather converted in a reversible electrochemical reaction either at ambient or at elevated temperatures, depending on the cell chemistry. Thus, the fuel cell can produce electrical energy directly out of the chemical energy of a fuel, only the fraction of energy that is not converted into electrical output power is converted into waste heat. This waste heat can be further utilized for domestic heating, supplying process heat to the industry or for driving a turbine cycle. Fuel cells have been invented as early as 1838 by Sir William Grove, but it was not until the 1950s that further-going research has lead to the first prototype fuel cells being produced and operated. The 1980s and 1990s have seen an increased interest in fuel cell technology, producing pre-commercial fuel cell modules operated in demonstration projects and field trials all over the world.

Fuel cells basically consist of two electrodes connected through an electrolyte and an external electrical circuit. Hydrogen – or a synthesis gas containing hydrogen - and oxygen or air are supplied to the two electrodes of the fuel cell where a catalytic reaction occurs, stripping the hydrogen molecules of electrons. The resulting positive ions move to the opposite electrode through an ion-conducting electrolyte (in an alkaline cell, negative ions move through the electrolyte), forcing the electrons to move through the external circuit. A number of different electrolytes are used, liquid electrolytes such as potassium hydroxide is commonly applied with an alkaline fuel cell, polymer membranes are utilized with PEM fuel cells, and ceramics are utilized in SOFC.

1.1 Fuel cell usage in mobile applications / Emploi des piles à combustible dans des applications mobiles

3.1.1 Fuel Cell Vehicles / Véhicules à pile à combustible

Fuel cells are a very promising option in replacing internal combustion engines and/or batteries in vehicle traction applications. In the last decades, a number of vehicles have been equipped with fuel cell units providing power to electric motors driving the vehicle. Most of these fuel cell cars can be referred to as more or less advanced proof-of-concept where the emphasis was not primarily on building a vehicle that is competitive for the commercial market. Nowadays, most major companies involved in the automotive industry can refer to their own fuel cell projects and fuel cell demonstration vehicles. Recent years have seen increased efforts in building vehicles that are able to match current internal combustion engine powered vehicles in performance, range and comfort. Advances in fuel cell as well as hydrogen storage technology have continuously lead to the development of smaller and lighter fuel cell power trains, almost matching the figures of current ICE power trains. The next step will thus be to move on to developing vehicles that do not only compare favourably with respect to performance data, but also with respect to the price. Making fuel cell vehicles commercially attractive will be a very challenging task. First steps of producing fuel cell vehicles on a smaller scale and utilizing these vehicles in fleet as well as individual passenger transport have already started, production on a larger scale is yet to come.

3.1.2 *Hybrid and mild hybrid fuel cell vehicles /* **Véhicules hybrides et semi-hybrides à pile à combustible**

Hybrid and mild hybrid vehicles are commonly referred to as an interesting option in vehicle propulsion, especially during the transition from conventional internal combustion engine drive trains towards the introduction of new and innovative propulsion concepts. The definition of a “hybrid” and a “mild hybrid” vehicle often cause some confusion. In both cases, the drive train consists of two sources of power such as a primary ICE and a secondary battery system in the case of current hybrid vehicles. In a "mild" hybrid, the battery pack (or any secondary power system) aids vehicle performance, but cannot drive the vehicle by itself.

Current mild hybrid car drive trains use an integrated starter/generator unit. When the vehicle stops, the main ICE shuts off. At acceleration, the system restarts instantly and the electric motor gives the vehicle a boost to take off. The electric motor can be used to power auxiliary devices such as the air-conditioner when the engine is off in some systems.

Another important feature implemented in some systems is the ability to use the generator unit for regenerative braking and therefore storing some of the vehicle’s kinetic energy in the on-board battery system.

With a primary fuel cell power system, the installation of a hybrid fuel cell/secondary battery hybrid concept suggests itself. In this, the battery pack could be used for start-up of the fuel cell unit, peak power supply and regenerative breaking. Vehicles could thus be quipped with smaller fuel cell units which is especially important with respect to the current costs of fuel cell modules. On-board fuel cells could also be operated without having to deliver strong variations of the requested power output immediately. Although fuel cells can follow a change in load very quick, it would still be favourable to operate early fuel cell units without these rapid changes in the output power.

3.1.3 Auxiliary Power Unit (APU) / Générateur auxiliaire d’électricité (APU)

Recent studies in the US revealed that heavy-duty truck engines idle about 20-40% of the overall time the engine is running [9]. The energy efficiency of an idling heavy-duty truck engine is very low (~3%) compared to the efficiency when operating on the highway (~40%) [10]. This causes additional emissions, additional wear and additional fuel and lubrication costs. It is therefore desirable to reduce or even avoid engine idling. This means that an independent APU is required [11].

Several technologies have been proposed for APU’s such as battery packs, direct fired heaters, adsorption coolers or APUs based on small diesel engines. From these technologies only the latter offers sufficient supply of both electrical power and heat. Interestingly, diesel engine APUs have had a rather limited acceptance in the trucking industry, because they are said to be heavy, expensive and noisy [12]. Solid oxide fuel cells (SOFC) are an interesting alternative to diesel engine APUs. They are expected to operate at high electrical efficiencies and low noise levels which means a considerable improvement of the driver’s comfort during rest periods. In addition SOFCs enable an easy utilization of hydrocarbon fuels which is essential for APUs. Finally, SOFCs are high temperature fuel cells which means that they provide useful heat which may be used to keep the truck’s engine block warm or to heat the driver’s cabin. Thus, SOFCs have received much attention as APU technology recently [13].

1.2 Polymer electrolyte fuel cells (PEMFC) / Pile à membrane polymère (PEMFC)

The simple construction and the good status of development have made PEM fuel cells the most attractive state-of-the-art fuel cell. A PEM fuel cell (Fig. 1) essentially consists of a membrane (approx. 50 μm), the electrode catalyst layers attached to each side of the membrane (approx. 15 μm), the gas diffusion layers (approx. 200 μm) and the bipolar plates, that have channels for distributing reactant gases over the cell area machined into the solid structure. The bipolar plate shoulders that are pressed onto the electrodes cover approximately half the geometrical electrode area.

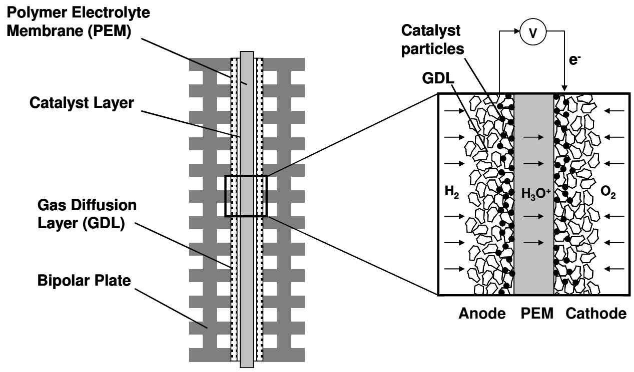

Fig. 1 Schematic of a PEMFC / Schéma d'une pile PEMFC

Polymer electrolyte fuel cells are developed primarily for mobile applications. They are able to generate high power output, operate at high efficiencies, and are very simple and robust due to their solid phase polymer membrane. Due to the fact that the electrolyte is solid and the operation temperature is approx. 60-80°C, sealing and cell assembly is less complex than for most of the other fuel cell systems. The low operation temperatures allows a fast start-up and a fast response to load changes. Platinum is used as primary catalyst, sometimes in conjunction with Ruthenium. The Pt loading has been reduced down to approx. 0,1 mg/cm^2 in state-of-the-art cells [14,15].

3.1.4 Performance

PEM fuel cell performance is primarily limited by the slow oxygen reduction kinetics, low conductivity of the polymer electrolyte, and membrane dehydration phenomena occurring at the interface between membrane and anode electrode. Fig. 2 gives an overview of state-of-the-art PEM single cell performance, showing the variation of the main sources of losses – the so-called overvoltages, referring to a loss in cell terminal voltage – with current densities up to 1 A/cm^2 [16]. In this, the cathode electrode overvoltage – shown in light orange – contributes the largest share to the total loss in cell output voltage. Anode electrode overvoltage is significantly smaller than with the cathode electrode, the membrane resistive overvoltage becomes a signification contribution only with larger current densities. The yellow area refers to the fraction of input chemical energy of the fuel (vs. HHV) that cannot to converted into electrical energy due to the thermodynamics of the overall cell reaction. Hence, the difference between this thermo-neutral voltage (approx. 1.48 V) and the reversible cell voltage (approx. 1.2 V) is lost for the conversion process and converted into waste heat.
The PEM fuel cell characteristics shown are for a cell operated with hydrogen and air at elevated pressures (5 bar cathode, 3 bar anode) and with an operating temperature of 80°C. PEM fuel cell stacks offering a similar single cell performance are commonly operated in automotive applications with peak stack output powers in the range of 50-70 kW.

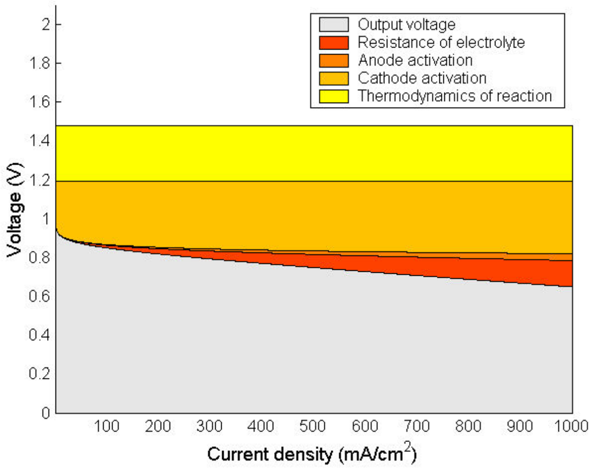

Fig. 2 Performance characteristics of a PEM fuel cell / Performances caractéristiques d'une pile PEMFC

3.1.5 Components R&D / R&D en composants

3.1.5.1 Electrolyte / électrolyte

The ion exchange membrane provides conductive paths for positively charged hydrogen ions to migrate through the membrane, while at the same time separating the reactant gases. The membrane is an electrical insulator to avoid short-circuiting of the electrodes. The conduction takes place via ionic groups located within the polymer structure. The actual ionic conductivity of the membrane phase is primarily determined by the local water content. Dehydration of the membrane causes a reduction of cell performance and may eventually even lead to anomalous cell degradation. Cell dehydration occurs primarily at the interface between anode electrode and membrane. Water molecules are dragged towards the cathode electrode via electro-osmosis. Hence, the anode input gas feed is commonly humidified to avoid membrane dehydration and allow for efficient operation of the fuel cell.

An often used material for the membrane is the perfluorosulfonic acid polymer Nafion® (DuPont Company), which consists of a backbone structure of Teflon® with perfluorinated side chains including sulfonic acid groups. The catalyst layer is in close contact with the membrane. It consists of catalyst particles supported on carbon structure and is in contact with the membrane phase.

The most important developments in recent years include the usage of ultra thin membranes, the development of composite membranes, the usage of carbon supported catalysts. However the resistance of the electrolyte is still strongly dependent on the humidity and the operation temperature of the electrolyte does not exceed 100°C. In automotive applications both points are very important. An operation temperature between 120-180°C is necessary to remove the heat from the stack with an adequate temperature difference to the cooling air efficiently. Due to the high dynamic load characteristic of a vehicle operation, the control system to keep a constant high humidity in the cell becomes very complex and it would turn out to be a great system simplification, if a new membrane was developed, which resistance is independent from the humidity of the reactants.

A number of investigations dealing with the development of high-temperature membranes are documented in literature. Popular approaches are to investigate organic/inorganic composite membranes, or polymer that absorb acid electrolytes. If polyvalent oxoacid is applied, the migration of protons within the membrane can occur with the Grotthus mechanism. Polybenzimidazol/ phosphoric acid membranes as well as the AB-PBI/phosphoric acid systems are the most popular approaches.

3.1.5.2 Catalyst layer / catalyseur

For oxygen reduction (and also for hydrogen oxidation) Platinum is used as catalyst. A multitude of Pt alloys have been developed, and it has been proven in phosphoric acid fuel cells that the alloys are more active and more stable than pure Platinum (Pt/Cr, Pt/Co, Pt/Ni). However, for applications in PEMFC, Platinum is still the catalysts of choice for oxygen reduction. A very interesting field has been opened by the worldwide research efforts in nano-ciences. The possibility of constructing nanostructured catalyst layers might be the appropriate tool to considerably reduce the catalyst loading in the future.

3.1.5.3 Gas diffusion layer

The porous backing layer or gas diffusion layer (GDL) acts as gas diffusor, provides mechanical support and as electrical pathway for electrons produced and/or consumed in the electrode reactions. The GDL is commonly based on carbon (carbon cloth, pressed carbon fibre, carbon felt) and incorporates a hydrophobic material such as polytetrafluoroethylene (PTFE) to create a non-wetting surface to prevent flooding of the pores.

3.1.5.4 Bipolar plates

The bipolar plates supply the electrodes with reactant gases, remove product gases as well as water and provide the electrical connection between adjacent cells of a stack. In addition, bipolar plates have to be gas tight, inexpensive, corrosion resistant in acid with oxygen heat and humidity, minimal in volume and weight and they have to be a good thermal conductor.

Several materials (graphite, carbon, carbon composites, steel, titan, aluminum, graphite-polymer composites) are investigated for the construction of bipolar plates. The cost should be low (<10$/kW US Department of Energy), the electric conductivity and the corrosion resistance high and the hydrogen permeability low. Basically steel would be suited as material for bipolar plate since it is non-porous and it can easily be machined. The disadvantages are the high specific weight and the low corrosion resistance against acidic electrolytes. In addition the surface becomes passivated with an electrically nonconductive layer. Recent developments have probably solved these issues and Japanese car manufacturers are going to use plates made of steel in the next generation of fuel cell cars. But as can be seen in the estimated costs of bipolar plates (see chapter 4), this "simple" part represents still a considerable cost factor.

1.3 Alkaline Fuel Cells (AFC) / Piles alcalines (AFC)

Alkaline fuel cells have been the best performer in the beginning decades of fuel cell development. In the last two decades a rapid change took place and many groups stopped developing alkaline fuel cell systems. From the few remaining groups ZeTek was the only one to produce small series of stacks and prototypes of hybrid vehicles. Also the prototypes demonstrated in practice, that the alkaline fuel cell is well developed, simple to operate and is inexpensive to manufacture, future investments in and usage of alkaline fuel cells are still uncertain. The main driving force for application of alkaline fuel cells for electric vehicles in future will be the cost issue,

Liquid potassium hydroxide is commonly used in a aqueous solution as electrolyte. The reaction product water and the waste heat generated are transported out of the cell with the liquid electrolyte. Alkaline fuel cell electrodes are built from an active layer and a hydrophobic layer, that are both sandwiched onto a metal grid for electronic conductivity. Catalyst loadings are in the order of 0,5 mg/cm^2, operation of the cell is usually at ambient pressures. Only few life cycle analysis have been conducted.

A number of catalyst materials have been investigated. Silver has been applied with the cathode electrode for oxygen reduction, Raney nickel has been applied with the anode electrode as well as Spinels and Perowskites.

A major advantage of the alkaline cell chemistry can be seen in the oxygen reduction kinetics being faster than with an acidic cell [19,17,18]. Thus, a Pt cathode (0,25 mg/cm^2) can operate at 0.868 Volts (RHE) with current of 100 mA/cm^2 in an alkaline solution (30 % KOH at 70°C). The same electrode produces only 0.63 Volta in a 96% phosphoric acid at 165°C. [19].

A major limitation for alkaline fuel cell operation in terrestrial applications is the sensitivity towards carbon dioxide within the electrode gas streams. The electrolyte will form carbonates when getting in contact with carbon dioxide. These carbonates can block up the porous electrode structures.

3.1.6 Performance

Alkaline fuel cells for terrestrial applications do not offer the high output power densities provided by rival cell technologies such as the PEMFC and the SOFC. Very high power densities have been demonstrated with alkaline fuel cells built for space applications, but the focus of research and development has clearly shifted from alkaline to PEM technology in the 1980s, so many promising approaches in designing cheap and efficient alkaline fuel cell systems have not been pursued any longer. Alkaline fuel cell stacks have been built and sold commercially by the Belgium company ZeTek, but they were built with outdated technology and thus do not compare well with the advanced PEMFC units.

Fig. 3 provides an overview of the losses occurring in an alkaline fuel cell utilizing reference data generated with one of the alkaline fuel cell stacks produced by ZeTek [20]. In this, the anode and cathode electrode overvoltage is derived from a fraction associated with the charge transfer overvoltage and a fraction associated with ohmic overvoltage for each electrode, and adding the voltage drop over the 1mm liquid electrolyte channel. Thus, the cathode electrode overvoltage contributes the major share of the reduction in cell voltage with the ZeTek stack. The fraction of chemical energy supplied to the cell that is lost due to the thermodynamics of the cell reaction is gain shown in yellow and is equal to the losses generated in a PEMFC operated at the same temperature.

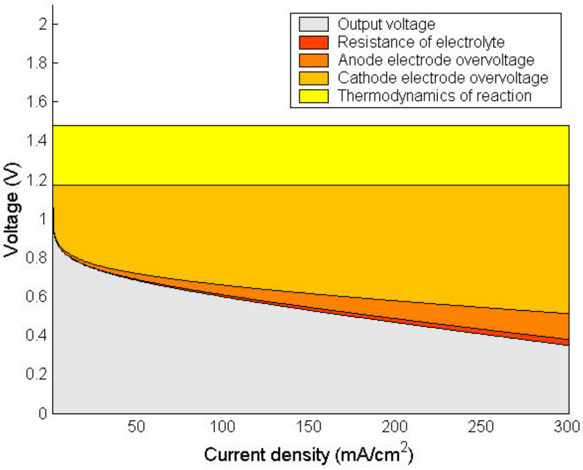

Fig. 3 Performance characteristics of an alkaline fuel cell / Performances caractéristiques d'une / Piles alcalines

3.1.7 Components R&D / R&D en composants

Potassium hydroxide (KOH) is commonly used as electrolyte. Other liquid electrolytes such as sodium hydroxide (solution of NaOH in water) offers some advantages compares to KOH with respect to costs, but the performance of the fuel cell system has shown to be reduced. Sodium hydroxide does offer the advantage of producing carbon dioxide as by-product. These small bubbles prevent the formation of crystals within the porous electrode structure and thus reduce the blocking of pores.

Several kinds of electrodes have been developed for AFCs. This includes different metal and carbon based electrodes [21,22]. Commonly gas diffusion electrodes are used. Inside the electrode the area for the reaction zone, where catalyst, carbon, the gaseous reactant and the liquid electrolyte is present has to be maximised. Currently the low power densities of AFCs are considered as a main disadvantage compared to other fuel cells.

For carbon dioxide removal from cathode-air soda lime is used. Soda lime is essentially a mixture of sodium hydroxide (NaOH) and calcium hydroxide $Ca(OH)_2$. Carbon dioxide is removed out of the air following the governing reactions written below:

$CO_2 + 2NaOH \rightarrow Na_2CO_3 + H_2O + \text{Heat}$
$Na_2CO_3 + Ca(OH)_2 \rightarrow 2NaOH + CaCO_3$

Some 0.6 kg of carbon dioxide can be removed with each kg of calcium hydride. Thus, some 1200 kg or 1000 Nm^3 of air can be cleaned (one kg of air contains roughly 0.5 g of carbon dioxide). Soda lime can not be regenerated, therefore this issue has not been solved satisfactorily yet.

Although the alkaline technology has been largely neglected in recent years (due to CO_2 poisoning issue), there are no obvious technical reasons to discount its potential for useful applications, especially considering the efforts towards a Hydrogen Economy which are made nowadays .

CO_2 in the reactant stream(s) has a distinct effect on the performance of AFC systems. The carbon dioxide poisoning is therefore a major issue for a future terrestrial application of AFCs [23]. Regenerative absorbers using molecular sieves could achieve the reduction of atmospheric carbon dioxide to acceptable levels. The requirement for dry air for these processes increases however both, their capital and operating costs. Another option uses liquid hydrogen to condense the carbon dioxide out of the air.

Another option is to use a solid ionomer alkaline membrane without free potassium cations that would enable a cell to run without the possibility of carbon dioxide poisoning. Or to operate the cell at higher temperatures, which increases the solubility of the K_2CO_3 in the electrolyte and prevent it from precipitation. Also the circulating of the electrolyte improves the AFC tolerance to carbon dioxide significantly.

Research has also to continue in the development of electrode materials to improve alkaline cell performance, with the goals to replace Platinum by nickel or silver and to improve the power density considerably.

3.1.7.1 System test at TU Graz / Université de Technologie de Graz système test

A test stand for alkaline fuel cells was developed and assembled at Graz University of Technology [24]. The control system was developed in co-operation with AVL List GmbH, Austria, the test cell was provided by ZeTek, UK, and the ammonia cracker was contributed by Apollo Energy Systems, USA. Fig. 4 shows the schematic diagram of the test bench. The test bench is subdivided into three main areas: the reformer with ammonia storage and initial fuel supply, the AFC with KOH storage and gas supply and the electrical load.

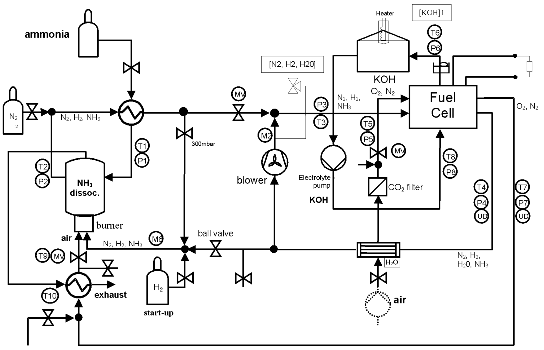

Fig. 4 Schematic Diagram of Test Bench / Schéma d'un banc d'essai

Hydrogen is used for the start-up of the ammonia dissociator. In a heat exchanger hot cracked gas containing hydrogen, nitrogen and traces of ammonia is cooled down to the working temperature of the AFC (anode in) and ammonia is preheated. A second heat exchanger heats up air for the burner using exhaust gas of the ammonia dissociator.
The CO_2 content in the air (cathode in) is reduced to below 10 ppm by using a soda-lime filter. Anode exhaust gas is partly recirculated and mixed with cracked ammonia gas and partly serves the burner of the ammonia dissociator as fuel. Cathode exhaust gas might be recirculated to the ammonia dissociator as well.

Due to the gas mixture the concentration of nitrogen increases in the anode loop as hydrogen is consumed. Therefore a much higher gas flow in the anode channel occurs than at systems with only hydrogen supply. The KOH storage is equipped with a heating device to keep the fuel cell at operating temperature. This is necessary because of the small output power of the AFC. At more powerful cells, a cooling system would be necessary. The electrolyte can be drained to an external reservoir when the fuel cell is shut off. It is absolutely necessary to keep the fuel cell filled with KOH during start-up and during operation to avoid damage of electrodes (if no electrolyte is in the chamber during operation hydrogen reacts directly with air on the electrode). The electrical load enables the dynamic real-time simulation of a down scaled vehicle in a driving cycle. No buffer battery was included in the set-up.

The performance of a fuel module and the overall efficiency of the complete system are shown in Fig. 5. The low-pressure AFC reaches a high overall efficiency due to less energy demand, particularly in compressing the air.

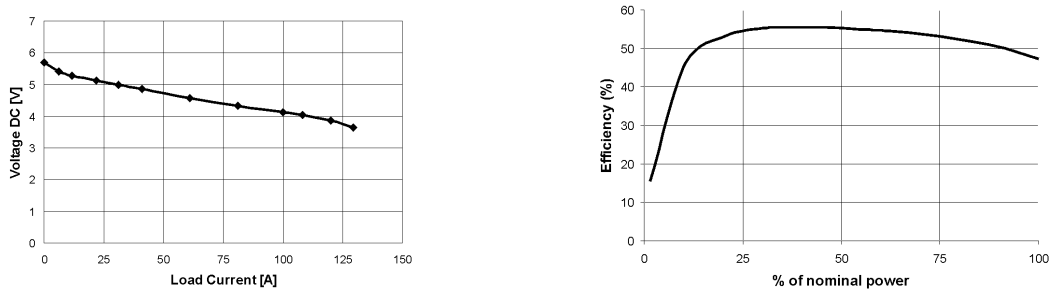

Fig. 5 Typical performance Mk2 module and efficiency of the complete Zevco fuel cell system [ZeTek, 1999] / Performance caractéristique d'un module Mk2 et rendement du système à pile à combustible Zevco

The principle of the control system is a HIL (Hardware in the Loop)-system as shown in Fig. 6. The grey block covers the hardware components: fuel supply, reformer, fuel cell stack, converter, battery, load, etc. This block is surrounded by control software: thermal management, vehicle management, etc. and also models of powertrain and vehicle are part of the software system. The dynamic electrical load for the fuel cell system is calculated with the simulated powertrain. In the labcar test stand the electrical load is directly connected to the fuel cell (without DC/DC converter) and acts as load only and not as power supply in case of recuperative braking.

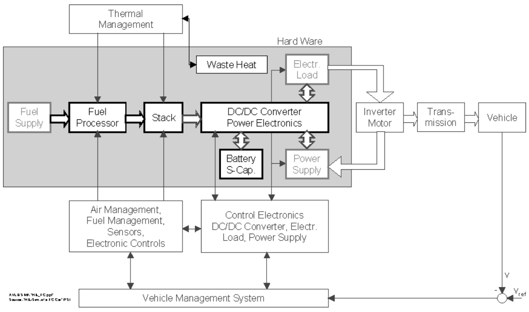

Fig. 6 HIL-System of fuel cell powered vehicle for labcar test stand (Graz University of Technology with the courtesy of AVL List GmbH) / Système HIL d'un véhicule à pile à combustible pour banc d'essai labcar (Université de Technologie de Graz avec l'aimable autorisation de AVL List GmbH)

The fuel cell stack shows a very good dynamic response in the New European Driving Cycle NEDC (under consideration of the manufacturer's restriction of a maximum current gradient of 20 As-1). Fig. 7 shows stack voltage, current and power within the NEDC (each diagram shows the NEDC profile in dashed line). The cracker shows a slow dynamic response, due to temperature drop under dynamic load. The layout of the heat exchanger systems and the gas channels have therefore to be further optimised [25]. The tests showed that cracked ammonia (nitrogen/hydrogen mixture) can be used in an alkaline fuel cell, and that the fuel cell is still able to respond to the NEDC [24].

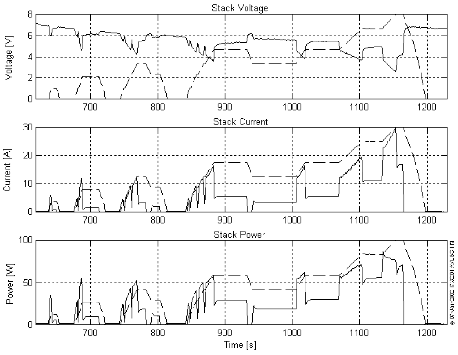

Fig. 7 Electrical Measurements of the Stack / Mesures électriques du module

1.4 Direct Methanol Fuel Cells (DMFC) / Piles directes à méthanol (DMFC)

Methanol is a readily available, liquid and comparably cheap fuel. The energy density of methanol is not substantially lower than with normal fuels. Especially the possibility to utilize liquid methanol fuel directly in a fuel cell is very advantageous with respect to mobile applications, where the fuel can be stored in an unpressurized and light vessel, and installing the required infrastructure is not too challenging.

The reversible cell voltage of a fuel cell operated directly on methanol is 1.21 Volts and is hence in the range of the reversible voltage of hydrogen (1.23 V). The major advantage of a direct-hydrogen fuel cell compared to a direct methanol fuel cell (DMFC) is the much faster anode reaction. In a hydrogen fuel cell, a two-electron reaction occur at the anode, converting one hydrogen molecule into two protons and two electrons. In a DMFC, the anode reaction produces six electrons and carbon dioxide as anode electrode reaction product. This reaction is inherently slower than the hydrogen reaction and also linked to producing intermediate products that can poison the catalyst material.

3.1.8 Performance

DMFCs are primarily operated in small-scale applications working at ambient pressures and near-ambient temperatures. The performance of a DMFC technology based fuel cell is significantly lower than that offered by a direct-hydrogen fuelled cell, although the same polymer electrolyte membrane is utilized and higher catalyst loadings are applied. The high gravimetric and volumetric energy storage density of a liquid fuel nevertheless makes DMFCs an attractive option for the portable power generation market.

Fig. 8 provides an overview of the losses occurring in a DMFC [26]. The cell is assumed to be operated at ambient pressures and an operating temperature of 80°C. DMFC performance is primarily limited due to the slow electrode kinetics, both on anode and cathode electrode. Performance of the cathode electrode is similar to that offered by state-of-the-art PEM fuel cells, but a large overvoltage is also encountered on the anode electrode, required for the electro-oxidation of methanol. Maximum output power densities achieved with the DMFC characteristics shown below are in the range of 100 mW/cm^2, which is considerably lower than the performance achieved with PEMFC technology, offering some 0.5-1 W/cm^2.

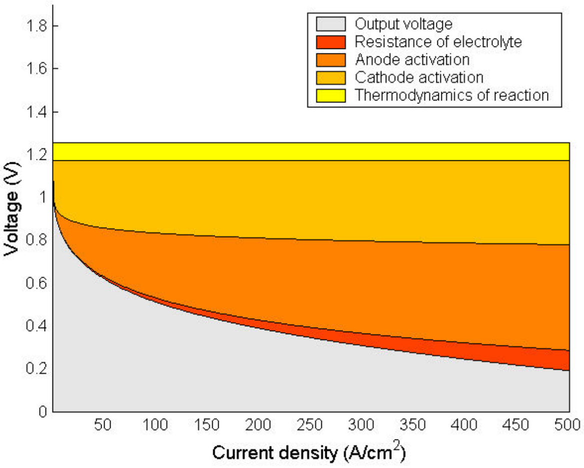

Fig. 8 Performance characteristics of a DMFC / Performances caractéristiques d'une pile DMFC

3.1.9 Components R&D / R&D en composants

Methanol can be supplied to the anode in liquid or gaseous state. A gaseous methanol feed increases the reaction kinetics at the cost of adding a mass penalty for auxiliary devices (e.g. evaporator, catalytic burner) and an increased system complexity. Due to the fact that DMFCs are primarily aiming at the portable power generation market where small units are operated in consumer electronics for instance, research and development is focused on liquid methanol systems. Thus, operation is limited by the boiling point of methanol, applying higher pressures would enable elevated operating temperatures but the overall efficiency would be reduced at the same time.

3.1.9.1 Anode catalyst

Platinum catalyses the electro-oxidation of methanol, but a couple of by-products produced within this electro-oxidation poisons the Platinum catalyst. Especially carbon dioxide – which is formed from methanol and water – poison and subsequently deactivate the active Platinum particles for the oxidation process.

The overall oxidation of methanol oxidation which is commonly referred to a six-electron-reaction due to the fact that six electrons are released for each molecule of methanol oxidised – is therefore only occurring very slowly at pure Platinum catalysts. A number of investigations on finding suitable options for avoiding catalyst poisoning and increasing the rate at which methanol is oxidised have been performed. Already as early as 1972, an addition of Ruthenium catalyst particles was suggested to avoid poisoning phenomena. Ruthenium is still used as the major catalyst material for methanol oxidation nowadays, utilizing in the order of 50 molar% Ruthenium for the best performance. The major efforts in achieving better catalyst performance are still focused on Platinum/Ruthenium mixtures, although some ternary (Pt/Ru/Os) and quaternary (Pt/Ru/Os/Ir) catalysts showed very good activities. Actual performance of different catalysts reported in literature is often very controversial [27,28].

3.1.9.2 Cathode catalyst

Catalysts applied with the cathode electrode of a DMFC are essentially very similar to the one used in a PEM fuel cell. Oxygen has to be reduced into water, accepting four electrons from the external circuit and four protons migrating through the membrane. Water is present in the cathode active layers both produced from the electrochemical reaction and coming from the membrane containing considerable amounts of liquid water in operation. Unlike with a PEM fuel cell, there is also a substantial amount of methanol coming from the anode electrode through the membrane, primarily driven by a gradient in concentration causing molecular diffusion as well as due to electro-osmotic drag pulling waster as well as methanol molecules along the gradient in electrical potential. This methanol coming into the cathode electrode active layer is converted into water and carbon dioxide, causing an additional consumption of oxygen, a loss in methanol fuel, and a reduction in the cathode electrode potential. This loss in electrode potential is especially disadvantageous due to the fact that the fuel cell output voltage is proportional to the fuel cell efficiency. Finding solutions avoiding or reducing methanol crossover through the membrane and conversion of methanol at the cathode electrode active sites is currently subject of research.

3.1.9.3 Electrolyte

Ionic conductivity of polymer electrolyte membranes currently utilized in PEM fuel cells depends on the amount of liquid water present in the membrane. Membranes show good ionic conductivities only if sufficient liquid water is present. Hence, a presence of liquid water is required and wanted. If methanol is present in the anode electrode of a DMFC, this methanol will also be dragged into the membrane, and the membrane will eventually be soaked with a mixture of water and methanol. Methanol and water will also be subject to similar transport phenomena concerning gradients in concentration, a hydraulic pressure gradient as well as electro-osmotic drag.

Utilizing membranes based on Polybenzimidazol (PBI) soaked with phosphoric acid can reduce the methanol crossover substantially [29,30]. Controlling the supply with fresh methanol as a function of output power required is another method of reducing methanol crossover [31]. The ideal solution would be to have a membrane that is selective towards the molecules present. Thus, different transport features could be achieved for water and methanol molecules, but the investigations are currently limited due to the similar behaviour of water and methanol [32,33,34].

Alkaline electrolytes show a better kinetic behaviour, but they continuously consume the electrolyte due to the formation of carbonates produced by carbon dioxide, one of the by-products of methanol oxidation. Polymer membranes conducting negative ions become chemically instable at temperatures above 60℃. Some aqueous carbonate electrolytes shoe a good theoretical behaviour [35].

3.1.9.4 Methanol-Crossover

Efficiency of a DMFC is limited due to the crossover of methanol molecules through the electrolyte. Methanol crossover causes a net transport of methanol that is not utilized in the anode electrode through the electrolyte and into the cathode compartment, reducing the potential of the cathode electrode and causing a loss in fuel. Methanol crossover is thus a very critical issue in DMFC operation.

Other than with the PEM fuel cell where crossover of fuel (hydrogen) is not connected to the migration of protons, these phenomena are linked within a DMFC.

In case of a DMFC operated in a vehicle, methanol crossover will be even more difficult to handle than with a small portable application. In transportation systems thin membranes are utilized to reduce the ohmic drop across the electrolyte and thus limit waste heat generation and loss in efficiency. If the thin Nafion 112 (ca. 50 µm) is utilized rather than Nafion 115 (ca. 125 µm), methanol crossover will increase by a factor of 2.5-3.5.[36]. Above that, an increase in operating temperature will also lead to an increase in methanol crossover [36,37].

Methanol crossover is essentially a function of the following parameters:

- operating conditions
- thickness of the membrane
- methanol concentrations
- structure of the electrode
- activity of the electrode

Methanol crossover through the membrane is equivalent to current densities in the order of 50 mA/cm^2 up to 500 mA/cm^2 (4 M methanol solution), depending on the concentration of methanol [38,39,40]. If the anode electrode is able to utilize methanol at the same rate as it is supplied, methanol crossover becomes very small and can actually be neglected [41,42]. Thus it is very advantageous if the methanol concentrations in the anode electrode active layer can be controlled as a function of the current drawn from the cell.

3.1.9.5 Innovative DMFC at TU Graz / Université de Technologie de Graz innovant DMFC

DMFCs, using (acidic and alkaline) liquid electrolyte without circulation, have been investigated in the past [43,44,45]. Though CO2 sensitivity was avoided, no solution was found to the problem of corrosion and performance reduction due to methanol crossover. We see the approach of a circulating electrolyte as an option compared to development of new membranes or methanol-tolerant catalysts whereby the circulating system includes the already discussed advantages. The cross leakage problem can principally be solved with circulating electrolyte. Other methods might reduce the efficiency, increase the resistance, endanger the catalysts or require low methanol mix percentages.

A new approach to solve the problem of methanol crossover in DMFC is investigated. It is based on the technology of circulating electrolytes developed for alkaline fuel cells. The aim is to set up a system to prove the advantages of circulating electrolyte in conjunction with DMFC and to optimise the design (electrolyte mass flow, arrangement of membrane or matrix) and minimise methanol crossover. The Direct Methanol Fuel Cells with Circulating Electrolyte (DMFC-CE) is capable to be shut down completely due to the removable electrolyte. Different separators and membranes has been tested to evaluate their possibility as methanol barrier. Topics of recent research work are:

- development of a new cell design which fulfils demands for testing of DMFC-CE;
- system development including a fully closed circulation loop for the electrolyte where the cross leaked methanol is separated;
- comparison of different electrolyte/membrane/electrode assemblies;
- investigate applicable anode and cathode catalysts;
- analysis of methanol cross-over effect during operation of different systems;
- modelling of the whole system to be able to analyse and optimise the system and its ancillary components.

A. System design

The electrolyte (1 M sulphuric acid) is pumped through the system by using a tubing pump. The electrolyte volume in the cell is exchanged approximately four times a minute. The experiments are carried out at temperatures between 45°C and 60°C. At the cathode pure oxygen at atmospheric pressure, at the anode a 2 molar methanol/water mixture, was used.
The middle part with the spacer in it consisted of three thin cemented PP-foils where the electrolyte canal was formed in the inner of these three foils.

The cell temperature is controlled by heating and cooling the liquid electrolyte and the water/methanol feed. This was done by a thermostat and flasks containing the both liquids and a thermostat controlled water circulation in an outer double-layer. To keep the concentration of the water/methanol solution and sulphuric acid constant it was necessary to have reflux condensers, where the water and methanol condenses back in the flasks.

B. Power increase compared to assembled PEM-DMFC

To evaluate the advantages using a liquid electrolyte assembly independent from the overall performance of the fuel cell it was necessary to design a PEM-DMFC made of the same components used in our liquid electrolyte cell. Compared to a DMFC with a PEM-membrane as electrolyte it could be shown that the assumed advantages of a liquid electrolyte system are given.

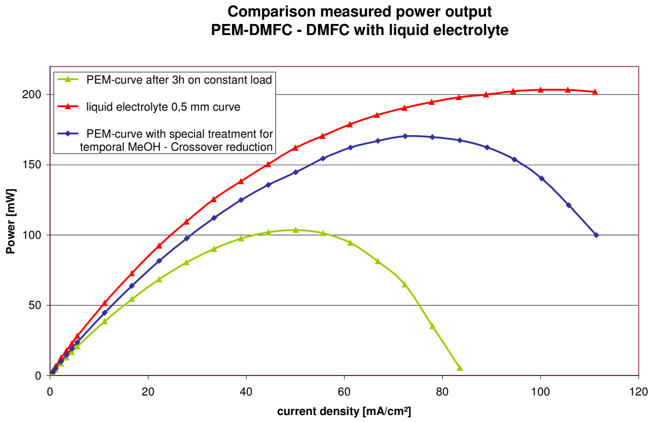

Fig. 9 Measured power output for PEM-DMFC and DMFC with liquid electrolyte / puissance de sortie PEM-DMFC et liquide électrolyte

In Fig. 9 the higher PEM-DMFC curve (middle) was obtained after the methanol feed was taken away and the fuel cell got spilled with deionised water until all the methanol was totally removed from the anode. After reattaching of the methanol feed, the measurement was done quickly, that methanol crossover through the membrane was reduced.

Comparing the curve derived from the measurement with the DMFC operated with an pumped liquid electrolyte to the power maximum of the PEM-DMFC in the case of the pretreated spilled fuel cell, the power improvement shown is significant.

C. Methanol crossover

To allow a certain methanol crossover from the cathode through the electrolyte canal, the electrolyte pump was stopped for 20 minutes. After that time a sample was taken from the electrolyte canal and the methanol level in the liquid electrolyte was measured using a headspace technology on a gas chromatograph.

The polarization curves were measured and the results are shown in Fig. 10 as the lower curves. The result of the oxidation of the diffused methanol at the cathode is a mixing potential, that decreases the cathodic potential, which results in loss of cell voltage.

The measured methanol concentration in the electrolyte was determined during this measurement by gas chromatography and was at the level of 0,8vol%.

In long term investigations using a constant load, the cell voltage and power drop depending on the slowly rising methanol level in the circulating pumped electrolyte was found and verified, because the methanol was not removed from the electrolyte.

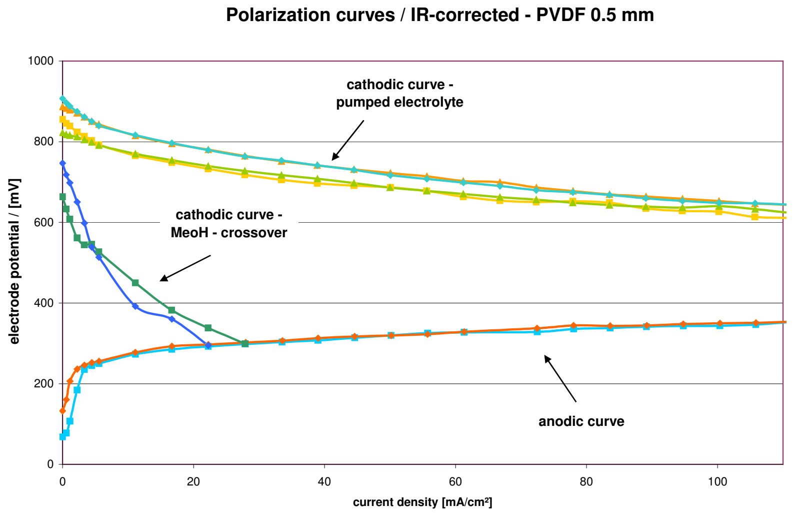

Fig. 10 Performance curves of DMFC cathode and anode with liquid electrolyte / performance de DMFC cathode et anode avec liquide électrolyte

Further following parameters of different polymer-electrolyte-membranes were obtained in the laboratory to complete the model for the methanol crossover in the PEMFC:

- water management in the membrane as function of temperature and humidity
- swelling characteristics and changes in the membrane dimensions as function of methanol concentration and temperature
- separated parameters of methanol crossover (diffusion, hydrodynamic permeability, electroosmosis) as function of methanol concentration, temperature, pressure and current desity
- investigation of the proton transport mechanisms
- electrochemical active surface area of a MEA
- porosity of gas diffusion electrodes.

1.5 Solid Oxide Fuel Cells (SOFC) / Piles à oxyde solides (SOFC)

An SOFC is completely built from solid elements. Thus, the problems associated with management of the electrolyte are avoided [46,47,48,49]. Due to the elevated operating temperatures in the range of 800°C, the cell open circuit voltage is in the order of 0.9 V. This reduction in the reversible potential is compensated due to the smaller losses encountered in operation. Efficiencies in excess of 50% can be achieved. Internal reforming of hydrocarbon fuels is possible due to the elevated operating temperatures.

Methane and other hydrocarbons are fed into the SOFC mixed with steam. These hydrocarbons are reformed into a hydrogen-rich synthesis gas which can be directly utilized within the anode compartment. Although a number of different gases are available for oxidation within the anode electrode compartment, investigations showed that primarily hydrogen is actually utilized in the electrochemical reaction [50]. Direct utilization of carbon monoxide is limited, but the water gas shift reaction causes a indirect conversion as more and more hydrogen is drawn out of the gas stream. Hence, a steam reforming catalyst must be applied in order to achieve the same performance of an electrode operated on synthesis gas and hydrogen. Applying too low steam to carbon ratios leads to carbon deposition, which can block up the fine pores of the electrode and reduce the fuel cell performance considerably. [51].

An SOFC is built from posour gas diffusion electrodes (Nickel-Cermet for the anode electrode, e.g. $LaMnO_3$ for the cathode electrode) and a solid electrolyte (yttrium-stabilized Zirconiumdioxid YSZ). Following the governing principles discussed with other types of fuel cell, hydrogen and oxygen are utilized in the electrode reactions, producing electrons that migrate through the external circuit.

SOFCs are primarily built for stationary applications. Small units are envisaged for CHP in residential applications, larger units are proposed for distributed power generation applications. Small SOFC units are also considered or mobile applications. Some R&D is currently performed aiming at the introduction of SOFC technology into small auxiliary power units (APU) for heavy duty trucks or even passenger vehicles.

3.1.10 Performance

The performance of SOFCs is closely linked to the operating temperature. High operating temperatures result in a good ionic conductivity of the electrolyte, fast reaction kinetics, and high output voltages. High operating temperatures are – on the other hand - very challenging with respect to balance of plant issues and system start-up for instance. Thus, research activities are currently focusing on developing SOFCs that operate satisfactorily with lower temperatures, in order to simplify balance of plant and operation, and still achieve a good cell performance.

Figure 11 shows the performance characteristics of an electrolyte-supported SOFC operated at 800°C and ambient pressures [52]. Due to the electrolyte-supported design assumed, the main source of losses is the ionic resistance of the electrolyte layer. The loss in cell terminal voltage due to the limited ionic conductivity of the electrolyte can be reduced by applying an anode-supported design where a thin electrolyte layer and a thick anode is applied. This leads to a better performance especially at lower current densities, gas phase transport of reactants into the catalytically active layer is reduced due to the thick diffusion layer, though.

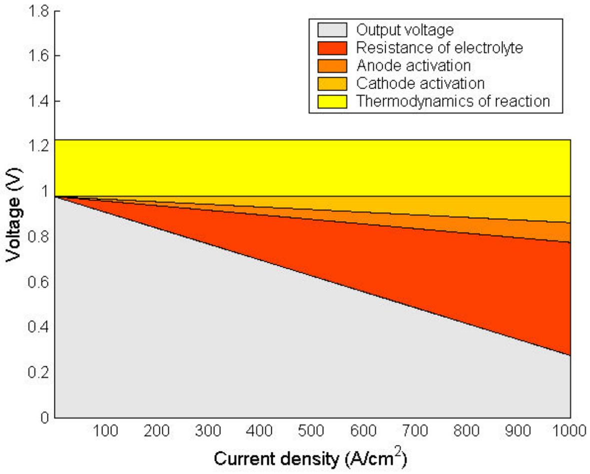

Fig. 11 Performance characteristics of an electrolyte-supported SOFC fuel cell / Performances caractéristiques d'une pile SOFC à électrolyte supporté

*3.1.11 Components R&*D / R&D en composants

R&D is currently focused on developing low-temperature electrolytes providing sufficient ionic conductivity for applications at temperatures below 800°C. Development of interconnector materials, flexible and highly-conductive ceramics and the production of large coatings for SOFC applications.

3.1.11.1 Interconnector

In an SOFC, the interconnector separates the individual cells of the stack, provides an electrical connection of the single cells and thus works the cells in series. Interconnectors have to be gas-proof, have a sufficient mechanical stability and the coefficient of thermal expansion must be similar to the coefficient of the electrolyte (in case of an electrolyte-supported cell) or of the anode (in case of an anode-supported cell) to minimize the mechanical stress developed when the cell is heated from ambient up to operating temperatures and back. A good interconnector material has also got to have a very good electrical conductivity while offering very low ionic conductivities.

3.1.11.2 Anode

Finding good materials for anode electrode of SOFCs is still subject of research. Work is currently focused on nickel-based materials. Nickel is utilized as catalyst material for reforming of hydrocarbons und can also be applied as catalyst for oxidation of hydrogen and carbon monoxide. The different thermal expansion coefficients of nickel and the electrolyte materials causes considerable problems. Thus, YSZ particles are mixed with anode powder to overcome the problems associated with thermal expansion. The coefficient of heat expansion is 16 x 10^{-6} 1/K for nickel, and YSZ offers values in the order of 10 x 10^{-6} 1/K.

3.1.11.3 Cathode

Perovskites or strontium-doped lanthanum manganite are usually utilized as cathode electrode material. Utilizing noble metals as electrode materials is not pursued due to the high costs associated. Currently, SOFC cathodes are primarily produced from Perovskites [53].

An increase in cell performance can be achieved by introducing catalyst particles into the interface between electrode and electrolyte. Palladium considerably increases the current densities for oxygen reduction for instance, a factor of 2-4 is achieved with Platinum. Electrodes with Ruthenium and Iridium do not show an increase compared to the untreated electrode.

3.1.11.4 Electrolyte

Only few of the ceramics conducting oxygen ions (O^{2-}) or hydrogen ions (H^+) can be used as electrolyte material in an SOFC. An electrolyte must offer ionic conductivities in the order of 0.1 S/cm (electrolyte-supported design) or 0.01 S/cm (electrode-supported design) und a very small electric conductivity (an electric conductivity in the order of 1 % leads to a loss in output voltage of roughly 1 % and a reduction in current density in the order of 5%).

Reducing the operating temperatures is a very effective means of reducing the costs of an SOFC. If an operating temperature of 700℃ can be achieved, metals can be used as interconnector materials. A ceramic interconnector costs roughly 130 Euro/kW, the same interconnector built from metals would cost only 7.6 Euro/kW [54].

YSZ offers an ionic conductivity in the order of 0,1 S/cm art 1000℃, and 0,001 S/cm at 620℃ [55].

3.1.11.5 Investigations of Auxiliary Power Units at TU Graz

At TU Graz in co-operation with AVL List GmbH a SOFC-APU system has been designed under following assumptions [11]. An idling truck engine typically generates about 1300 W electrical power as pointed out in [1]. Therefore, an SOFC stack with a nominal stack power of 1650 W is considered. 350 W are required for the air compressor and the water pump. The reformer is chosen to be an exothermic reformer (auto-thermal reformer or catalytic partial oxidation reformer). It turned out that the temperature at which the synthesis gas leaves the reformer is high enough for an immediate feed through of synthesis gas to the SOFC anode. Compressed and pre-heated air is supplied at the cathode side. Off gases leaving the SOFC stack are mixed and the remaining fuel content is completely combusted in the catalytic burner. The enthalpy of the exhaust gas after the burner is subsequently used to pre-heat the cathodic air feed, to vaporize the water for the reforming process and to provide useful heat. The gas is then vented to the environment.

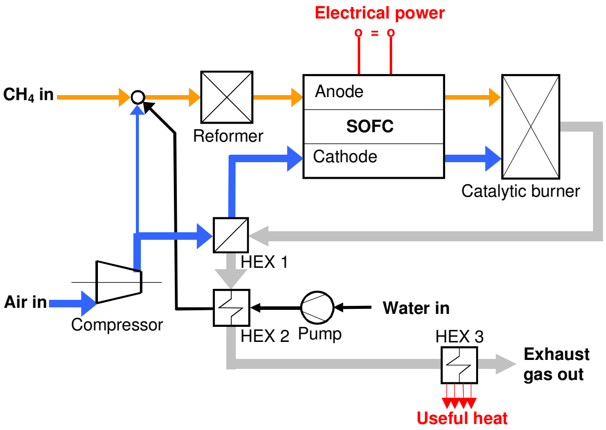

Fig. 12 SOFC APU layout[11] / Schéma d'une APU de technologie SOFC

The energy balance of the SOFC-APU is illustrated in Fig. 13 The lower heating value (LHV) of the methane input flow corresponds to 3.78 kW. 1.3 kW of this total energy input are converted into usable electrical power by the APU. 1.28 kW are useful heat if the exhaust gas is vented to the environment at 150℃. In this case the waste heat equals 1.20 kW if an ambient temperature of 25℃ is assumed. Based on above figures the electrical and total efficiency of the APU can be calculated to 34 % electrical efficiency and 68 % total efficiency. The electrical efficiency in case of engine idling is estimated to be about 3%, and a figure of 19% is given for diesel engine APUs. [1].

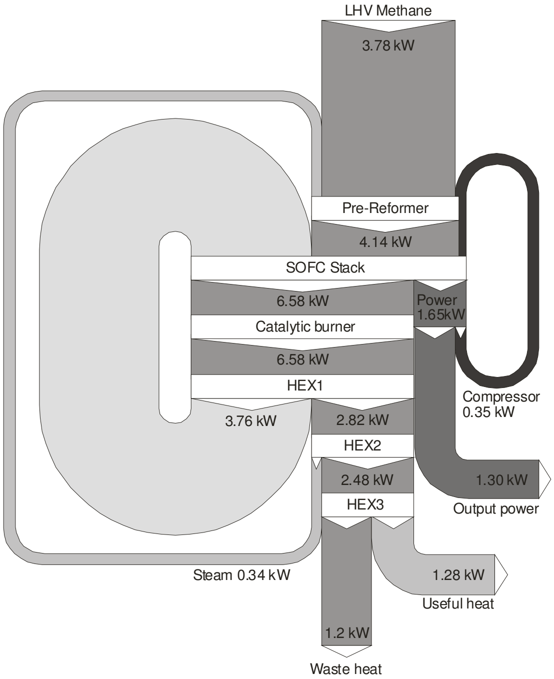

Fig. 13 Energy flow chart of the SOFC-APU (Sankey diagram) / Schéma d'énergie d'une APU de technologie SOFC

In both cases the electrical efficiency of the SOFC APU obtained from the thermodynamic analysis is significantly higher. It is interesting to note that 3.76 kW of heat must be recycled from the exhaust gas in order to bring the cathode air flow from 57°C to 650°C (HEX1). This means that the exhaust gas is cooled down from 846°C to 371°C. Designing heat exchangers that meet these requirements is a challenging and expensive task. An APU layout made up of individual system components as shown in figure 1 is suitable for principal system analysis as presented in this paper, or for feasibility studies on laboratory scale. In order to develop end-product solutions, the degree of system integration must be substantially increased to improve the thermal management of the APU and to reduce system complexity and costs.

4 Production cost and noble metal content (PEMFC) / Coût de production et teneur en métal précieux (PEMFC)

One of the key issues in fuel cell research and development will be to reduce the production costs. Tsuchiya et al. [56] calculated a number of different scenarios for the year 2020 and derived the costs of fuel cells will be reduced down to 15-145 €/kW due to mass production and increased cell performance. A moderate scenario based on 5 million fuel cell vehicles by the year 2020 estimates fuel cells costs to be 38 €/kW. In this, an increase in cell performance up to 4 kW/ m^2, as well as a reduction of platinum loading down to 1 g/m^2 and costs for stack assembly in the order of 94 € are assumed for a 50 kW fuel cell stack. Costs can be further broken up into 17 €/m^2 for the membrane, 49 €/m^2 for the electrodes, 57 €/m^2 for the bipolar plates, and 6 €/m^2 for auxiliary devices.

The safe worldwide reserves of Platinum Group Metals (PGM, primarily Pt, Pd and Rh) are assumed to be 100 million kilograms for the upper 1.2 km of the Earth crust. The majority of these reserves are in South Africa (>60%), Russia, Zimbabwe and the US. Platinum is currently utilized in the following applications: industry (310,000 kg), jewellery (62,000 kg) and catalysts for vehicles (93,000 kg). Despite of the safe reserves, here are still some concerns if sufficient PGM metals can be mined to satisfy the increased demand with a wide-scale introduction of fuel cell vehicles. The price for Rhodium has peaked at US$ 225/g due to a delivery bottleneck [57,58,59,60]

The following assumptions were made in order to estimate if sufficient amounts of Platinum are available for a wider introduction of fuel cells [61]. Assuming that less than 1% of the vehicles sold by 2010 will be powered by a fuel cell, some 500,000 fuel cell cars will be produced (50 million vehicles were built in 2000 some, the number of cars will increase to roughly 60 million by 2010). Assuming a noble metal content of 0,5 mg/cm^2 (anode and cathode) and a maximum power density of kW/m^2, one gram of Platinum will be consumed per kW of fuel cell stack peak output power. Due to advances achieved until 2010, the Platinum content will probably reduce down to 0.4 g(Pt)/kW by 2010 [61]. Thus, a 75 kWe stack will require 30 g of Platinum. This sums up to a total of 15,000 kg Platinum required for the 500,000 fuel cell vehicles assumed for the year 2010. Johnson Matthey estimates an additional 400 kg of Palladium, 200 kg Rhodium and 1100 kg of Ruthenium will be required.

Current vehicles require approximately 6-7g PGM metals [62]. Hence, substituting 500,000 internal combustion engine powered vehicles with fuel cell vehicles will thus also reduce the current PGM consumption by 3000 kg PGM metals a year.

Comparing the amount of Platinum required for an introduction of fuel cell vehicles and the amount of safe reserves of PGM metals shows that sufficient resources are available. Measures have to be taken now to increase the world Platinum production now in order to avoid a substantial increase in PGM metal prices with an increased utilization in fuel cell. Recycling of fuel cell catalyst materials will be the second major issue.

5 Fuel pathways for mobile applications / Combustibles envisagés dans les applications mobiles

1.6 Alkoholkraftstoffe / Alcools carburants

A number of different alcohols is utilized as fuel in vehicles, primarily Methanol (CH_3OH) and Ethanol (CH_3CH_2OH). These alcohols can be utilized within an internal combustion engine without requiring any major modifications. Thus, adding alcohols into normal fuels is commonly seen as quick and easy step into utilizing fuels derived from biogenous sources, as envisaged by the European Commission.

Methanol is currently produced primarily from carbon dioxide and hydrogen as well as carbon monoxide and hydrogen that are derived from coal gasification or from natural gas. Methanol production could also be based on biogenous sources, making this approach a very interesting option with respect to agricultural issues. Up to date, methanol is not produced from biogenous sources, yet.

The second very interesting option is to utilize Ethanol. Ethanol is already added into gasoline fuel in the US as well as in Europe. Some 5 billion liters of Ethanol are consumed per year in the US alone. This figure is approximately 1% of the total fuel required in transportation applications. Roughly 48,000 vehicles are operated on E-85 in the US (85 % Ethanol, 15 % gasoline).

Ethanol is also utilized as fuel in Brasil (40% of the vehicles are run on Ethanol), the remaining vehicles are operated on a mixture of Ethanol (22%) and gasoline (78%) [63].

Some 2 million cubic meters of Ethanol have been produced in the European Union in the year 2000.

Bioethanol is competitive against gasoline due to the exemption of petroleum tax granted. The price of crude oil would have to rise up to 60 US$/Barrel before Ethanol becomes a competitive option. With the tax exemption granted, Ethanol becomes some 20.3 Cents cheaper than gasoline with a crude oil price as low as 10 US$/Barrel.

1.7 Sodium borohydride / Borohydrure de sodium

Hydrogen is produced if a mixture of sodium borohydride ($NaBH_4$) and water is fed to a catalyst. The reaction product Natriummetaborat ($NaBO_2$) is equivalent to Borax, a substance commonly found in cleaning agents. Some 2.4 liters of hydrogen are produced b each gram of sodium borohydride.

1.8 Ammonia / Ammoniac

Ammonia is a colourless gas with a typical, penetrating odour. Ammonia is easily transportable and storable and can be liquefied with small pressures in the range of 800–900 kPa at ambient temperature. Ammonia is extremely soluble in water and organic substances such as Ethanol, Acetone and other hydrocarbons [64]. Dozens of chemical plants produce Ammonia primarily from natural gas in a simple process – the so-called Haber-Bosch process – primarily for applications in the agricultural field as fertilizer.

1.9 Hydrogen / Hydrogène

Hydrogen has all the qualities one could wish for a new energy vector (economical to produce, easy to transmit and store, renewable and non-polluting, and more efficient than current fuels), so it is not over-ambitious to foresee that it will play an important role in the international energy panorama within a few decades. The timeframe is, as with any major infrastructure and technology change, unclear. By 2050 up to 50 % of energy generation could come from renewable sources, with hydrogen a significant component in the fuel mix [65]. Worldwide initiatives like the High Level Group of the European Commission, the World Energy Network (WE-NET) project in Japan or the U.S. Department of Energy Hydrogen Research and Development programme aim to establish technologies for constructing a hydrogen energy network on non-carbonaceous renewable energies such as hydraulic, solar and wind energies.

The introduction of a hydrogen infrastructure for transport will also overcome the main impediment to expand fuel cell vehicle use – the non-existent of a fuelling infrastructure – and will in this way also increase the energy conversion efficiency at the end-user considerably. A wide range of technologies for hydrogen production are under development. This includes central, on-site and on-board production technologies of hydrogen from renewable and from fossil fuels in the short-term. In principle, all primary energy carriers could be utilized for hydrogen production (Fig. 14). In this, the utilization of hydro- solar- and wind power requires to produce electrical energy first, which is then converted into hydrogen.

Also when the vision of our future with hydrogen becomes clear – a transport sector based on renewable energies with no CO2 emissions in the whole production chain, the short tern scenario including the usage of fossil fuels for the transition period is very difficult to evaluate in terms of emissions and efficiencies.

Therefore several studies were performed to investigate the influence of different technologies for several fuel pathways (well to wheel) to estimate the emissions and to evaluate their potentials [66,67,68,69,70].

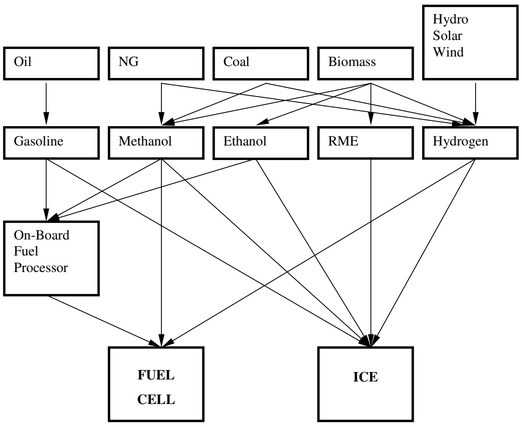

Fig. 14 Selected alternatives of fuel for transport applications / Combustibles alternatifs sélectionnés pour des applications mobiles

The current and future emission scenarios for different fuel pathways have been investigated by several study groups. To discuss the basic challenges of the introduction of a new fuel carrier, a short excerpt based on the study of GM for Europe 2010 is given here [68]. The data shown in Fig. 15, give an overview of the energy use and greenhouse gas emissions of 12 fuel pathways from well-to-tank. The WTT energy use is lowest for petroleum derived fuels, gasoline and diesel, which are slightly above 1 MJ/MJ. Below 1.5 MJ/MJ falls CMG (compressed methane gas obtained from anaerobic digestion of biogenic wastes). Between 1.5 and 2.0 MJ/MJ are methanol and several CGH_2 production pathways. In the range of 2.0 to 3.0 MJ/MJ fall LH_2 obtained from EU Mix NG, CGH_2 obtained from steam reformed biogas and ethanol from sugar beet. Ethanol obtained from poplar plantation is positioned at about 3.0 MJ/MJ. The most inefficient way shown, is CGH_2 produced in onsite electrolysis from EU electricity mix (the same for LH_2 would be even approx. 5.4 MJ/MJ).

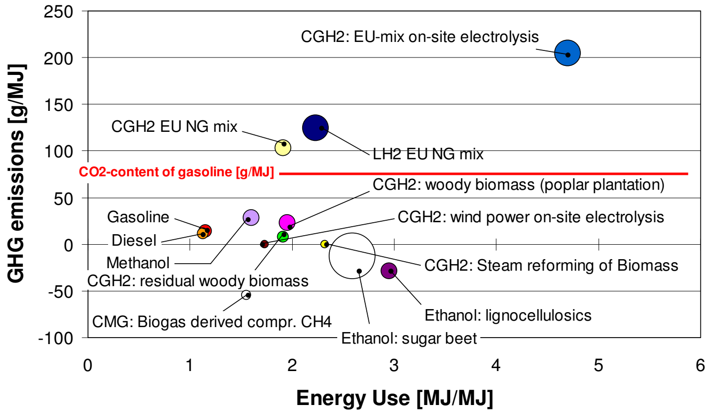

Fig. 15 Selected pathways of renewable and non-renewable energy: Well-to-tank (WTT) energy use [MJ/MJ] and WTT Greenhouse Gas (GHG) emissions; CGH_2 (Compressed Gaseous Hydrogen), LH_2 (Liquid Hydrogen), NG (Natural gas) [68] / possibilities renouvelable et non- renouvelable énergie

For GHG emissions, only the emissions during the supply are considered (as an example the CO_2 content of gasoline is given in the figure, to show the emissions of CO_2 during combustion). The best pathway with respect to GHG emission is ethanol from poplar plantation with about –30 g/MJ and ethanol with sugar beet with between about –40 g/MJ and +10 g/MJ. CGH_2 obtained from steam reforming of biogas or from onsite electrolysis using wind power have almost 0 g/MJ WTT GHG emissions. CGH_2 from residual woody biomass lies in the order of diesel from crude oil and CNG from EU NG Mix is comparable to gasoline from crude oil (15 g/MJ). Methanol causes approx. 25 g/MJ, significantly higher are LH_2 or onsite CGH_2 obtained from EU NG Mix, both above 100 g/MJ. CGH_2 obtained via onsite electrolysis from EU electricity mix has the highest WTT GHG emissions, about 200 g/MJ.

The results for WTT can be summarised in:

- Biomass derived fuel supply pathways show the highest complexity and the widest range of results,
- Fuel supply pathways from renewable energy sources lead to drastically reduced GHG emissions
- Hydrogen derived from renewable electricity represents a significant improvement with respect to GHG emissions.

The investigation of the Tank-to-wheel (TTW) emissions and energy efficiencies [68] prove the principal advantages of fuel cells compared to conventional energy conversion technologies. The efficiencies are considerably improved by the usage of fuel cells and the GHG emissions are drastically reduced due to the higher efficiencies and lower emissions.

6 Conclusions

Even in the B1-scenario of the International Institute for Applied Systems Analysis (IIASA), which is based on very optimistic assumptions regarding the future utilization of renewable energy sources, hydrogen will still be produced primarily from fossil energy sources in the year 2100. Electrical power, on the other hand, will be produced from renewable energy sources and nuclear power plants. Hence, the results derived from the scenario suggests to utilize renewable energy sources for power generation rather than for the complicated production of hydrogen fuel. Due to the fact that hydrogen will be produced primarily from natural gas in the short- and medium-term, a wider utilization of hydrogen has to be evaluated with respect to the efficiencies achieved with utilizing natural gas directly.

Methanol will also be produced primarily from fossil energy sources due to the limited agricultural areas that can be utilized for producing biogenous energy sources for methanol production. A wider utilization of methanol in transportation applications will lead to a small reduction of local greenhouse gas emissions. The conversion steps required for methanol production will - on the other hand - cause additional emissions, resulting in a net increase in emissions compared to operating vehicles directly on natural gas.

A wider application of fuel cells is advantageous due to the increased conversion efficiencies achieved with rival power generation technologies such as gas turbines for instance. Operating fuel cells in vehicles causes an overall reduction of greenhouse gas emissions only if the hydrogen fuel is either produced from regenerative energy sources – which seems a realistic approach only with small numbers of vehicles – or if a surplus of electrical energy derived from carbon dioxide free production (i.e. essentially nuclear power) is available.

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