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Fuel processors for fuel cell vehicles

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Abstract

Hydrogen is the fuel of choice for today's fuel cells with polymer electrolyte. However, for its successful application in vehicles there are some open issues, for example the missing hydrogen infrastructure and difficulties with regard to vehicle storage tanks. This makes on-board fuel processors together with liquid fuels both attractive and necessary. Different types of fuel processors for fuel cell vehicles are described. Some examples together with developmental challenges are presented. © 2002 Elsevier Science B.V. All rights reserved.

Keywords: Fuel cell vehicles; Fuel processors; Fuel infrastructure

1. Introduction

Today fuel cells with a polymer electrolyte are mostly regarded as the best solution for a vehicle with a fuel cell engine. For this type of fuel cells hydrogen is the fuel of choice. Of course there are other and very promising developments like direct methanol fuel cells or fuel cells with different electrolytes, but up to now they do not play a major role in the transportation sector. The two fundamentally different ways to supply the fuel cell with hydrogen are either to store hydrogen in a high-tech tank in the vehicle or to produce hydrogen on-board by processing a liquid fuel, which is stored in a much less sophisticated tank. The article will discuss some pros and cons of the hydrogen solution first and then describe several different types of fuel processors for fuel cell vehicles by presenting some examples. An outlook on some of the technical challenges, which still need to be addressed, is given at the end of the article.

2. Some remarks about the need of fuel processors in a hydrogen world

Why should fuel processors deserve consideration? It seems to be obvious to supply fuel cells directly with the fuel they are designed for. By doing this many successful demonstration vehicles have been built since a couple of years. Fuel processors are often described as bulky and heavy, requiring long start-up times and providing no dynamics. Also, the general consensus seems to be that fuel processors reduce vehicle efficiencies, all while increasing fuel production cost. The infrastructure requirements that accompany fuel processors seem to be much more critical than the requirements for using fuel cells and hydrogen, especially because the fuels for fuel processors are often regarded as transition fuels being on the market only for a limited period of time. Many people believe, that in some 20 years from now the hydrogen economy will be on the way and it will last for many decades.

2.1. Weight and volume

The issues around weight and volume can be discussed by looking at two well-known fuel cell vehicles, DaimlerChrysler's Necar 2 and 3 (Fig. 1). Both vehicles have a 50 kW fuel cell engine inside, but the first one uses gaseous hydrogen as fuel while the other one uses liquid methanol in conjunction with a fuel processor. Necar 2 appears to be a fully usable minivan with enough room for six passengers as well as some luggage. What has been left of the Mercedes A class in the Necar 3 is a two-seater with no trunk at all.

Taking a closer look at both vehicles, it becomes clear that Necar 2 is considerably larger than Necar 3. There is a difference in the basic design. In addition to this, Necar 2 has two hydrogen pressure vessels on its roof in a dedicated compartment, sporting a volume of more than 750 l. The fuel cell systems, disregarding the fuel storage and the fuel processor, are more or less the same for both vehicles. They are located either below the floor or under some of the seats. A direct comparison of the storage systems including the fuel processor shows that Necar 3 offers a slightly greater range than Necar 2 at a comparable volume!

Of course a storage tank for liquid hydrogen is much smaller, but it is possible to make a similar comparison of DaimlerChrysler's Necar 4 (liquid hydrogen) and Necar 5 (methanol fuel processor). The question of how to store

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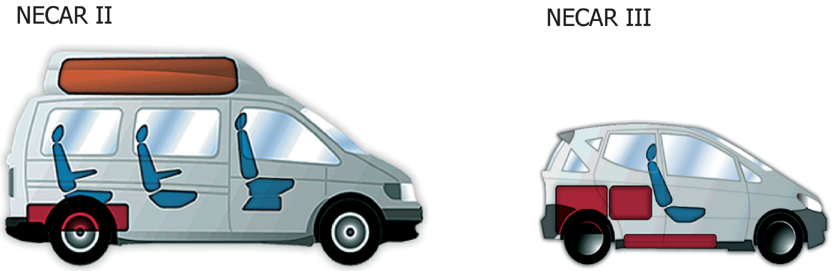

NECAR II			NECAR III		
Fuel Cell System	Power (2 Stacks)	50 kW	**Fuel Cell System**	Power (2Stacks)	50 kW
	Voltage Range	180 - 240 V		Voltage Range	185 - 280 V
Pressurized Tanks	Volume	2x140 l	**Tank**	Volume	38 l
	Pressure	250 bar			
Drive Train	Electric Drive		**Drive Train**	Electric Drive	
	cont.	33 kW		cont.	33 kW
		max 45 kW			max 45 kW
	Range	> 250 km		Range	> 300 km
Gross Vehicle Weight		2660 kg	**Gross Vehicle Weight**		1750 kg

Fig. 1. DaimlerChrysler's fuel cell vehicles Necar 2 and 3 [1,2].

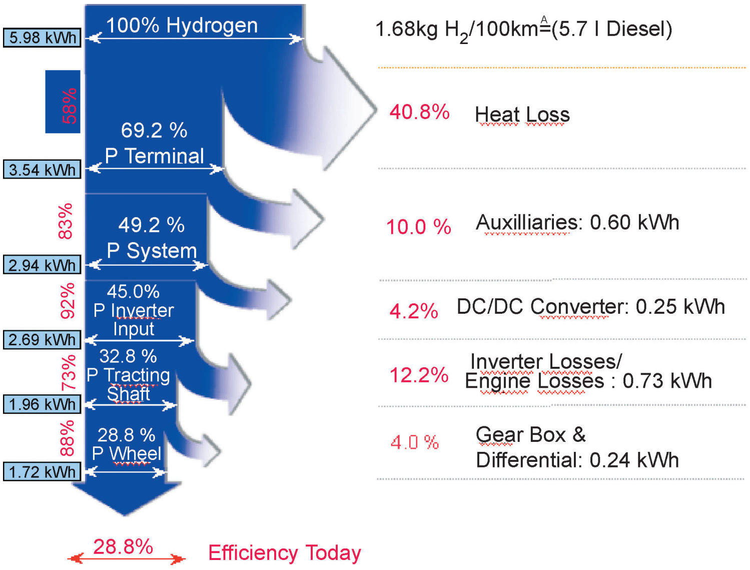

Fig. 2. Energy flow in the Necar 2 [3].

hydrogen in a vehicle still remains unanswered, and the demand for an acceptable range for fuel cell vehicles requires liquid fuels together with fuel processors. Only a new breakthrough technology would be a game changer in this situation.

2.2. Emissions

The hydrogen-fuelled vehicle is regarded as a "zero emission vehicle (ZEV)," while the methanol-fuelled vehicle "only" complies with the Californian "SULEV" standard.[1] The reason for this are very low hydrocarbon emissions of the fuel processor. CO_2 is not an issue in California. In a well-to-wheel scenario the result of this comparison might look different, especially with both fuels being produced from renewable resources (including CO_2!). In this case both fuel cell vehicles should perform very similar. Methanol is here only used as one example and the other liquid fuels can be discussed with equivalent results in a similar way.

2.3. Infrastructure cost

A comparison of the infrastructure cost shows that hydrogen fuel stations will cost at least 10 times as much as a fuel station for a new liquid fuel (e.g. methanol or ethanol). Naturally, in the case of today's already wide-spread used gasoline or diesel there would be no cost at all, which is almost also the case for a special new "fuel cell grade" gasoline.

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[1] SULEV: 1.0 g/mile CO, 0.11 g/mile NMHC, 0.02 g/mile No_x, 0.01 g/mile PM over the FTP 75 cycle (ZEV: all 0.0 g/mile).

Its cost would be in the same order of magnitude of those for a completely new fuel, maybe lower. The cost for introducing a completely new liquid fuel can also vary greatly depending on the comfort that the fuel station is offering to its customers, as can be seen at the broad variety of gasoline fuel stations today and in the past. If opting for hydrogen fuel stations, however, immense expenses simply cannot be avoided since these fuel stations require dedicated tanks and dispensers to handle the fuel safely while operating at high pressures or very low temperatures. It would only make sense to build up a hydrogen infrastructure if it was perfectly clear that it would last for many decades. This, however, is not clear today because of safety reasons and because no suitable hydrogen storage system for vehicles exists.

2.4. Availability

Taking a look at today's availability of the above mentioned fuels shows also that liquid fuels are closer to reality than hydrogen. Besides today's gasoline and diesel large production capacities especially for methanol and in increasing amounts for ethanol are already available that are sufficient for hundreds of thousands of fuel cells vehicles. This is not the case for hydrogen, where almost no overcapacities exist and the necessary production plants would have to be built up. On the other hand, new capacities for clean synthetic gasoline and diesel are already planned and will be installed in the next years. These fuels may be used in fuel cell vehicles, too.

Of course there are many more reasons to why fuel processors are beneficial and necessary for fuel cell vehicles but that would be outside the scope of this article. Instead, we will continue by taking a closer look at some of the technical issues surrounding using fuel processors in a vehicle, starting with considering the aspect of efficiency.

3. Thermal efficiencies of fuel processors in fuel cell vehicles

The efficiency of a fuel cell vehicle is determined by many parameters. Many of them have nothing to do with the fuel cell engine itself or even with a fuel processor. An analysis of the energy flow in Necar 2 shows that the efficiency of the fuel cell system of close to 50% is lowered to 28.8% due to the drive train and its individual components (Fig. 2). The inverter and the engine cause an efficiency loss of 27%. This is typical for all fuel cell-driven vehicles and of course in a similar way also valid for conventional ICE-driven vehicles. On the other hand, this offers a quite reliable way to define efficiency by dividing the energy brought to the road by the energy leaving the storage tank in the vehicle. One should not forget, however, the big influence of vehicle components such as inverters, gear boxes, or even tires.

A realistic discussion about the efficiency of a fuel cell engine needs to start with the interfaces to the vehicle. The definition that is used at Xcellsis is based upon interfaces to the fuel storage, the vehicle control unit, a 12 V battery, a DC delivery port for the traction motor inverter, the connections to the car radiator and to the outside air with an inlet, and an outlet to the exhaust pipe. Everything within these limits is defined as a fuel cell engine (Fig. 3). The Xcellsis efficiency values are based upon this.

Hence, a system with a fuel processor is much more complex, in particular when considering its many internal interfaces in addition. When discussing fuel processors, those interfaces become very important (Fig. 4). Because it is very easy to calculate a good efficiency for the fuel processor resulting in a bad efficiency of the fuel cell stack or vice versa. This can be continued for the fuel cell engine and its relation to the vehicle. However, this is more or less useless because the task is to optimize the vehicle efficiency as a whole and not just the efficiency of a single component by lowering the efficiency of another component. So, it does not make a lot of sense, in regards to system aspects, to talk about the efficiency of "the reformer" alone as long as this word is not used for the complete fuel processor within its well-defined limits.

Fuel processors have two important interfaces to the fuel cell system:

1. An interface to the air supply of the fuel cell to import the air for the air-consuming components such as partial oxidation or preferential oxidation reactors. The air compressor is usually not considered in the efficiency calculations for the fuel processor, but the hydrogen consumed by the air is considered since it leads to efficiency losses.
2. The second important interface is to the fuel cell that is embodied in the fuel processor. The fuel cell can be considered a hydrogen consumer with an efficiency of less than 100%, which means that some hydrogen is leaving the fuel cell again and flowing back into the fuel processor.

The second interface is now considered in more detail. The system schematic in Fig. 4 shows a fuel processor that needs thermal energy for the vaporizer and the reformer, wherefore a catalytic burner is included. This burner can be used to burn the excess hydrogen that is leaving the fuel cell. The fuel cell will always deliver some hydrogen because it cannot be operated as a dead-end unit with a reformate containing more than pure hydrogen. How much hydrogen a fuel cell can utilize depends on its internal structure (e.g. non-uniform flow distribution will result in lower utilization) and on the performance of the MEA's. Usually the utilization can be lowered but not easily increased. As long as the fuel processor needs all the hydrogen leaving the fuel cell there are no efficiency losses. But if the fuel processor is already exothermic, as it is in the case of a partial oxidation process the excess hydrogen causes efficiency losses based on the hydrogen utilization of the fuel cell stack. The fuel processor efficiency including heat losses can vary between

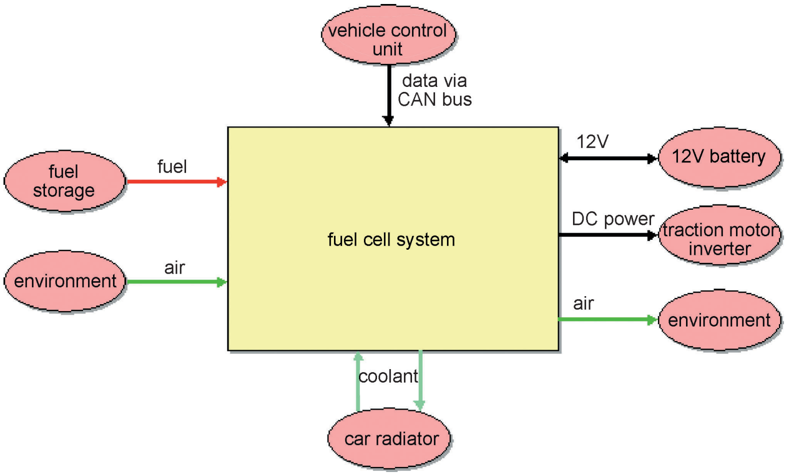

Fig. 3. Schematic view of a fuel cell engine.

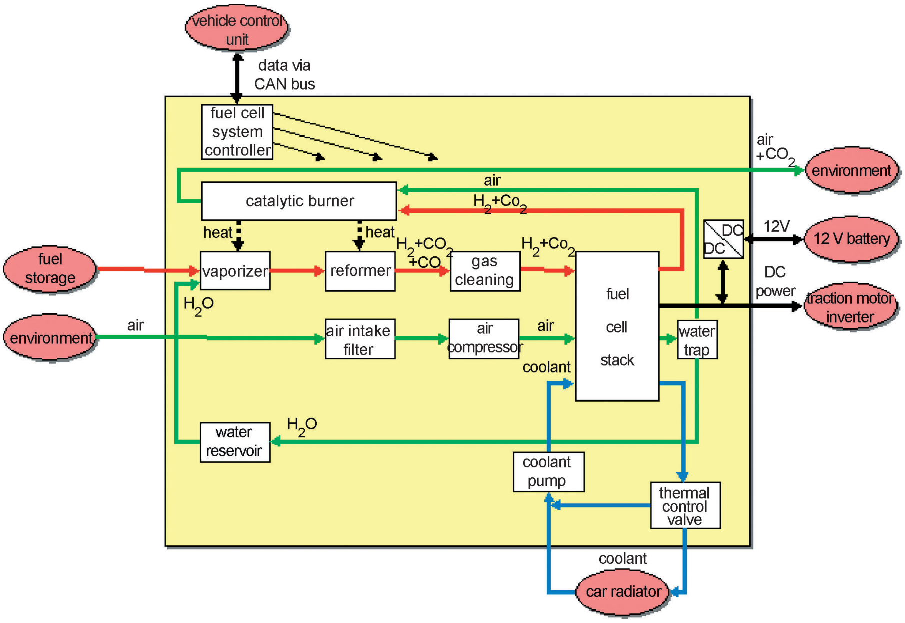

Fig. 4. Schematic view of a fuel cell system with fuel processor.

63 and 85% with the highest efficiency at 100% utilization of the fuel cell (Fig. 5).

The possible need of hydrogen by the fuel processor depends of course on the fundamental process of the hydrogen production. Depending on the amount of oxygen present in the process the fuel processor can be operated either exothermically or endothermically, varying from partial oxidation via autothermal reforming to pure steam reforming. Therefore, a fuel processor that can be operated under varying conditions is very helpful in optimizing the fuel cell engine's efficiency during the varying conditions of a drive cycle.

This situation can partly be avoided by using a hydrogen separation membrane in the fuel processor. Assuming that such a membrane is 100% selective for hydrogen as

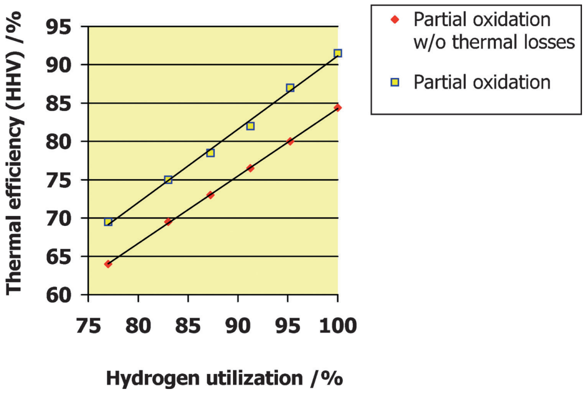

Fig. 5. Impact of hydrogen utilization on fuel processor efficiency (partial oxidation of methanol) [4].

in the case of a non-porous palladium membrane we have a pure hydrogen system on the fuel cell side. This eliminates the utilization problem with a single stroke but does not remove the need for making use of the unused reformate leaving the membrane unit on the fuel processor side. Hence, the previous discussion regarding exothermic and endothermic fuel processors comes up again.

There are other possibilities to use the excess hydrogen, including turbines, expanders, or even additional fuel cells, but they only add complexity to a system that is already regarded as very complex by many people.

As a general conclusion of the discussion about fuel processor efficiencies it can be stated that

1. fuel processors with steam reformers are the most efficient followed by those with autothermal reformers;
2. fuel processors based on pure partial oxidation offer the lowest efficiency.

4. Operating temperatures

Another aspect is the choice of the operating temperature of the fuel processor, and this is where the chemical structure of fuel plays an important role. Basically, there are two alternatives for a fuel processor:

1. it can be operated at a low temperature (250–300 °C) with all components (vaporizer, reformer, gas purification and perhaps catalytic burner) operating more or less at this temperature; or
2. the individual components operate at different temperature levels (250–800 °C) depending on their individual chemical reactions.

The first version can only be realized with fuels such as methanol or DME, which are easily processed. The second alternative is valid for all so-called "multi-fuel" systems with a high-temperature step for cracking fuels such as methane or hydrocarbons, followed by at least one shift unit, and a further gas cleaning unit, for example, preferential oxidation. Today, most high temperature fuel processors for vehicles use autothermal reforming or partial oxidation in the high temperature step.

Systems based on first alternative can be integrated to a higher degree than those based on second alternative. The Xcellsis ME 50-3 and ME 75-5 fuel processors may be regarded as examples showcasing the benefits of a high degree of integration.

5. Examples for fuel processors for vehicles

5.1. Low temperature fuel processors

The ME 50-3 fuel processor that is built into Necar 3 is a system with a low degree of integration. It has one component per function and makes you think of a small chemical plant with two boxes—one for the reformate production, containing catalytic burner, vaporizer, and steam reformer connected by a loop of thermal oil—as well as a second box with the water-cooled selective oxidation unit. It is perfectly clear why Necar 3 became a two-seater with these boxes in the trunk. Nonetheless, it was hereby possible to demonstrate that a vehicle with a fuel processor can be operated dynamically without an auxiliary battery or hydrogen storage. The dynamics were possible by means of a special vaporizer as a heat exchanger using μ-technology.

The ME 75-5 fuel processor is much more integrated, featuring components with several functions (Fig. 6):

1. vaporizer and superheater in a combined catalytic burner/vaporizer,
2. reformate production in a combined reformer and preferential oxidation reactor,
3. start-up and dynamics in a novel reactor allowing exothermic, autothermal, and endothermic operation.

This technology allows us to design a very compact fuel processor as one part of a fuel cell engine with in total four boxes that all fit together under the floor of a vehicle, as demonstrated in the Necar 5 as well as other vehicles.

5.2. High temperature fuel processors

High temperature fuel processors are necessary for fuels that are based on hydrocarbons with very stable chemical bonds. This is the case for methane with its very high symmetry and for molecules containing carbon–carbon bonds as in the case of LPG, gasoline, and diesel-type fuels. It is well known, in the chemical and petrochemical industry, that those compounds can be handled in very efficient processes that are more or less state of the art. All varieties from steam reforming to partial oxidation are possible and are also used within the corresponding surroundings of a large plant. In principle, there is always a step needed to produce a syngas consisting of hydrogen and carbon monoxide, which is then enriched in hydrogen in one or more shift stages. The hydrogen-rich reformate is then cleaned in additional steps such as preferential oxidation, membranes, or pressure swing adsorption.

The challenge enters when wanting to transfer these processes into a car. All of the above mentioned processes operate at different temperatures and sometimes also at different pressure levels. This means that there is a need for heat exchangers and maybe also for additional compressors in our system. Then, one has to deal with the possibility of soot formation, which will always happen if something gets out of control. The important parameters for a successful system operation are:

1. temperature control during start up, shut down and operation, especially during load transients;
2. control of S/C;
3. control of O/C (if necessary).

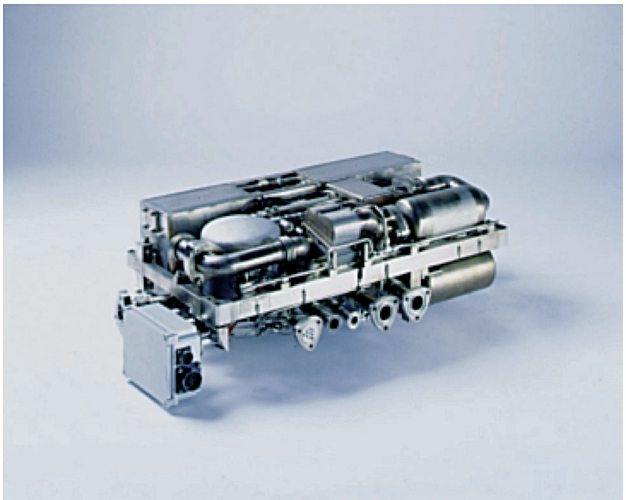

Fig. 6. ME 75-5 fuel processor.

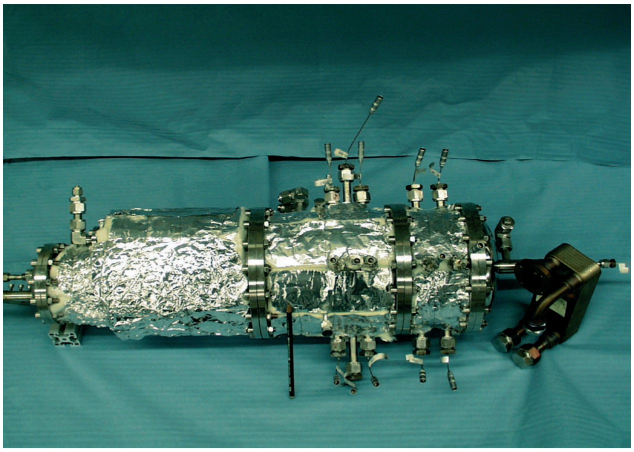

Fig. 7. Example of a gasoline reformer.

The necessity of large amounts of process water adds further complexity to the system as there are additional means needed for recovering the water and cleaning it before it can be used again in the fuel processor. Of course this is also the case for a low temperature system, but in the latter case we need less water.

Compact gasoline fuel processors can be constructed based on autothermal high temperature technology with integrated heat exchangers and shift stages (Fig. 7). Xcellsis demonstrated this technology both in 5 and 50 kW fuel cell lab systems (MF 5-1 and MF 50-1). A packaging study shows that, with our current level of knowledge, fuel processors can only be designed with an integration level similar to Necar 3. Until now we have yet to integrate such a fuel processor into a vehicle. Our simulations show that such a vehicle would need a large battery to ensure drivability. Further, it would have difficulties to compete with a very good diesel ICE (as hybrid or not) in terms of efficiency.

So, maybe high temperature fuel processors are better suited for non-mobile applications including fuel stations or in combination with large electric batteries to offer vehicles with better efficiencies than today's gasoline ICE and lower emissions than today's diesel ICE.

6. Development challenges

Nevertheless, fuel cell vehicles featuring any kind of fuel processor still present us with challenges, which need to be met until commercial fuel cell vehicles with fuel processors will be on the streets.

1. The interfaces between the fuel processor and the peripheral components:
   - pump and flow controller-dosing accuracies (stoichiometries);
   - water trap efficiencies (contaminants, thermal shocks);
   - contaminants (e.g. lubricants, corrosion products);
   - condensates;
   - catalyst particle emissions (valve operation).
2. The interfaces between the fuel processor and the fuel cell stack:
   - CO content during cold start and mode transition;
   - temperature and humidity control of reformate;
   - high-anode stoichiometries;
   - load-variable anode and cathode stoichiometries.
3. Control issues:
   - H_2-stoichiometry control (e.g. balancing of H_2-generation and consumption);
   - optimisation of dynamic response time;
   - reduction of dynamic losses;
   - control of mode transitions.

7. Conclusions

Hence, the overall and possibly thought-provoking conclusions are as follows:

1. Although the use of hydrogen seems very attractive at first glance both in terms of the fuel cell vehicle and of infrastructure, there are good reasons for the use of a liquid fuel together with an on-board fuel processor. Today and in the foreseeable future it is an open issue when a hydrogen economy will be installed for commercial use of fuel cell vehicles.
2. A discussion about fuel processor efficiencies has to be based upon a clear definition of the interfaces to the surrounding system. The chemical technology used for the hydrogen production (steam reforming, partial oxidation or autothermal reforming) has a strong impact on the efficiency of a fuel cell engine.
3. Low temperature processes are advantageous from the technical point of view for vehicular applications. High temperature processes are also possible, but they require additional efforts in the vehicle. From today's point of view, however, they may be better suited for stationary

applications than for vehicle drive trains, but there is no clear decision.

Although the technology has been successfully demonstrated since some years, there are still several challenges to be met for all types of fuel processors in vehicle systems, but there are no insurmountable hurdles any more. Fuel processors are more than just an option for fuel cell vehicles. They seem to be essential for a successful commercialization in a mass consumer market.

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