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ABSTRACT:
The ECN fuel cell related R&D program on fuel cells is linked to the stationary market and the automotive market. This paper will summarize our R&D activities for the automotive market.
The role of fuels cells in two transport application area's will be described: the development of dedicated hydrogen based platforms in combination with advanced electricity storage for special logistic applications and the APU market for passenger cars and trucks, as well as for ships and airplanes. The associated aspects of hydrogen transport and storage, as well as the reforming of logistic fuels and bio-fuels to hydrogen will be described with some illustrative examples.
These examples show that an integrated approach using applied catalysis, chemical reactor design and engineering, process simulation, control modelling and electrical engineering is required to address all aspects of the development of fuel cell technology for automotive applications.

The paper concludes with a summary of the important environmental and economic drivers that influence the fuel cell market application.

KEYWORDS: *Fuel Cells, Transport, Hydrogen, Reforming, Auxiliary Power Unit*

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1 Introduction

Fuel cells are electrochemical devices that convert hydrogen to electricity and heat. Fuel cells are generally seen as one of the important enabling technologies for introduction of the hydrogen economy. One of the important applications of fuel cells is the transport sector. Fuel cell technology is a clean and silent technology and enjoys a higher well to wheel efficiency than technology based on internal combustion engines. The main obstacles to be overcome for large-scale commercial application op fuel cells are still a too high cost, the lack of a hydrogen fuel infrastructure and the lack of dedicated balance of plant components.

1.1 Fuel cells for transport applications

The principle of a fuel cell is well described elsewhere (1,2); in this paper only the fuel cells that are considered for transport applications will be briefly described.

Today, Proton Exchange Membrane Fuel Cells (PEMFC) are the most widely used fuel cells in transport applications. It is estimated (3) that more than 90 % of all fuel cell vehicles on the road have been equipped with a PEMFC. The PEMFC fuel cell enjoys a high power density at its operating temperature of about 70 ^{0}C, which provides an attractive property for start-up purposes. There are still some drawbacks of the current state of the art PEMFC, one issue concerns the low CO tolerance. Most PEMFC stacks require CO levels well below 10 ppmv. Higher CO levels result in loss of fuel cell performance. A second aspect is the operating temperature of the PEMFC that may result in unacceptable large radiator area for cooling the fuel cell, in particular in hotter climate area's of the world (4). These two aspects have led to a research effort devoted to proton conducting membranes at elevated temperatures (5,6). It is expected that these developments will take a long time before those will be used in systems for practical applications (7). This type of high temperature PEMFC can be regarded as second generation. The conventional low temperature PEMFC stacks will be the first product in practical applications of today, yet requiring relatively pure hydrogen. PEMFC stacks in transport applications are used in a hybrid configuration with electricity storage devices, such as batteries or super capacitors.

For auxiliary power units in vehicles, also SOFC fuel cells are being considered as candidates. SOFC's are characterised by their high operating temperature, the state of the art ytria stabilised zirconia
based SOFC has an operating temperature of 800-1000 ^{0}C. There are two basic configurations of SOFC, a tubular concept and a planar concept. The tubular SOFC concept is regarded as suitable for large-scale stationary applications. For automotive applications a planar SOFC concept is preferred, due to the higher power density. An extensive review on high temperature SOFC has been written by Kendall et al (8).
The SOFC application for transport use is limited to the APU in vehicles, the SOFC concept is not seen as viable for traction purposes, due to the specific requirements that are associated with traction, in particular start-up behaviour and dynamic load changes.

1.2 Hydrogen infrastructure

A key issue for the use of fuel cells in the transport sector is the availability of hydrogen as a fuel. Without a widespread network of hydrogen stations there is no large-scale introduction of fuel cell cars and without a significant amount of fuel cell cars there is no incentive to make heavy investments in a hydrogen infrastructure. There are different scenario's being investigated to find solutions for this chicken & egg problem.

It is generally expected (9) that until 2030 hydrogen production from fossil fuels with carbon capture and sequestration (CCS) will be the most important production source in Europe, with renewable hydrogen slowly being phased in. Hydrogen infrastructure build-up will likely comprise both central and on-site hydrogen production.

In some localized area's it would be possible to link existing large-scale hydrogen production facilities with a pipeline network. Such a scenario has been proposed in Germany (10).
In some area's of the world there is already a network of hydrogen stations; in California there are now 15 stations in operation and 16 planned (11).

1.3 Hydrogen storage

The on-board storage of hydrogen is critical for the transport applications of fuel cell vehicles. Critical issues involve the possible driving range (that should be comparable with gasoline cars), the volume (that should not occupy too much storage volume), the weight and the cost.

The state of the art hydrogen storage options include compressed hydrogen, liquid hydrogen and metal hydrides. The DoE has published targets for on-board hydrogen storage devices (13).The energy storage density for hydrogen (in MJ/l) amounts to 2.8 for hydrogen as gas (345 bar) to a value between 7 and 12 for meta hydrides, 8.4 for liquid hydrogen and 31.1 for gasoline as the reference value.
Hydrogen storage in nanotubes or in zeolites is also under investigation (14,15).

1.4 On board fuel reforming

On-board fuel reforming is an option to utilise the existing liquid hydrocarbon fuel infrastructure and to produce the hydrogen on-board a vehicle. In this way, on-board fuel reforming could be an intermediate solution, in anticipation of the establishment of a hydrogen infrastructure, providing a seamless transition to a hydrogen based infrastructure. The reformer–FC system that would be needed is obviously more complex than an on-board hydrogen-FC system.

The reforming for automotive applications takes places with an air assisted primary reforming, indicated as Catalytic Partial Oxidation (CPO) or as Autothermal reforming (ATR). In the case of CPO air is being applied together with a hydrocarbon fuel, in the case of ATR air is applied together with steam and a hydrocarbon fuel. The gastreatment consists of shift conversion and CO removal. The exhaust of the PEMFC fuel cell is cionverted in an after burner reactor.

The on-board reforming system presents a number of technical and economical challenges hat include volume targets, weight targets, start-up requirements (16).
These requirements translate in a number of goals for the reformer system and in particularly for the catalysts used in the different reactors:

- Higher catalytic activity than conventional pelletized catalysts
- Good stability for dynamic operation and fast load changes
- Non pyrophoric properties, ability to operate under air atmosphere
- Good sulphur resistance
- Small size and low weight (high space velocities)
- Low costs
- Fast and safe start-up/shut-down
- High turn-down ratio's
- Low pressure (0-few barg)
- Minimum number of heat exchangers
- Integration of heat exchangers with reactors

In table 1 the properties of conventional reforming catalysts and reforming catalysts tailored for automotive application are compared.

Table 1. Fuel processor targets* compared to industrial practice

*Targets: 50kWe Fuel Processor for automotive applications DOE/Freedomcar

	Fuel Processor for automotive application	**Industrial Hyrogen plant**
Size steps	GHSV (hr^{-1})	GHSV (hr^{-1})
Reformer	200,000	1,000
Water gas Shift reactor I (HTS)	60,000	1,000
Water gas Shift reactor I (LTS)	60,000	3,000
CO removal	150,000	3,000
Pressure (bara)	1-10	> 30

Although a number of companies have abandoned the idea of on-board reforming, Nuvera, together with Renault is developing a concept (STAR) based on an on-board auto-thermal reformer for passenger cars (17).

2. Hydrogen Fuel cell systems for traction

Hydrogen fuel cell systems are now being introduced in the market as fleet demonstrations (18), FC cars enjoy well to wheel (WTW) efficiencies that more than double the WTW efficiencies of ICE based cars (19). It is generally accepted that FC cars will penetrate the market with specific fleet owner vehicles (buses, taxis) and particular niche applications (forklift trucks, vehicles for internal transport, etc.) or in situations where local conditions (for example air pollution in urban area's) provide a drive to embark on fuel cell technology. Mass introduction of FC cars is expected from 2015 onwards. The examples given below cover some of the niche applications.

2.1. Special transport applications: the ECN HydroGEM vehicle

ECN has converted is a GEM (supplied by Global Electric LLC) electric vehicle to a hybrid fuel cell system that generates approximately 5 kW of electric power. The GEM vehicle is typically used in local transport such as inland parks, logistic in-company area's, etc.

This vehicle is equipped with a PEMFC stack of 5 kWe (manufactured by ECN) and includes all balance of plant equipment such as a dedicated air blower (Vairex Corporation), a hydrogen recycle compressor (H2 systems Inc.), a dedicated DC/DC converter (ECN design), a humidifier (PermaPure), hydrogen storage (Dynetek Corp.) and includes a MABX (dSpace) process controller. The system design, engineering, procurement, assembly, control design and testing was executed by ECN.

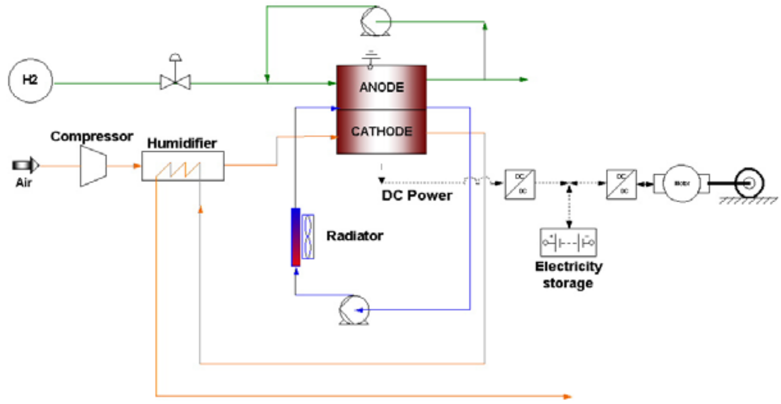

Figure 1. Simplified Process Flow Diagram of the hybrid Fuel Cell/Electricity storage system

Figure 1 gives the process configuration of the H2-PEMFC system with the main components. Hydrogen will be stored on board the vehicle and delivered to the fuel cell anode. The unused hydrogen in the anode will be recycled to maximise the fuel efficiency. The required cathode air is compressed and humidified and fed to the fuel cell cathode. The cathode outlet will be used to humidify the inlet. The cooling of the fuel cell stack is carried out with demi-water. Heat is rejected to the atmosphere via a radiator.

2.2. Hydrogen based application for a scooter

There are a number of drivers for the development of Hydrogen Fuel Cell Scooters. In Europe, there are more than 20 million two-wheel vehicles on the road. These vehicles are mostly (85-90 %) used in urban area's. Therefore, use of hydrogen fuel cell technology for scooters will help considerably in reducing the pollution in urban area's, in particular NOx, SOx and small particles. ECN has been involved in a major European project (FRESCO) for the development of a fuel cell based scooter.

Scooters are popular vehicles in cities and urban area all over the world. Reducing the emissions and energy use of these vehicles would mean a great asset in improving air quality and preserving the environment, including the reduction of noise. Highly efficient fuel cells running on hydrogen and feeding an electric motor form the preferred technology for the longer term when patrol and other fossil fuels will be more and more replaced by sustainable hydrogen as energy carrier. By participating in a development project on this technology, Piaggio extends its activities in environmentally benign propulsion systems that have been started with its leading role in hybrid technology. In hybrid technology normal internal combustion engines are used, but in a very efficient way. Because these engines still use petrol, hybrids can already be used today and thus offer short-term environmental improvements. The longer-term fuel cell technology will be the step to vehicles with no emissions at all.

In the FRESCO project Piaggio teamed up with the Energy research Centre of the Netherlands (ECN) that built the "electrochemical engine" comprising the fuel cell, Selin Sistemi Spa from Italy that realized the electric motor and its electronic control, and CEA Valduc from France that developed the on-board hydrogen storage tank and the refilling facility. The Universities of Pisa and Florence, and ESMA Company from Russia delivered additional services and components to the consortium. Although the FRESCO project officially ended in July 2005, an additional effort was made by the project partners and University of Pisa to enhance the vehicle performance and effectiveness, that has lead to new tests made in February, 2006, in which vehicle acceleration, maximum speed and range were verified. Earlier fuel cell developments for scooters were limited to small devices for charging an on-board battery. In the FRESCO project, propulsion relied completely on the fuel cell alone. Because of its relevance for preserving the ecosystem, the development was partially funded by The European Commission in its fifth framework program (Contract nr ENK6-CT-2001-00565).

In March 2006 the testing was completed with drive testing on the outside test circuit with 10 laps and a driving speed exceeding 50 km/hr.

3. APU systems for transport applications

Fuel Cell APU systems are considered for a number of applications that include passenger cars, trucks, trains and aircraft. An important driver for the APU application is related to the increased electricity demand for auxiliary power use. Hereafter, a few examples of developments in this area at ECN will be discussed.

3.1. Diesel based system for naval applications

Internationally, there exists a lot of interest for the use of fuel cells for military naval applications. The role of fuel cells in this application is to provide the on-board power supply. One of the advantages is the reduction in pollution in harbour area’s.
ECN has been involved in a WEU project (DESIRE), together with Germany, UK and Turkey. This project was carried out in the period 2001-2005. The DESIRE project is dedicated to the development of a fuel processor for the conversion of F-76 marine diesel into hydrogen rich gas suitable for fuel cells. In phase 1 of the project the preferred system was selected: A steam reformer (SR) fuel processor unit in combination with a PEM fuel cell. Test experiments were executed with the fuels shown in table 2.

Table 2: Comparison of diesel specifications

	F-76	City Diesel (Aral)	Hydroseal (G 232 H)	Units
Density	820 - 880	820 - 845	805 - 820	kg/m³
Recovery at 350 °C	> 85	> 85	100%	% v/v
Flash point	> 61	> 55	> 101	°C
Kin. Viscosity	1.7 - 4.3	2.0 - 4.5	3 - 4	mm²/s
Pour point	< -6		-50	°C
Cloud point	< -1			°C
Sulphur content	< 1.0	< 0.001	< 0.0001	% m/m
Ash content	< 0.010	< 0.01		% m/m
Carbon residue	< 0.20	< 0.30		% m/m
Copper corrosion	< 1		< 1	class
Cetane number	> 45	> 51		-
Water reaction	< 2			ml

Water content		< 0.02	< 0.005	% m/m
Lubricity at 60 °C		< 460		µm
Aromatics		< 11	< 0.03	% m/m
Sediment on storage	< 10			mg/l
Additives	To be agreed	Standard		

The role of ECN was focused on the system development and to the testing of all the required components in a line-up configuration. This was executed in the existing 25 kWe Fuel Processing Test Installation at ECN. Figure 2 presents a block scheme of the test installation.

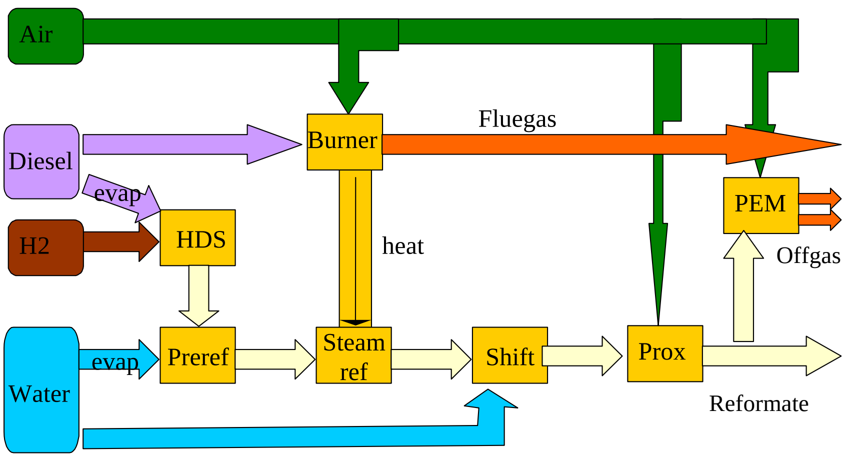

Figure 2. Simplified block diagram 25 kW demonstrator for diesel reforming

The results from the desulphurisation and pre-reforming tests conducted at ECN revealed that at the operating pressure of the unit (max. 4 bar) the desulphurisation of F-76 marine diesel with 0.12 wt% S was not successful: desulphurisation down to 300 ppm S was obtained whereas a sulphur level < 0.1 ppm S is required.
Small scale tests with City Diesel (10 ppm S, imported from Germany) revealed some catalyst deactivation, although the conversion levels were sufficient. Therefore the demonstration experiments at 25 kWe scale were performed with Hydroseal G 232 H (from Total-Fina-Elf) as well as with City diesel. Hydroseal is a C13-C15 aliphatic solvent with aromatics content < 100 ppm and with < 1 ppm S. The City diesel applied contained < 10 ppm S and was imported from Germany. It was found that Hydroseal as well as City diesel was converted to a reformate gas with < 10 ppm CO at all operating conditions. This resulted in a reformate pure enough to be applied to run a fuel cell. A side steam of reformate product was provided as fuel to a 2 kWe PEM fuel cell stack (ECN).

The overall fuel processor efficiency turned out to be 82% at full load. This efficiency is defined as (flow*LHV) of the reformate, corrected for the hydrogen supply for the HDS divided by the (flow*LHV) of the diesel fuel to the process and the steam reformer burner.

The 25 kW demonstrator at the ECN premises successfully showed the conversion of diesel fuel to a hydrogen rich gas, suitable for use in a PEM Fuel Cell. The results led to directions for improvements of the system, especially with respect to the use of NATO F-76 diesel fuel. Clearly, the sulphur removal from diesel fuels is a critical issue to be solved before wide-spread use of fuel cells in this application area will occur.

3.2. Kerosene based system for aircraft application

Fuel Cell technology for auxiliary power application in aircraft is a challenging option of the future and is driven by an increased electrical energy demand driven as a result of a modern cabin infrastructure that will occur in the next aircraft generation. At the same time a reduction of fuel consumption, exhaust- and noise emissions of aircraft will be required.

The demands of the aircraft industry to application of fuel cell technology are ambitious in comparison to automotive applications, especially with regard to safety, reliability and to the operating conditions of low temperature and low pressure at elevated altitudes.
For the next aircraft generation, pneumatic and hydraulic systems will be replaced step by step by electrical, electro-pneumatic and electro-hydraulic systems. Therefore, a highly efficient primary electric power source is needed. Fuel cell systems are the most promising opportunity to meet that demand.

ECN is involved in a European project named CELINA (FUEL CELL APPLICATION IN A NEW CONFIGURED AIRCRAFT, contract AST4-CT2005-516126), under project coordination of Airbus and executed with 20 partners in Europe.

The main objective of the CELINA project is the investigation of the behaviour and assessment of the limiting conditions of a PEMFC fuel cell system as APU in an aircraft in terms of different system parameters such as performance output, thermal management, mass flow, cooling and air supply. Dynamic simulation models will simulate the fuel cell system under the aircraft operating conditions. The dynamic model will be verified by testing as far as possible. The target is to identify all technical issues, which have to be modified to reach an airworthy design.

The role of ECN in this project lies in the modeling of the APU system, together with experimental verification of the reformer components at a scale of about 5 kWe. The modeling implies both steady state modeling (using Aspen One) and dynamic simulation (using Matlab Simulink) for a 50 kWe APU capacity. The modeling of the reformer section will be integrated into the overall APU model for the aircraft.

A number of conceptual variations have been studied and a conceptual architecture has been identified.
The system performance has been calculated for various operating modes, with the load and altitude as main parameters

For operation at 41,000 ft altitude the nett efficiency is some efficiency points lower. This is mainly related to the higher parasitic power requirements of the air compressor.

3.3. Bio-diesel based PEMFC system

Bio-fuels are fuels that are in principle CO_2 neutral and are considered both as fuels for internal combustion engines as well as fuels that can be converted to hydrogen for use in fuel cells. In Europe, bio-diesel is typically produced from field crops, whereas elsewhere in the world bio-diesel is mostly made from recycled cooking oil.

ECN, together with other partners, carries out a European research project (contract ENK6-CT-2002-00612) on the development of a bio-diesel based fuel processor for an APU for a vehicle. The conceptual design of a 10 kWe system has been described elsewhere (20).

In this project, a considerable amount of process intensification has been applied, in particular with regard to the integration of reactors and heat exchangers. As example, The ATR reactor has been integrated with steam generation and steam superheating.

The reformer system has been assembled by Scandiuzzi s.r.l. in Italy (one of the partners in this project) and is subject to verification and testing at the ECN facilities in Petten in the summer of 2006.

4. Environmental and economic aspects

The main incentive for the use of Fuel Cell cars lies in the improvement of fuel economy and in the reduction of air polluting emissions in comparison to gasoline based ICE cars. In addition, the tendency to become more independent on foreign oil supply is an important political driver for the introduction of FC cars.

It is generally accepted that gasoline hybrid ICE based cars have a Tank to Wheel (TTW) efficiency that is equivalent to the FC hybrid car TTW efficiency (21). The hybrid hydrogen ICE car has an efficiency that is lower than the FC hybrid car efficiency.
The hybrid gasoline ICE cars still do emit pollutants like NO_x, SO_2, hydrocarbons and small particles and will still be dependant on foreign oil supply. These aspects will differentiate this gasoline hybrid ICE configuration from the hydrogen based configurations.

5.Conclusions

Fuel cells are important enabling devices to utilise the hydrogen that will be available in the upcoming hydrogen economy. Fuel Cell cars offer significant benefits in fuel economy and in reduction of polluting emissions in comparison to the ICE based cars. Hybrid ICE cars offer fuel economies that are comparable with FC cars, but FC cars have considerable lower emission levels of NO_x, SO_2, hydrocarbons and small particles. Use of fuel cells in transport applications provides significant benefit in terms of energy saving and pollution reduction Yet there are still further efforts needed on component and system level to produce reliable and cost-effective fuel cell systems for transport applications.
An integrated approach using applied catalysis, chemical reactor design and engineering, process simulation, control modelling and electrical engineering is required to address all aspects of the development of fuel cell technology for automotive applications.

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