HYDROGEN FUEL CELL VEHICLE AND A STAND-ALONE RENEWABLE ENERGY-BASED REFUELLING STATION

Master's Thesis
University of Jyväskylä
Department of Biological and
Environmental Science
Master's Degree Programme in Renewable
Energy
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20th August, 2007

JYVÄSKYLÄ UNIVERSITY - Faculty of Mathematics and Science -
Department of Biological and Environmental Science -
Master’s Degree Programme in Renewable Energy

Pakkanen Atte: Hydrogen fuel cell vehicle and a stand-alone renewable energy-based refuelling station

Master´s Thesis: 107 p., 2 appendixes (2 p.)
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August 2007

Key words: hydrogen, fuel cell, hydrogen vehicle, hydrogen storage, metal hydride storage, electrolysis, renewable energy, alternative transportation fuels

ABSTRACT

Energy use for transportation purposes and the number of passenger vehicles is increasing rapidly around the world. Fuelling infrastructure based on hydrogen provides an attractive alternative to predominating fossil fuels. This thesis project includes research on hydrogen fuel cell vehicle and its local refuelling station based on hydrogen production from renewable energy sources. Hydrogen produced via electrolysis is stored in portable metal hydride containers to be used in a fuel cell hybrid vehicle. The test vehicle is equipped with a battery unit and two 6 kW electric motors.

The targets of this thesis project were to test and estimate the driving range of the vehicle, to examine the behaviour of the metal hydride containers under changing temperatures and determine the rate of hydrogen production via electrolysis. Also hydrogen leak tests to assure safe testing procedures in the future were included to these initial tests.

The driving range tests resulted 7.6 km with full batteries only. According to the calculations, this range limit can be doubled with one full metal hydride container and a polymer electrolyte fuel cell used in this study. The metal hydride tests resulted that temperatures under 10 °C around the H_2 container create a risk of PEM fuel cell malfunction. The hydrogen production rate under normal conditions was measured 530 cm^3 / min with a nominal pressure of 6.7 bars. Leak tests proved that safe operation of the system is possible, but requires a careful system built-up. The equations produced by these leak tests can be used to estimate the fuel losses due to leakage in the future experiments.

JYVÄSKYLÄN YLIOPISTO - matemaattis-luonnontieteellinen tiedekunta
bio- ja ympäristötieteiden laitos - uusiutuvan energian maisteriohjelma

Pakkanen Atte:	Vetyauto ja uusiutuvaan energiaan perustuva tankkausasema
Pro gradu –tutkielma:	107 s., 2 liitettä (2 s.)
Työn ohjaajat:	professori Mohanlal Kolhe ja TkT Margareta Wihersaari
Tarkastajat:	professori Mohanlal Kolhe ja FT Jussi Maunuksela
Elokuu 2007	

Hakusanat: vety, polttokennot, vetyauto, vedyn varastointi, metallihydridivarasto, elektrolyysi, uusiutuva energia, vaihtoehtoiset liikennepolttoaineet

TIIVISTELMÄ

Liikenteen energian kulutus ja maailman autokanta kasvavat nopeasti. Vetyyn perustuva liikenneinfrastruktuuri tarjoaa houkuttelevan vaihtoehdon nykyiselle fossiilisiin polttoaineisiin perustuvalle tekniikalle. Tämä pro gradu -työ liittyy koemittakaavan laitteistokokonaisuuteen, jossa vetyä tuotetaan elektrolyysillä hyödyntäen uusiutuvaa energiaa. Tuotettu vety varastoidaan metallihydridisäiliöihin ja käytetään edelleen polttokennoauton polttoaineena. Tämä koekäyttöön rekisteröity ajoneuvo on varustettu kahdella 6 kW sähkömoottorilla, ja sillä voidaan ajaa myös ilman polttokennoa käyttäen pelkkiä akkuja energiavarastona.

Tässä työssä oli tarkoitus tutkia tämän kevytajoneuvon pisintä mahdollista ajomatkaa, vetysäiliöiden käyttäytymistä eri lämpötiloissa sekä vedyn tuotantonopeutta elektrolyysillä. Tähän työhön sisältyi myös vuototestejä, joista saatua tietoa voidaan tulevaisuudessa hyödyntää auton tehokkuuteen ja turvalliseen käyttöön liittyvissä tutkimuksissa.

Ajoneuvon ajomatkaksi täysillä akuilla ilman polttokennoja ja vetyä saatiin 7,6 km. Laskennallisesti tämä matka voidaan kaksinkertaistaa yhdellä täydellä vetysäiliöllä ja laitteistoon kuuluvalla 500 W PEM polttokennolla. Vetysäiliöillä tehdyissä testeissä paljastui, että jo alle 10 °C lämpötila voi aiheuttaa toimintahäiriöitä PEM polttokennon toiminnassa johtuen vetysäiliön paineen alenemisesta alhaisissa lämpötiloissa. Elektrolyysillä tuotetun vedyn valmistusnopeudeksi saatiin käytössä olleella laitteistolla 530 cm^3 / min, joka mitattiin 6,7 baarin paineessa. Vuototestit osoittivat, että turvallisen vetylinjan rakentaminen on mahdollista, mutta vaatii huolellisuutta. Tutkimuksessa määritettyjä yhtälöitä voidaan käyttää tulevaisuudessa vuotohäviöiden arvioimiseen vedyn kulutusta seurattaessa.

CONTENTS

NOMENCLATURE

ABS	Acrylonitrile butadiene styrene
AFC	Alkaline fuel cell
AKE	Ajoneuvohallintokeskus (The Finnish Vehicle Administration)
CSV	Comma Separated Values
FCV	Fuel cell vehicle
FCHV	Fuel cell hybrid vehicle
GHG	Greenhouse gas
H/M ratio	Hydrogen atoms per metal atom –ratio
HEV	Hybrid electric vehicle
HHV	Higher heating value
ICE	Internal combustion engine
LHV	Lower heating value
LP	Liquefied petroleum
MH	Metal hydride
NPT	National Pipe Taper thread
PEM electrolyser	Polymer electrolyte membrane electrolyser
PEMFC	Polymer electrolyte membrane fuel cell
PV	Photovoltaic
PVC	Polyvinyl chloride
RE	Renewable energy
STP	Standard temperature and pressure
WTG	Wind turbine generator

1 INTRODUCTION – Arrival of the hydrogen age?

The modern world is currently facing two great challenges that are strongly connected to each other: energy use and the growth of population. The energy consumption per capita is increasing continuously. In 2004, total primary world energy consumption was approximately 11.7 Gtoe (Marbán & Valdés-Solís 2007), and this figure is estimated to grow exponentially. The situation is made more complicated by the fact that also the amount of energy consumers is increasing rapidly. Despite the ongoing renewable energy promotion, fossil energy sources are clearly dominating the current energy sector. Of all the fossil fuel consumed worldwide, transportation accounts for a great share. Despite the fact that alternative fuels for transportation have been available even before the invention of petroleum fuelled ICE, the fossil fuels predominate clearly the modern transport sector.

Taking to account these facts, the introduction of a permanent alternative fuel for transportation is a necessity rather than an option. Hydrogen is one of the potential fuels for the future. The interest on hydrogen has been increasing lately because of the substantial benefits it possesses. Hydrogen can be used in various end-use applications and it can form a basis for a non-polluting energy infrastructure. Once integrated with small scale renewable energy sources, hydrogen provides efficient independence from centralized electricity production and combustion of fossil fuels. The growing interest on environmental issues and a search for an alternative for fossil fuels have launched numerous hydrogen energy programs worldwide. In transport sector, almost every major auto manufacturer has introduced their vision of a hydrogen vehicle of the future.

During the spring 2005, a small scale hydrogen vehicle project was started in the premises of the University of Jyväskylä. In addition to the hydrogen fuel cell hybrid test vehicle, the concept included PV panels and wind turbine which provided power to hydrogen production unit. The hydrogen produced was stored in two portable metal hydride units which acted as fuel tanks for the vehicle. This project was established to provide an experimental set-up of stand-alone refuelling system and a test vehicle which are completely based on renewable energy. The additional scientific value of the project comes from the northern location of the facility. Once the system is built, it can serve as a demonstration project for students and researchers in the university.

The preparation for this thesis work started by building the RE production system and by installing the equipments. The aim of this thesis was not to analyse the whole system, but to make initial tests to the components of the system and measure the driving range and performance of the hydrogen fuel cell vehicle. The components included an electrolyser, PEM fuel cell, alkaline fuel cell, metal hydride containers and other necessary equipment and measuring devices. The component testing provides a basis for the system improvements and for the development work in the future. It was obvious from the beginning that the test vehicle with the existing fuel processing equipment does not compete with modern automobiles in a technical sense. The final aim of the whole scheme was to provide a practical example that oil and grid independent and non-polluting shopping trip is possible in northern parts of Europe. This thesis work will form a basis for this scheme.

In this graduate thesis, the technologies discussed include hydrogen manufacturing, storage and its utilization in fuel cells. Safety issues concerning hydrogen use in vehicles is also included. In the chapter 6 the research targets of this thesis work are discussed in more detail. Nearly all the results are based on own experiments which were planned and realized during the spring 2006. A secondary research material was used only in determining the power consumption of the vehicle at different driving speeds.

Most probably, the research on this hydrogen test scheme will continue after this project. The hydrogen vehicles are rapidly increasing their minimal share of the global vehicle fleet. Also it would not be hard to make a list of topics requiring intensive research concerning the whole project. Hydrogen as an energy carrier has gained lot of notice lately and a myriad of hydrogen economy scenarios have been sketched during the last decades. Of all statements supporting the hydrogen transition, one is worth mentioning in the first pages of this thesis work. Ironically, it comes from the representative of a company which is strongly related to oil industry:

> “The Stone age did not end because we ran out of stones, and the Oil age will not end because we run out of oil”. Don Huberts, Shell Hydrogen (National Hydrogen Association 2007a)

2 ENERGY USE AND TRANSPORTATION

2.1 The challenge of the energy need

> "While sustainability is a state, sustainable development is a process of development, from where we stand today towards that ideal state." (Everard 2005, 28).

A quick look to the global energy industry reveals that the current situation is far from that ideal state described by Everard. The fact is that the world population will continue its rapid growth in the near future. At the middle of the year 2006 the population of the world was 6,555 millions and the expected population in the year 2050 is over 9,000 million (Population Reference Bureau 2006, 5). The question is how can world of almost 10 billion people be provided with adequate supplies of energy. To avoid possible global catastrophes due to short-sighted energy policies, this energy problem has to be solved efficiently, cleanly and in sustainable manner. Although economic growth, according to Lehtilä (2001, 191), is the key factor driving energy demand in the future, a permanent balance between economic growth and the environment is achievable.

World energy consumption has quadrupled between 1950 and 1992, from 76 500 Pj to 311 000 Pj (Imboden & Jaeger 1999, 65). Some researchers estimate that the climbing rate of world energy use is about 2 % (Lahidji et al. 1999, 8), but there are estimates of even more rapid growth in global energy use. According to International Energy Agency, nearly 60 % growth in energy use can be expected over the next 20 years (Tuhkanen, Hirvonen, Savolainen & Honkatukia 2001, 29). Commercial energy use per capita is highest in North America, where the consumption rate is 10 kW per head. Europeans consume energy at rate of 5 kW, and people in newly industrialising regions at 2 kW per head (Imboden & Jaeger 1999, 64). These figures are lower for majority of the world citizens. For example, commercial energy consumption rate is said to be 80 times lower in Africa than in the U.S. (McNelis, van Roekel & Preiser 2002, 195). Finland's energy intensity is one of the highest in the IEA countries, both per capita and per unit of GDP (IEA 1999, 15), mainly due to the fact that Finland has the coldest climate in Europe.

In the turn of the millennium, 95 % of the world's commercial energy production was based on fossil fuels (Imboden & Jaeger 1999, 64). Despite the pollution they create, fossil fuels are predicted to remain the predominant primary energy source, especially in developing countries. This creates a possibility that CO_2 emissions will grow even faster than the primary energy use (Tuhkanen et al. 2001, 29).

It has been roughly estimated that in order to stabilize the atmospheric concentrations of GHGs at a reasonably safe levels, the reduction of greenhouse gases produced by industrialized countries should be about 80 % by the end of the century (Lehtilä 2004, 191). This will certainly not happen without utilizing renewable energy sources. The facts presented above have accelerated the development work on RE technologies. The key issues for increasing energy production from RE sources relate to cost-competitiveness of RE technologies and their integration into existing energy system.

There are several definitions to renewable energy, but the main idea is that the energy source will not run out when used at a certain rate. Indeed, as MacCready (2004, 229) states, all currently used energy sources that are derived from the sun are renewable. But on human scale, it is not possible to wait millions of years for fossil fuels to be regenerated. Burning wood and biomass are renewable on human timescale, but still there is a possibility to consume it too fast and make it run out. Hydro, wind and solar are truly renewable energy sources on any time-scale. Wahlström et al. (1992, 16) have estimated that the rate at which modern man uses the fossil fuel resources exceeds the natural regeneration rate 600 000 times. This is the fundamental and principal difficulty of the energy industry currently prevailing (Kovács & Vignais 2001).

There are still many prejudices about RE technologies. The most common of them are that RE technologies will be an economical burden or they do not work efficiently in practical applications. In the long run, it is feasible to use RE sources in both technical and economical sense. In fact, the RE technologies are so well developed, economical and reliable, that the transition to a sustainable energy future should be the most important goal of governments and the world community. (El Bassam & Maegaard 2004)

2.2 Transportation and GHG emissions

> ”The car has come to be widely perceived in modern societies as a mean of freedom, mobility and safety, a symbol of personal status and identity, and as one of the most important products in the industrial economy.” (Intergovernmental Panel on Climate Change 2001, 331)

As defined above by IPCC, the energy consuming apparatus moving people from place to place is here to stay. Currently, this is not done without significant contribution to global GHG emissions. According to Laurikko (2001), transport is responsible for some 30

% of global CO_2 emissions. Also global transport sector, including air traffic, has been estimated to be the fastest growing source of CO_2 emissions (Kara 2001, 11). The reason for high amount of emissions is that the clear majority of all fuels used for transportation purposes are derived from fossil energy sources. Sperling and Cannon (2004a, 1) states that half of all oil used worldwide is for transportation. Due to the rapid growth in the sector, also the oil consumption is inevitably increasing. This has been estimated to be the main contributor for the increases in global oil consumption (Tuhkanen et al. 2001, 49). Modern road traffic has also negative environmental impacts on local and regional air quality, which are mainly due to exhaust gases containing NOx, CO and partially burnt hydrocarbons HC (Fergusson, Haines & Skinner 2005, 375).

Little over hundred years ago, when oil was found economically feasible and abundant source of energy for transportation, the growth of oil use in the sector has been exponential. For example, according Jakle and Sculle (1994), in the 1970s only in the U.S., the amount of gasoline fuelling stations had increased to 225,000 pcs (Kurani, Turrentine, Heffner & Congleton 2004, 40). Today, every minute 100 new cars or trucks are built worldwide and nearly every one equipped with petroleum fuelled ICE (Sperling & Cannon 2004a, 4). Rapid growth is expected to continue, since many highly populated countries such as India and China, are just getting ready for the culture of private cars (Bekkeheien, Øystein, Klovening & Stokholm 1999, 99).

In Europe, the European Commission expects CO_2 emissions from transport to rise to around 1113 million tonnes by 2010, which is 50% more than in 1990. The main contributor to this is the road transport, which accounts for 84 % of transport-related carbon dioxide emissions. (EU 2003) At the moment the CO_2 emissions of an average vehicle in EU are 160 gCO_2/km. A legislative limit of 130 gCO_2/km is on the planning state (Yläjärvi 2007, 18).

Also in Finland, all modes of transport, especially passenger transport by car and goods transport by road, are expected to continue their growth until at least 2030 (Lehtilä 2001, 198) and the energy consumption of the transport sector, according to Laurikko (2001), is predicted to grow 30 – 60 by the year 2020 (Pöllänen, Kallberg, Kalenoja & Mäntynen 2006, 18). According to the Finnish Vehicle Administration (AKE), in the end of 2006, there were nearly 2.5 million passenger vehicles (Norrman, S. e-mail 9th March 2007). Finland has one of the highest ratios of private cars per inhabitants of the IEA countries, which is mainly due to the size of the country and low population density (IEA 1999, 29).

The transport sector accounted for 17 % of Finland’s energy consumption in 2005, and of all CO_2 emissions produced in Finland in 2004, transportation accounted for 22 % (Pöllänen et al. 2006, 18). Passenger vehicles account for nearly 50 % of traffic–related and about 10% of total CO_2 emissions (Lehtilä 2001, 206). Also in Finland, the transport sector was undoubtedly the greatest consumer of oil leaving behind among others the industrial and residential sector (IEA 1999, 48).

2.3 The search for an alternative fuel

2.3.1 Historical view on fuel changes

Taking a look at the facts presented above, it seems obvious that something has to be done to cut emission levels of transport. The search for an alternative fuel and also for improved conventional technologies is getting more intense. Last major fuel chance in transport sector was the introduction of gasoline vehicle more than a hundred years ago. In the time before that the main travelling method was a horse ride. For example, in 1900, there were 20 million horses used for transportation in the United States, which at that time exceeded the number of motorized vehicles by a factor of 10,000. The change to gasoline ICE vehicles demanded huge changes in many fields, e.g. in agriculture, since roughly one-quarter of all U.S. cropland was used to grow hay for horses. (Sperling & Cannon 2004a, 3.) Ever since, the growth of petroleum use has been exponential. According to Jakle and Skulle (1994) only in the United States the number of gasoline refuelling stations had increased to 225,000 pcs in 1970s (Kurani et al. 2004, 40). The reason for this major change of fuel culture was the easily exploitable energy content of oil, its abundance and low price at the time. There are also other motives that have resulted in fuel changes in history. For example, in Finland during the period of 1939-1944, over 43 000 vehicles were converted to use pyrolyzed wood as a fuel (Hoffman & Kara 2004, 34). The motive for this was the shortage of oil supplies due to the Second World War.

2.3.2 Improvements of internal combustion engine

The first serious sign to put oil consumption under question was the oil crisis in the 1970s. As a result, overall fuel efficiency of vehicles begun to improve and new technologies to bring down the emissions from gasoline burning ICEs were springing up. According to Mendler (1992), these techniques include among others advanced transmission, variable valve control, stoichiometric fuel-air mixtures and engine heat retention (Bekkeheien et al. 1999, 102). From the mid-1970s, according to Bekkeheien et al., fuel efficiency of an average European car had improved by approximately 30 % by the change of the millennium. The emissions of gasoline vehicles have been minimized by using e.g. the three-way converter that reduces all three major exhaust pollutants, which include CO, HC and NO_x (Laurikko 2001, 180). In addition, the emissions can be cut by optimizing the burning process, but the inevitable truth is that even the perfect optimization and technology can not prevent CO_2 emissions as long as the fuel contains any carbon. There has been also reverse cultural development according to transport emissions. There is a common trend among people in the western world to buy larger cars with more and more energy consuming accessories. This does not help the situation with GHG emissions. According to Bekkeheien et al. (1999, 98), this trend towards ever-larger cars cancelled the effect of energy efficiency improvements in seven European countries during the nineties.

2.3.3 Competitors to the present ICE technology

The main competitors to the present ICE technology are different hybrid vehicles and later fuel cell vehicles FCVs (Kara 2001, 11). According to Chalk and Miller (2006, p.73), the transition to hydrogen-powered fuel cell vehicles will occur during the next 10-15 years. Others believe that hydrogen FCVs should be viewed as technologies after 2030 (Romm 2004b). In the meantime, hybrid electric vehicles HEVs, will increase rapidly. HEVs, like Toyota Prius, combines electric traction motor and ICE engine to improve the overall fuel efficiency. This drive train choice is not new, even though it has recently gained such attention. A patent for a petrol-electric hybrid vehicle was filed already in 1905 (Why the future is hybrid 2004, 2), but then conventional ICE ruled over for reasons other than environmental. Currently, the emissions of Toyota Prius are 104 gCO_2/km (Yläjärvi 2007, 18).

The Alternative Motor Fuel Act of 1988 in the U.S. was one of the first laws addressed for alternative fuel development (Sperling & Cannon 2004a, 15). Today, Japan is a country with serious intentions to move away from petroleum fuelled transport. Ministry of Economy, Trade and Industry has announced a plan to have 5 million fuel cell vehicles in 2020. EU has targeted, that 15 % of the total on-road fleet would be hydrogen fuelled vehicles by 2030. (Solomon & Banerjee 2004, 791.) In Finland, the growing interest on alternative fuels is clearly seen from the statistics. According to AKE, the number of passenger vehicles with a fuel or propulsion technology classified as "unknown", increased +523 % in one year between 2005 and 2006 (Norrman, S. e-mail 9.3.2007). The vehicles categorized to this category mainly include different gas fuelled vehicles. (Malin, J., AKE, phone conversation 14th March 2007).

Today, in addition to different hybrids and hydrogen fuelled FCVs, other alternatives to conventional petrol and diesel cars include liquefied petroleum gas LPG, compressed natural gas CNG, biofuels, bioethanol, electricity stored in batteries (Fergusson et al. 2005, 376), methanol and numerous mixtures of different gases. An example of such gas mixture is called Hythane, or HCNG, which is a compressed mixture of hydrogen and natural gas (Alcorn 2007).

However, in the near future alternative fuels are inevitably increasing. There will be many different fuel and propulsion combination available on commercial scale before entering to large scale RE-based hydrogen transportation, which is considered as the ultimate goal by many researchers.

2.4 Hydrogen economy

> "Electricity is the great energy carrier of the twentieth century. Hydrogen will emerge as the great energy carrier, alongside electricity, in the twenty-first century" (Scott 2004, 21).

The term "hydrogen economy" was used already in 1970 by electrochemist John O'M Bockris (National Hydrogen Association 2007b) and since then it has been under intensive research. Almost all industrialized countries have launched intensive research programmes on hydrogen technologies. Among these countries, Iceland has set itself to a role of a pioneer. It has announced an ambitious plan to transfer to a hydrogen energy economy by 2030 (Solomon & Banerjee 2004, 791). Building a hydrogen economy, three major issues to be addressed are public health, global warming and energy security (DeCicco 2004, 213-223). The interest on hydrogen is increased by the fact that it is appropriate fuel for vast variety of end uses, from the production of heat in buildings to power generation in transport applications (Hart 2004, 231). Hydrogen can also be transferred via pipelines and converted to electricity with good efficiency. Hydrogen energy development is strongly connected to the use of renewable energy sources, since without the use of RE sources hydrogen economy is not any less polluting than current energy system. Development of fuel cell technology is another key factor in promoting the use of hydrogen.

Hydrogen is not an energy source. It is a medium for storing the energy produced from primary energy sources. This makes hydrogen and electricity very similar concerning the energy transfer and storage. They can both be regarded as an energy carriers of truly clean and flexible energy system, since they itself are independent from primary energy sources. They both can be produced from various different common materials, including water, natural gas and coal (Sperling & Cannon 2004a, 2), and both can be converted to each other. Main difference is that electricity can transport energy without transferring material, unlike hydrogen. Hydrogen is now in the same position than electricity a little over century ago, when it replaced the direct use of the power from a steam engine. (Scott 2004, 23) However, hydrogen does not replace electricity in the future, but they will work together in some kind of synergy. Electricity will be converted to hydrogen when energy storage is needed and hydrogen will be converted back to electricity when e.g. a fuel cell vehicle needs power to its traction motor (Sperling & Cannon 2004a, 8). The obstacles that hydrogen energy transition is facing include lack of infrastructure, high cost, lack of vehicles to operate on the fuel and prejudices about safety and reliability issues (McNutt & Rodgers 2004, 166). The lack of infrastructure and vehicles pose a classic chicken or egg dilemma, which all new fuel innovations have to face. As DeCicco (2004, 213) has stated: “What comes first, the vehicle or the fuel?”

3 HYDROGEN AS A FUEL

3.1 Properties of hydrogen

Hydrogen is the most abundant element in the universe. Once harnessed, it has numerous advantages as a fuel. In this chapter properties and availability of hydrogen are discussed, including some safety issues and comparison to other common fuels.

3.1.1 The brief history of hydrogen

The first record of hydrogen production was done by physician Paracelsus in the 15th century. As a distinct element it was discovered in 1766 by British scientist Henry Cavendish. (Dunn 2004, 241.) Later, in 1788, the newly found element was named after the Greek words ´hydro’ and ‘genes’, meaning “water” and “born of”. In 1800, it was discovered that applying electric current to water, hydrogen and oxygen could be produced. The process was later called as electrolysis. (National Hydrogen Association 2007b.) After the manufacturing process had been discovered, pure hydrogen was used for fuel in the first modifications of ICE engine in 1807 (Cleveland 2004, 658). The vast interest on hydrogen technologies was initiated by the First World War, mainly due to uncertainties in access to the conventional energy supplies (Dunn 2004, 242). Currently hydrogen is used mainly in industrial processes. In 2004, the industrial use of hydrogen corresponded 3% of worlds primary energy consumption. The main consumers of hydrogen are chemical, metal and oil refining industries. (Helynen et al. 2004, 304.)

3.1.2 Hydrogen as an element

It is estimated that four billion years ago, hydrogen dominated the Earth’s early atmosphere. Today the atmosphere on Earth is an oxidizing one in which oxygen predominates. This is a result of a global redox cycle, where water was split into hydrogen and oxygen and hydrogen atoms escaped into space. Chemically speaking, hydrogen is strong reducing agent, which means that in proper conditions it reacts eagerly with oxygen releasing energy. (Cammack 2001, 21.)

Hydrogen accounts for approximately 75 % of the entire universe by weight. Of all molecules in the universe hydrogen accounts for more than 90 %, making it clearly the most abundant element in the universe. (Dunn 2004, 241.) It is also the element with the lowest atomic weight (table 1), consisting only one proton and one electron. Pure hydrogen at the standard temperature and pressure is odourless, colourless, tasteless and non-poisonous gas. Despite its simplicity and abundance, hydrogen does not occur naturally in its elemental form. It readily combines with other elements, such as oxygen to form water, which covers 70 % of Earth's surface (Manahan 1994, 5), and hydrocarbons that make up the majority of our present day fuels. One cubic meter of water contains 111 kg of hydrogen, which means that in theory the H_2 reserves are limitless (Mikkonen 2003, 62).

Table 1. Selected properties of hydrogen. The low-temperature properties all refer to hydrogen which has the parahydrogen concentration corresponding to its low-temperature equilibrium value (McGraw-Hill 1992, 582).

Property	Value
Atomic weight	1.00797
Molecular weight	2.01594
Melting point	-259.34 °C
Boiling point at 1 atm	-252.87 °C
Density of gas at 0 °C and 1 atm	0.08987 kg/m^3
Density of liquid at -252,87 °C	70.8 kg/m^3
Density of solid at -259,34 °C	85.8 kg/m^3
Critical temperature	-240.17 °C
Critical pressure	12.8 atm
Critical density	0.0312 g/cm^3
Specific heat at constant pressure and 25 °C	3.42 cal/(g)(°C)
Thermal conductivity at 25 °C	0.000444 cal/(cm)(s)(°C)
Viscosity at 25 °C	0.00892 centipoise
Specific gravity	0.0695 compared to air

3.1.3 Energy content and benefits of H_2

In a way, hydrogen has been used as a fuel since the dawn of history, not in its elemental form, but combined in other fuels, such as wood, coal, oil and natural gas. When considering the development of fuel use through time, the hydrogen content in the predominant fuel has been increasing as the technology had evolved (figure 1). According to this fact, pure hydrogen seems to be the fuel of the future.

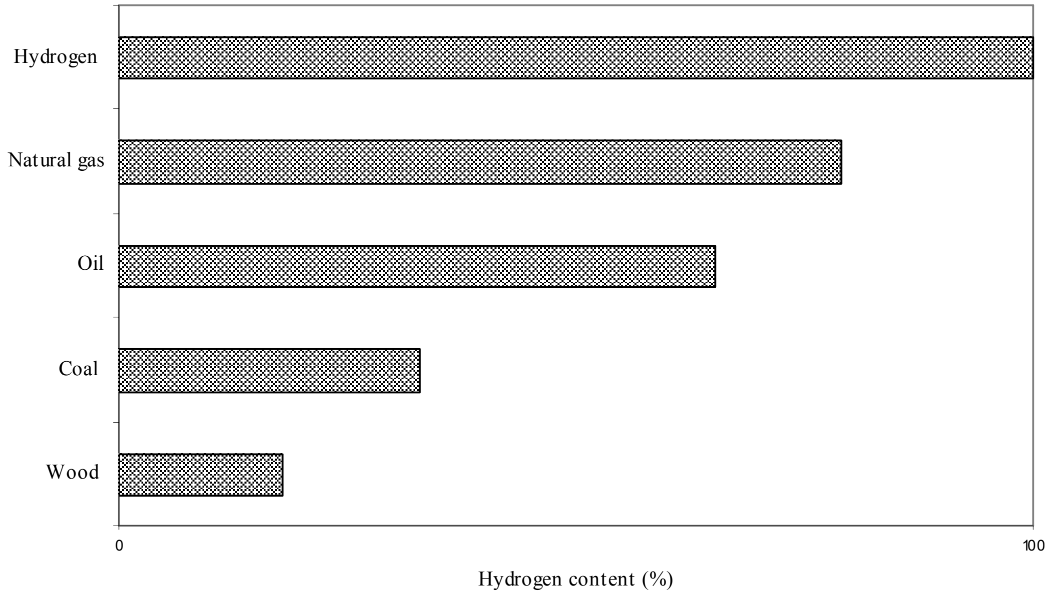

Figure 1. Hydrogen content in common fuels. As technology had evolved hydrogen content in predominant fuel has been increasing. (Cook 2001, 2)

Hydrogen is flexible and rather easy to store, which make it suitable fuel for many applications. It can either be burned as a substitute for gaseous fossil fuels or be converted directly into electricity by a fuel cell in an electrochemical process that exceeds the efficiency of conventional electricity generation.

Hydrogen has the highest energy density per unit weight than any other chemical fuel. The energy content of H_2 is around 120 MJ/kg (table 2), which is three times higher than that of gasoline (43 MJ/kg). For comparison, coal has the energy content of 27,9 MJ/kg (Kara et al. 2004, 380), which means that hydrogen contains more than four times more energy by unit weight than coal. When comparing gravimetric energy densities of all common fuels, hydrogen rises to number one position, but it ranks last when the energy density by volume is considered. This is due to the minimal atomic weight of hydrogen. When most of the other fuels like gasoline, diesel and ethanol are liquids at ambient temperatures, bimolecular hydrogen is in gaseous form. For example, at STP hydrogen takes up some 2800 times more space than gasoline containing an equivalent amount of energy.

Table 2. Heating values of different transport fuels (McGraw-Hill 1992, 582; El Bassam & Maegaard 2004, 190-191; Steinberger-Wilckens 2005, 506).

Property	Hydrogen	Methane	Gasoline
Density (kg/Nm3)	0.08987 *	0.6512	749
LHV (MJ/kg)	119.88	50.04	43.04
HHV (MJ/kg)	141.91	55.5	45.7

* Density of gas at 0 °C and 1 atm

Compared with natural gas, the next lightest gaseous fuel relevant in power production, hydrogen has a volumetric heating value of roughly three times smaller than that of natural gas. In gravimetric comparison, hydrogen contains energy more than twice as much than natural gas.

When used in a low temperature FC system and the output water is in liquid form, the higher heating value HHV of hydrogen, which is 142 MJ/kg, can be utilized. In this case, 1 kg of hydrogen accounts for 4,3 liters of gasoline (Ronkainen et al. 2005, 6). The interest about hydrogen is based mainly on its use in FCs, but also its utilization as an ICE fuel is considered promising. In both end-use technologies, FC systems and combustion, the main benefit of using hydrogen as a fuel is its friendliness to the environment. It burns cleanly and with high temperatures. For example, when burned in pure oxygen, the temperature of 2700 °C can be measured (Arvonen & Levonen 1989, 124). And since the fuel contains no carbon, there are no resulting CO, CO_2 and HC emissions, only by-products being heat and water.

When hydrogen is burned with air, which is about 68 % of nitrogen, some NO_x compounds are formed. The amount of nitrogen oxide emissions is still significantly smaller than in fossil fuel combustion (Mikkonen 2003, 62). By using e.g. PEMFCs and pure hydrogen as fuel, NO_x emissions can be eliminated due to the low operating temperature (Laurikko 2001, 187).

In addition to the benefits mentioned in the chapter 2.4, hydrogen as a fuel possesses advantages such as (Steinberger-Wilckens 2005, 505; Mikkonen 2003, 62):

- high energy density by weight
- negligible emissions if produced by using RE sources

- it can be sold outside electricity market as industrial gas
- it has ignition energy less than 10 % of the value for hydrocarbon fuels, which is a benefit in catalytic combustion but also requires some safety concerns
- it can be blended with natural gas to 15 – 20 % mixture without a need to modify the burners
- it can be used as an energy carrier in places where electric cables are undesirable.

Disadvantages of hydrogen are the difficulty to store it in a vehicular application due to its low energy density by volume. For example, according to liquid storing methods for vehicular fuels, hydrogen is far more difficult to liquefy than e.g. natural gas and it possesses lower energy density (Laurikko 2001, 184). Hydrogen also causes problems with ferric materials, causing embrittlement of the material (Steinberger-Wilckens 2005, 507).

3.1.4 Combustion characteristics of H_2 and safety issues

The wide self ignition limits of hydrogen-air mixtures create a safety concern to be taken care of. Anyway, the hazards associated with the use of hydrogen are not greater than those with conventional fuels (Steinberger-Wilckens 2005, 507). Due to its low weight, hydrogen disperses in the atmosphere with high diffusivity, which reduces the risk of explosion. Even in the case of explosion, hydrogen has considerably lower explosion energy than e.g. methane CH_4 and propane C_3H_8. Some characteristics relating to ignition and explosion hazards of different fuels are presented in the table 3.

What comes to hydrogen, there is one safety concern less than with the conventional fuels. Gasoline and oil are toxic and poisonous to human and wildlife when released into the environment. Hydrogen spills would evaporate almost immediately leaving only water vapour to the atmosphere. Conventional fossil fuels like heavy oil and gasoline, on the other hand, require immense cleanup efforts with the result being that most of the spilled gasoline or oil seeps into the surrounding ecosystem causing severe harm. It is true that hydrogen possesses wide range of ignition limit with air, but the minimum concentration which is needed for ignition, is less than that of gasoline. Even though hydrogen does not warn its user of possible leaks with a smell like gasoline and other hydrocarbon fuels.

Table 3. Properties of hydrogen, NG and gasoline fuels relating to combustion and safety issues (IEA Greenhouse Gas R&D Programme 2007, 33; McGraw-Hill 1992, 582; Laurikko 2001, 184)

Property	Hydrogen	Natural gas	Gasoline
Boiling Point (°C)	252.9	-161.0	27 - 225
Self ignition point (°C)	585.0	540.0	228 - 501
Flame temperature (°C)	2045	1875	2200
Limits of flammability in air (vol-%)	4 - 75	5.3 - 15	1.0 – 7.6
Minimum ignition energy (μJ)	20.0	290.0	240.0
Limits of detonation in air (vol-%)	13 - 65	6.3 – 13.5	1.1 – 3.3
Explosive energy (kg TNT/m^3 gas)	2.02	7.03	44.22
Diffusion coefficient in air (cm^2/s)	0.61	0.16	0.05

Safety risks followed by a fuel leakage are very different depending on the fuel in question. While gasoline or propane is leaking from their container, there is a possibility for them to congregate near the ground increasing the risk of explosion. In the case of hydrogen leak, the safety risk may occur when hydrogen is accumulating to unventilated spaces in the upper parts of the vehicle or room. The safe handling of hydrogen is studied widely and there are numerous technologies to improve it, e.g. the odorants added to H_2 gas to help the users to detect the leaks (Kopasz 2006). Also storage methods with minimal risk of explosion are available on the market, such as various types of metal hydrides.

Despite the decades of safe use of hydrogen, there can be found some prejudices about the hydrogen derived from the past. The two most famous ones are the hydrogen bomb and the Hinderburg disaster. Cammack and Rao (2001, 221) have titled this old belief as the “Hinderburg disaster syndrome”.

3.2 Electrolysis as hydrogen production method

3.2.1 Hydrogen production at present

Hydrogen can be produced from primary energy sources in many ways. Either directly or indirectly, most of the hydrogen at present is produced industrially by conversion of fossil fuels (Cammack 2001, 202). The most important or at least the most common way to produce hydrogen is steam reforming of natural gas. Other production methods include electrolysis and gasification of solid fuels, such as coal, biomass or heavy fuel oil. Electrolysis is considered to be the ideal production technology, but the major disadvantage of this method is its cost. In 2003, the cost of electrolysis was 3-4 times higher than in steam reforming. (Mattila et al. 2001, 122.)

According to U.S. Department of Energy, annual rate of global hydrogen production is approximately 500 billions of Nm^3/year, of which 48 % is derived from natural gas and only 4 % is produced by electrolysis (Romm 2004a, 72). In the United States only, natural gas is the source of 95 % of the hydrogen produced. According to Romm, hydrogen demand has been growing 10 - 20 % per year since 1990s, with the biggest contributor to the increasing demand being the oil refining industry. (Romm 2004b; 2004a, 72.)

3.2.2 Principles and the efficiency of electrolysis

Electrolysis is the technical name for a process in which electricity is used to split water into its constituent elements, hydrogen and oxygen. The splitting of water is done by passing an electric current through water. The water electrolysis is thus the exact reverse of a hydrogen fuel cell. Electrolysis technology can be implemented at any scale whenever there is a supply of electricity, and it can be easily utilized close to the points of demand. (Newborough 2004.)

For the efficiencies of the electrolysers, many different figures can be found from literature depending on how these efficiencies are being calculated. Roy, Watson and Infield (2006) states that there is five different descriptions for the electrolyser efficiency, such as stack efficiency, voltage efficiency, overall efficiency, energy efficiency and finally, water to hydrogen conversion efficiency. Newborough (2004, 2) states, that modern electrolysers can reach conversion efficiency up to 90 %. According to Steinberger-Wilckens (2005, 507), If LHV is used for the calculation, the efficiency is 65 %, which means that the electrolytic production of 1 Nm^3 of hydrogen at atmospheric pressure would take 4.2 – 4.8 kWh of energy. This efficiency can be raised to 85 - 90 % by using high-temperature and pressurised electrolysis as well as advanced materials (Steinberger-Wilckens 2005, 507). Romm (2004b, 76) calculated that typical commercial electrolysers with an energy efficiency of 70 % require about 50 kWh of energy per 1 kg H_2. Latest development on electrocatalysis has resulted in even more efficient systems. Marshall, Børresen, Hagen, Tsypkin and Tunold (2006, 436) reported that an energy consumption of 3.75 kWh Nm^{-3} H_2 at 1 A cm^{-2} and an efficiency of 94,4 % is being obtained.

The polluting level and CO_2 emissions coming from the activity depend on the origin of the electricity used for the process. Electrolysers have small but increasing share in the market for the same reason as it is for the fuel cells i.e. the cost. Schroeder (2004, 121) lists three benefits of the electrolysis of water. First thing is the purity of electrolytic hydrogen, which is the best fuel for fuel cells. Another reason is that the electrolysis technology is relatively mature and therefore unexpected problems are not likely to happen. Third benefit, which is also stated by many other researchers, is that the electrolytic hydrogen production is the only practical way to produce hydrogen from renewable energy sources.

Three types of electrolysers can be listed: Alkaline, proton exchange membrane PEM and the solid oxide electrolysers (Newborough 2004, 7). The latter is sometimes called as high temperature electrolysis (Mikkonen 2003, 64), due to its operating temperature of 800-1000 °C. The PEM electrolysers, which are discussed in more detail in the next chapter, are well suited for distributed H_2 generation and their functioning is well proven with numerous existing applications (Newborough 2004, 7).

3.2.3 PEM electrolysers

Proton exchange membrane (PEM) electrolysis (figure 2) has been successfully used since 1970s in submarine and spacecraft applications to generate oxygen and hydrogen (Schroeder 2004, 121). Greater energy efficiency, higher production rates, and more compact design that PEM electrolysers offer, are the advantages over the traditional technologies (Marshall et al. 2006, 432).

The PEM water electrolysis cell consists of a polymer layer on which the anode and cathode are bonded. The polymer layer acts as an electrolyte and a gas separating membrane. The electrodes are normally composites of electrocatalytic particles. On anode and cathode, different electrocatalysts can be used, such as IrO_2 at anode and Pt at cathode. Hydrogen is produced by supplying water to the anode where it is decomposed into oxygen, protons and electrons. The protons are transported through the membrane to the cathode while the electrons exit the cell via the external circuit, which supplies the driving force for the reaction. At cathode the electrons and protons are combined and high purity hydrogen is formed. (Marshall et al. 2006) The purity of the hydrogen produced is up to

99.999 % (Barbir 2004, 668). Reactions in the PEM electrolysis cell are the following (Marshall et al 2007):

Anode: $2\ H_2O \rightarrow 4\ H^+ + O_2 + 4e^-$

Cathode: $4\ H^+ + 4e^- \rightarrow 2\ H_2$

Cell: $2\ H_2O \rightarrow 2\ H_2 + O_2$

The voltage applied must be DC voltage and higher than a thermoneutral voltage 1,482 V. Similarly to fuel cells, individual electrolyser cells may be constructed into a stack in order to get the desired output at a reasonable stack voltage. Other equipment required for electrolyser include power supply regulator, water supply system, water circulation pump, water-gas separators, heat exchanger, controls and safety instrumentation. (Barbir 2004, 662.) Water fed to the anode must be desalinated and demineralised (Steinberger-Wilckens 2005, 507). Theoretically from 1 litre of water 1.24 Nm^3 of hydrogen can be produced. According to Barbir (2004, 667) actual water consumption is about 25 % higher, since both gases leave the electrolyser wet and some water is lost due to oxygen exhaust and periodic hydrogen purge.

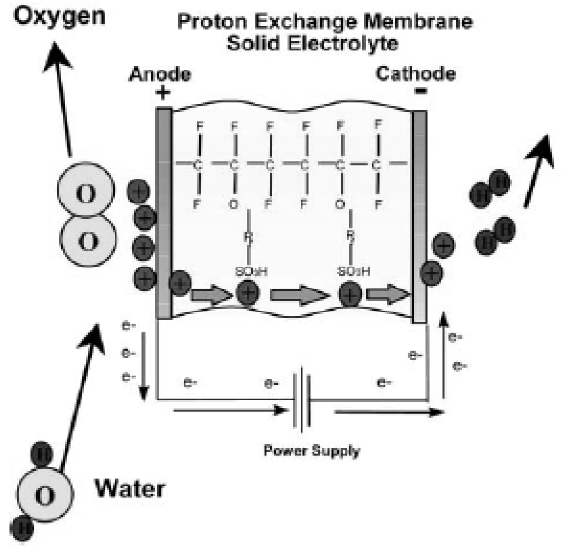

Figure 2. Schematic representation of a PEM electrolysis (Barbir 2004, 662)

PEM electrolysers are kept as a promising technology for clean hydrogen production. In addition to their other benefits mentioned before is that they can cope with transient variations in electrical power input, which is a major benefit when utilizing intermittent renewable electricity supplies (Newborough 2004, 8).

4 HYDROGEN IN TRANSPORT APPLICATIONS

4.1 Metal hydride storage of hydrogen

In this chapter, the on-board storage methods of hydrogen are discussed concentrating on the metal hydride storage. The general requirements for an on-board fuel and a comparison of different storage options are also discussed briefly.

4.1.1 Different hydrogen storage technologies

Hydrogen can be stored by using a variety of technologies. These technologies include compressed H_2 gas, cryogenic containers, metal hydrides (MHs), chemical hydrides (Christodoulou 2003), carbon nanotubes, glass microspheres (El Bassam & Maegard 2004, 172), charcoal absorption, zeolith storage and fullerenes (Mikkonen 2003, 72). Every method has its advantages and weak points, such as low temperature needed for liquid storage and safety risks occurring in high pressure gas storage (Steinberger-Wilckens 2005, 508). Solid state hydrogen storage, such as metal hydrides, carbon based storage and glass microspheres are safer and have important potential considering the on-board storage alternatives.

4.1.2 Requirements for an on-board fuel storage system

The main criteria for any fuel for vehicular applications are safety, efficiency and reasonable energy density by volume or weight. The potential properties of hydrogen as a fuel were introduced in chapter 3, but to store it in a moving vehicle and meet the driving ranges and other performance criteria of current fossil fuel based ICEs is not a simple case. H_2 storage issue is considered to be the key in achieving market success for FCVs.

As Chalk and Miller (2006, 79) defined, energy density is critical matter considering the driving range and other performance criteria of an H_2 vehicle. The energy density of current fuel systems is about 6 kWh l^{-1}, which means that the requirement for a FCV with improved fuel economy and comfortable storage system would be 2.7 kWh l^{-1}. Considering the energy density by weight, the requirement would be 3.0 kWh kg^{-1}, and this must be met with reasonable cost. Currently, there is no hydrogen storage technology that meets this requirement. New storage materials are needed, and currently the focus is on hydride and carbon-based materials. (Chalk & Miller 2006, 79.) Ross (2006, 1084-1085) defines additional targets for the hydrogen fuelled vehicle and its storage method. The most significant of those targets is that the full tank system should possess 6 % gravimetric hydrogen density and the system recharge time should be less than 5 minutes. In addition, the H_2 should be available at an overpressure of few bars and without a need to heat the container to 50 °C or higher.

4.1.3 Metal hydride mechanisms and materials

There are several metal hydride compounds that are considered promising in solving the problem of H_2 storage on-board. In MH storage, hydrogen is chemically bonded to one or more metals and can be released through heating or a catalyzed reaction (Romm 2004a, 97). To form a metal hydride, hydrogen splits into atoms at the surface of the metal and then enters the metallic lattice in atomic form. After that, hydrogen atom diffuses through the metal moving from interstitial site to another, and finally forms a hydride phase with a more or less ordered hydrogen sub-lattice. The metal hydrides such as lanthanum nickel hydride ($LaNi_5H_6$) were first studied in 1970s by Buschow and Miedema. (Ross 2006, 1085)

There are two possible ways of hydriding a metal. Those are direct dissociative chemisorption (1) and electrochemical splitting of water (2). The reactions are the following (Sakintuna, Lamari-Darkrim, Hirscher & Dogan 2006, 2):

$$M + \frac{x}{2} H_2 \rightarrow MH_x \tag{1}$$

$$M + \frac{x}{2} H_2O + \frac{x}{2} e^- \rightarrow MH_x + \frac{x}{2} OH^- \tag{2}$$

Metal and hydrogen usually form two different kinds of hydrides, α-phase and β-phase. At α-phase only some hydrogen is absorbed and at β-phase hydride is fully formed. The ability of different metals to dissociate hydrogen depends on surface structure, morphology and purity. An optimum storage material is required to have a long list of favourable properties which include (Sakintuna et al. 2006, 3):

- high hydrogen capacity per unit mass and per unit volume
- low dissociation temperature
- moderate dissociation pressure

- low heat of formation to minimize the energy necessary for hydrogen release
- low heat dissipation
- reversibility
- limited energy loss during charge and discharge
- fast kinetics
- high stability
- cyclibility
- low cost and high safety.

Recently, lots of research is done to develop hydride materials towards the optimum. $LaNi_5H_6$ hydrides mentioned earlier are not very promising alternatives to provide a substitute for petrol and diesel since all AB_5 compounds, in which $LaNi_5H_6$ also belongs, can provide only about 1 % hydrogen by mass (Ross 2006, 1085). The aim of the developers is therefore to find lighter metal lattices that will work as well as these AB_5 materials while satisfying the gravimetric efficiency requirement. The metals, which are especially interesting due to their light weight, include Li, Be, Na, Mg, B and Al. They all form a variety of metal-hydrogen compounds, including lithium imides, sodium alanates and borohydrides. Another benefit these metals possess is that they are capable of binding a good number of hydrogen atoms compared to metal atoms, in many cases at the order of $H/M = 2$.

Magnesium hydrides are promising candidates for on-board hydrogen storage material due to their high storing capacity by weight and low cost. Mg-based hydrides also possess good functional properties, such as heat-resistance, vibration absorbing, good reversibility and recyclability. The main disadvantages of some Mg-based hydrides are slow kinetics and high hydrogen desorption temperature up to about 300 °C. Reasonably, these facts reduce their efficiency and applicability in vehicular use. (Sakintuna et al. 2006, 3) Progress has been made also with other materials than light metals. For example, an organometallic compound made from rhodium and hydrogen, recently introduced by Dr. Weller at Bath University, can store and release hydrogen at room temperature (Storage metal reduces hydrogen car problems 2006, 50).

Solid hydrides have certain advantages, but metal hydrides can also be stored in a water based solutions (Romm 2004a, 97). The example of a successful use of water based metal hydride storage is a process demonstrated by McClaine, Tullman and Brown (2005) where they utilized MgH_2 slurry and water. In this process MgH_2 was hydrolyzed to $Mg(OH)_2$ and hydrogen. (Saxena, Drozd & Durygin 2006, 3.)

All hydrides can be classified either reversible or irreversible. Reversible hydrides are generally solid alloys or intermetallic compounds that can release hydrogen under specific pressures and temperatures. Reversible hydrides can be replenished again by adding pure hydrogen at a filling station. Irreversible hydrides undergo certain chemical reactions to form by-products, and they cannot be replenished without chemical processing. (Romm 2004a, 98.)

4.1.4 The storing capacity and durability of a metal hydride storage

The storage density of MHs has been studied a lot and a wide range of storing capacities can be found in the literature. What makes the analysis more complex is that the figures are sometimes given in gravimetric densities, and sometimes volumetric energy density is used. In 1995 Hagström, Lund and Vanhanen studied the applicability of six hydrogen absorbing pseudobinary AB_5 and AB_2 alloys at near ambient temperatures, finding out the storage capacities for the alloys within the range of 1.4 – 1.5 wt%. Similar figures were found by Anttila (1993), when he measured a maximum of 1.18 wt% of hydrogen for a AB_5 compound. Steinberger-Wilckens (2005, 509) states as well that total amount of hydrogen absorbed to MHs is generally 1 – 2 %. The seemingly low figures derive themselves from the low weight of hydrogen and the high weight of metal alloys. According to Carpetis (1988) some hydrides are capable of storing 5 – 7 wt% of hydrogen but require unloading temperatures of 250 °C or higher (Steinberger-Wilckens 2005, 509). Ross (2006) reports, that certain sodium borohydride can achieve 7.5 % gravimetric yield of hydrogen. Magnesium hydride, MgH_2, is said to have the highest energy density by weight of all reversible hydrides applicaple for hydrogen storage. According to Sakintuna Sakintuna et al. (2006, 3), storing capacity of MgH_2 is 7.7 wt%. For a comparison, pure water contains 11.1 wt% of hydrogen, but its decomposition requires much thermal, electric or chemical energy. This is the reason why the figures of gravimetric hydrogen content of different materials itself does not reveal much of its usefulness.

Although the state-of-the-art materials in metal hydrides are still far away from the aimed target for the transportation application, there has been some successful development to improve the storing capacity and performance of MHs. The kinetics of MHs have been improved by adding an appropriate catalyst into the system and as well as by a process called ball-milling. Ball-milling creates fresh surfaces during MH processing and it leads to increased surface area and formation of micro- and nanostructures in the material. (Sakintuna et al. 2006, 1)

The durability of the MH storage tank is directly proportional to the purity of the hydrogen, because the hydriding process becomes less effective as the impurities are deposited in the crystal lattice (Steinberger-Wilckens 2005, 509). Impurities that are usually accumulated, by physisorption or by chemisorption, in the MH surface include H_2O, OH^-, O^{2-}, C, $-CH_2-$ and other molecules existing in the atmosphere. The surface of the MH material must therefore be activated periodically to clean the surface from impurities. Activation restores the storing capacity of the MH to a certain level, and it can be done by using e.g. heating and vacuum. (Christodoulou 2003)

4.1.5 Metal hydrides compared with other storage options

The advantages of the MH storage are high energy density by volume and safety. The volumetric energy density of MH storage is 55 – 60 kgH_2/m^3, which is even higher than that of liquefied hydrogen (Mikkonen 2003, 67). Due to the weight of the storage material, the gravimetric energy density is therefore 15 – 72 gH_2/kg (Helynen et al. 2004, 305). This would mean that storing 5 kg of H_2 would require storage tank of 70 – 330 kg depending on the hydride material. This is one of the main disadvantages of the MH storage. The discharge temperatures of 300 - 350 °C are also too high for vehicular applications in conventional MHs with high storage capacity (Cammack & Rao 2001, 225). The kinetics can also become an obstacle with the use of MHs, since some of them are slow to absorb hydrogen in fuelling while others are slow to release it during their use (Romm 2004a, 98). To improve the performance of MHs with slow discharge, a gas vessel used as buffer storage can be attached to the MH storage system (Christodoulou 2003). Different H_2 storage methods are compared in the table 4. The minimum values of H_2 content for metal hydride and carbon nanotube storage are the ones achieved in near ambient temperature and pressure and higher values correspond to situation with high temperatures and pressures (Mikkonen 2003, 74).

Having the highest gravimetric H_2 content, liquefied storage has its own disadvantages such as net losses of about 30 % due to liquefaction and inevitable boil-off rate which is approximately 1 % day^{-1}. In addition, the method becomes costly due to the maintaining the storage temperature and pressure. (Ross 2006, 1085.) The value of gravimetric density of liquefied hydrogen depends naturally on the vessel materials used. Sakintuna et al. (2006, 1085) gives a liquid storage of hydrogen a gravimetric yield of 10 wt%, which is little more pessimistic than represented by Mikkonen.

Table 4. Comparison of different hydrogen storage methods (Mikkonen 2003, Sakintuna et al. 2006).

Storage technology	Hydrogen content (wt%)
Compressed gas	11.3
Liquid	25.9
Metal hydrides	2.0 – 7.6
Carbon based storage	0.2 – 10.0
Zeolite	0.8
Glass microspheres	6.0
Fullerenes	6.0
Chemical storage	8.9 – 15.1

The same material dependence issue is naturally the case also for the gas cylinders. For example, a conventional steel hydrogen vessel achieves about 1 % gravimetric hydrogen (Ross 2006, 1085). According to volumetric density, the MHs have significant advantage compared to compressed gas storage. For example, 1 liter MH container can store 1000 liters of hydrogen under conditions of 10 bars and 25 °C. In the same conditions, the same amount of H_2 stored as a compressed gas would require a vessel of 100 liters (Christodoulou 2003). Yet the compressed gas and liquid H_2 storing methods are relatively advanced technologies, it is not expected that they will fully meet the requirements for on-board vehicular storage technology (Sakintuna et al. 2006, 1085).

4.2 Fuel cells

In this section the performance and principal functions of fuel cells are explained. The emphasis is on PEM and alkaline fuel cells. The efficiencies of ICE and fuel cells are also discussed briefly.

4.2.1 Fuel cell performance

The first fuel cell was developed by Sir William Grove on 1839, which was some 40 years before the invention of the ICE engine (El Bassam & Maegard 2004, 174). Despite the fact, a more intense development work on fuel cells started not until the late 1950s, and soon small FC systems were developed for the US Gemini and Apollo space programmes in the 1960s (Everett & Boyle 1998, 412). Since then, different FC technologies are being developed for numerous applications, often categorized as portable, transportable or stationary depending on their size and function (Mattila et al. 2001, 118). In this work, the emphasis is on the transportable (kW range) fuel cell systems.

Fuel cell is an electrochemical device that converts chemical energy directly into electricity by combining hydrogen and oxygen in a controlled system. Pathapati, Xue and Tang (2004, 5) defines a fuel cell as "fluid-solid-heat-electrochemistry coupled complex system". Basic structure of a fuel cell consists of an electrolyte layer in contact with a porous anode and cathode on either side (EG&G Technical Services, Inc. 2004). The simple reaction inside an acid electrolyte fuel cell is a reverse to the electrolysis of water (Larminie & Dicks 2003, 2):

At the anode: $2\,H_2 \rightarrow 4\,H^+ + 4\,e^-$

At the cathode: $O_2 + 4\,e^- + 4\,H^+ \rightarrow 2\,H_2O$

A gaseous fuel, hydrogen in this case, is continuously fed to the anode. Air or pure oxygen, which acts as an oxidant, is fed continuously to the cathode. Electrochemical reactions taking place at the electrodes produce the electric current. (EG&G Technical Services, Inc. 2004.) Fuel cell components and characteristics are quite similar to an ordinary battery, but in a fuel cell there is no cycle of discharge and recharge. A battery will stop producing electrical energy when the chemical reactants are consumed and it needs to be recharged again by an external energy source. Fuel cell is an energy conversion device that can theoretically produce electricity as long as the fuel and the oxidant are fed to the electrodes. Fuel cell, like any other device has a limited lifetime. Considering the FC lifetime, the aim for automotive fuel cell system is to be competitive with the current ICE engines, i.e. 5000 h lifespan under heavy load cycling. Despite the intense development work, the performance of current FC systems decreases substantially after approximately

1000 h. (Chalk & Miller 2006, 78.) The main reason for the limited lifetime of fuel cells are corrosion and malfunction of components (EG&G Technical Services, Inc. 2004). The degradation of the catalyst layers and membranes is also accelerated by the variations in cell potential and relative humidity levels (Chalk & Miller 2006, 78).

4.2.2 Advantages and disadvantages of a fuel cell

Fuel cells possess many advantages and disadvantages compared to other power sources. Forrai, Funato, Yanagita & Kato (2005) list four reasons which contribute to the global interest on fuel cell technology: Efficiency, simplicity, low emissions and silence. In general, FCs are more efficient than combustion engines and small FC systems can be just as efficient as large ones. This is important point in building small-scale and transportation FC systems. The efficiencies are discussed in more detail within the next chapter. According to Forrai et al. (2005) the simplicity of the fuel cells, the fact that in FCs there is few if any moving parts, can lead to long lifetime and reliability. Generating electricity with low emissions is one of the benefits of the fuel cells, the only by-product of hydrogen fuelled low temperature FC being water. In the case of fuel cell systems, the possible emissions from the fuel manufacturing must not be forgotten. The fourth benefit mentioned by Forrai et al. (2005) was silent operation. Because electricity is produced straight from the fuel without any mechanical conversion energy, FCs are silent compared to combustion engines. This is an important fact in automotive and in local power generation applications.

Even if it was just mentioned that the simplicity of FCs is the key to the long lifetime, there are other opinions about the fuel cell lifetime expectations found in literature. For example, Chalk & Miller (2006) have noticed that the major challenges to the FC development are their durability and cost. There are promising evaluations about the continuous increase in FC lifespan. Hottinen (2003) estimated that the FC lifetime expectation of 5000 h will not be difficult to achieve. The FC lifetime is expected to be improved by developing the durability of conventional fluorocarbon membranes. The cost issue is the other major challenge to FC commercialization, but the gap between the ICE cost and the FC system cost has been decreasing during the recent years. In 2003, the FC cost was estimated to be over ten times that of ICE (Mattila et al. 2001). Three years later, Chalk and Miller (2006, 78) estimated that the FC system would be only about a factor of five higher in cost than ICE. In this estimation, they applied cost savings for high volume manufacturing. The major contributors to the FC costs are the catalysts, membrane and the bipolar plates. In this field as well, there are promising development issues to continue the trend of FC cost reduction. One of the promising matters in this field is the development of the hydrocarbon-based membranes which could equal the fluorocarbon membrane performance at lower cost. (Chalk & Miller 2006, 78.) It must be remembered that the cost is always a problem of new innovations, for example, when T-Ford was introduced to the market it was expensive, but the cost was rapidly decreased by the mass production.

4.2.3 Efficiency of a FC system

The efficiency of a FC system is not a simple thing to define. The case is simpler in defining the maximum efficiency of e.g. a wind-driven generator or a heat engine. The maximum efficiency gained from wind generator can be estimated by using the Betz Coefficient (0.58) and the heat engine maximum efficiency can be calculated by using the Carnot limit [(T1 – T2)/T1]. Defining the maximum possible efficiency for FCs is not so clear. (Larminie & Dicks 2003, 31) Here are some equations and points to describe the efficiencies of a FC system.

In general, the efficiency of a system is defined as the energy produced by the system divided by the input energy. In this case the input energy meaning the chemical energy of the fuel integrated over time. For a fuel cell, this means that (Noponen 2003a, 3)

$$\eta_{theoretical} = \frac{\Delta G}{\Delta H}$$

where, ΔG = Gibbs free energy

ΔH = Reaction enthalpy of the fuel

For example, the theoretical efficiency for PEMFC in standard conditions (1 bar, 25 °C) is (Noponen 2003a, 3):

$$\eta_{theoretical} = \frac{\Delta G}{\Delta H} = \frac{-236kJ/mol}{-283kJ/mol} = 0{,}83$$

The efficiency limit is decreasing when the temperature increases. This could lead to conclusion that the lower operation temperatures are better for fuel cells, but the other losses occurring in the FC operation must be taken into account as well. For example, the voltage losses are nearly always less at higher temperatures. Also the waste heat is more useful when the FC is operating at higher temperature. And though it was mentioned in the previous chapter, that FCs have higher efficiency limit than heat engines, this is not always the case at higher temperatures. (Larminie & Dicks 2003, 33-34.)

In a real fuel cell system other factors affecting to the overall efficiency must be considered. The conversion efficiency is defined as the total efficiency of the fuel cell. There the theoretical, voltage and current efficiencies are taken into account to give the conversion efficiency. The voltage efficiency of the fuel cell is decreased by loss mechanisms, which include activation losses, ohmic losses and mass diffusion. The current efficiency is affected by the possible hydrogen penetration through the membrane or leakage through the gaskets. (Noponen 2003a, 3) According to Forrai et al. (2005, 674) the FC system efficiency is highly dependent on the temperature, humidity and air flow. Also to maintain an optimal operation conditions, parameter identification, diagnostics and control is required. The main losses of the hydrogen fuelled low temperature fuel cell are the activation and ohmic losses and they are normally similar in magnitude (Forrai et al. 2005, 674).

In literature, there are many researches that compare the efficiencies of the ICE and FC systems. According to Chalk and Miller (2006), fuel cells are projected to have energy efficiency twice that of ICE.

4.2.4 PEM fuel cells

The abbreviation PEM means proton exchange membrane fuel cell (Larminie & Dicks 2003, 67). In the literature this type of fuel cells are also being called as polymer electrolyte fuel cells PEFC or solid polymer fuel cells SPFC (Larminie & Dicks 2003, 67). The charge carrier in PEMFC is H^+ ion, which travels through the electrolyte during the electrochemical process (figure 3). PEMFC system operates at moderately low temperatures (approx. 80 °C) and the usual size of a PEMFC system ranges from 1 W to 10 MW (Mattila et al. 2001, 119). PEMFC system is suitable for both stationary and automotive applications, and according to Pathapati et al. (2004, 4), it is a suitable candidate as a substitute of traditional ICE power source. This is mainly due to its combination of simple design, high power-to-weight ratio and low operating temperature

(Bekkeheien et al. 1999, 103). One more reason that can be regarded as a benefit is the electrical efficiency of a PEMFC, which lies between 40 – 60 % (Kolhe 2004).

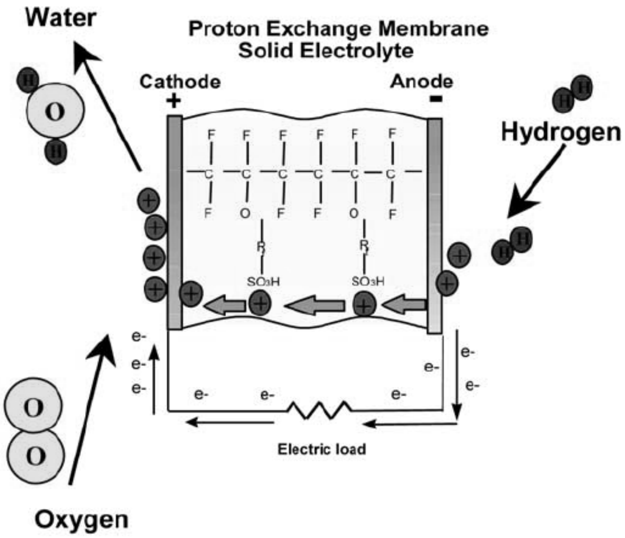

Figure 3. Schematic representation of a PEM FC (Barbir 2004, 662)

Despite all the good characteristics, PEMFCs have some disadvantages. Wang, Nehrir and Shaw (2005) have developed dynamic models for PEMFCs and have noticed that they cannot respond to electrical load transients as fast as desired, which is mainly due to their slow internal electrochemical and thermodynamic responses. Dynamic modelling of the FC systems predicts the transient phenomena in a FC system. For example, the dynamic model developed by Pathapati et al. (2004) can predict the transient response of cell voltage, temperature of the cell, hydrogen and oxygen out flow rates and cathode and anode channel temperatures and pressures under sudden changes in load current. The models are used for example to utilize the automotive FC systems in a more effective way.

The voltage of a single fuel cell is only 0.7 volts (figure 4), which is why the individual cells are connected in series to increase the voltage of the system. Typical components within the PEMFC stack include ion exchange membrane electrolyte, porous backing layer, two electrodes and a cell plate hardware that delivers the fuel and oxidant to the reactive sites. (EG&G Technical Services, Inc. 2004.) The catalyst layer i.e. the electrodes are in good contact with the membrane electrolyte, which is typically 50–175 μm thick. The heart of the cell, which is magnified in the figure 4, can be called as membrane electrode assembly MEA. (Gottesfeld et al, 2004.) The humidification level of the MEA is a key factor that affects mass diffusion and proton transportation between the electrodes. Too much water leads to the flooding of the MEA and too low humidification level leads to higher internal resistance. Final humidification level depends on the fuel cell temperature, and it can be controlled by humidifying the inlet fuel and air. (Pathapati 2004, 5)

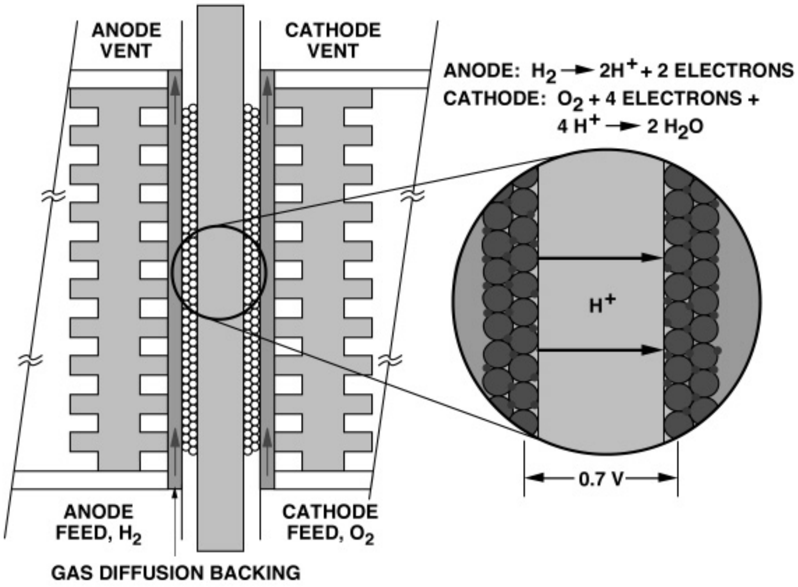

Figure 4. Cross section of PEM fuel cell. (Gottesfeld 2004)

4.2.5 Alkaline fuel cells

The development of alkaline fuel cells (AFCs) started in 1930s and by 1952 the first tests on a 5 kW alkaline fuel cell was completed by F.T. Bacon. In this fuel cell, 45 % KOH was used as an electrolyte. (EG&G Technical Services, Inc. 2004) The early development of AFC was related to space technology but nowadays the emphasis is on the small off-road vehicular and stationary applications (Lin, Kirk & Thorpe 2006, 475). In road transport applications AFC have lost their popularity to PEMFC solutions. The benefits of AFC compared to the PEMFC are smaller activation losses and the absence of costly noble metal catalyst platinum in its electrodes (Mikkonen 2003, 44). Lin et al. (2006, 474) adds the high electrical efficiency and reliability at low current densities to AFCs list of benefits. The electrical efficiency of advanced AFC system can be nearly 70 %.

The typical electrolytes in AFC are sodium hydroxide NaOH or potassium hydroxide KOH. The latter is more popular due to its high solubility. (Mikkonen 2003, 44.) The charge carrier in AFC is an OH^- ion and the chemical reactions are (Cook 2001, 4):

Anode reaction: $2\ H_2 + 4\ OH^- \rightarrow 4\ H_2O + 4\ e^-$

Cathode reaction: $O_2 + 4\ e^- + 2\ H_2O \rightarrow 4\ OH^-$

Overall reaction: $2\ H_2 + O_2 \rightarrow 2\ H_2O$

AFCs are very sensitive to carbon dioxide CO_2, which creates one of the main problems for the AFC applications. In the poisoning of AFC electrolyte, the carbon dioxide reacts with the hydroxide ion in the electrolyte to form a carbonate (Cook 2001, 6):

$$2\ KOH + CO_2 \rightarrow K_2CO_3 + H_2O$$

This results in reduced OH^- concentration in the electrolyte, lower diffusion rates and reduced mass transport. The overall efficiency of the AFC is therefore reduced when e.g. air is used as an oxidant without CO_2 removal. The situation is more critical in AFCs with immobilized electrolytes than in the case of circulating liquid electrolytes. (EG&G Technical Services, Inc. 2004)

4.3 Hydrogen vehicles

Hydrogen can be used as an automobile power source in two completely different ways: By using fuel cells (FCs) or burning the hydrogen in internal combustion engine (ICE). In this thesis work, only fuel cell powered vehicles are studied in detail. The history, schematic lay-out and efficiencies compared to conventional drive trains are represented in this chapter. At the end of the chapter there is also a quick look to the nowadays FCV market.

4.3.1 Hydrogen vehicles in history

For the first time hydrogen was used as a transportation fuel in a hydrogen balloon in 1783 by a French physicist Jacques Charles (National Hydrogen Association 2007b). As mentioned in chapter 3.1.1, hydrogen was also the first fuel for internal combustion engine when it was invented in 1807. The first fuel cell vehicle was a 20 horsepower tractor introduced in 1959 by Allis-Chalmers Manufacturing Company (HistoryWired 2006). It was powered by alkaline fuel cell (Cropper & Jollie FuelCellToday 2002) consisting of 1008 individual cells (Science Service 2007). According to Science Service, it was not pure hydrogen that was used as a fuel but a mixture of different fuel gases, largely propane.

Figure 5. Hydrogen powered fuel cell passenger vehicle on public roads built by K. Kordesh in 1970 (Kordesh et al. 1999)

In 1970, K. Kordesch modified Austin A40 to a hydrogen fuelled FC vehicle (figure 5). According to Kordesch et al. (1999) the vehicle was used on public roads for three years and it was powered by liquid hydrogen fuelled AFC together with seven series connected lead-acid batteries acting as an additional energy source during hill-climbing and acceleration. On-board the vehicle there was a hydrogen/air FC system with circulating KOH electrolyte. The main functional idea of the propulsion technique was that the FCs would work only during actual driving. This avoided electrolyte damage related to shunt currents and also some open circuit problems. Hydrogen was stored in six hydrogen tanks each weighting 13 kg. The total amount of hydrogen storage capacity was 25 m^3 of H_2 at a pressure of 150 bars. (Kordesch et al. 1999.) A more serious work on alternative fuels begun in 1973 after the OPEC oil embargo and the resulting petroleum crisis. Some twenty years after, Daimler Benz demonstrated NECAR I, which is said to be the worlds first modern FCV. The demonstration took place in 1994 in Germany. (National Hydrogen Association 2007b)

4.3.2 Fuel cell vehicle (FCV)

Fuel cell vehicles can be classified to two main categories, though the line drawn between the categories is not clear in every case. Principal difference between the two groups is that the others use FC system directly as a power source without a battery (FCV) and the others use battery or supercapacitor in addition to the FC system. The latter is called fuel cell hybrid vehicle (FCHV). A so called range extender type fuel cell vehicle can also be designed with low power FC whose function during operation is to charge batteries, which are designed to provide full power needed to move the vehicle (EG&G Technical Services, Inc. 2004). The propulsion system of a fuel cell vehicle is different from conventional ICE vehicle in many ways. Conventional propulsion (figure 6) is based on continuous fuel combustion process and a separate transmission system. The batteries in conventional vehicles are used only to power lights and ancillaries.

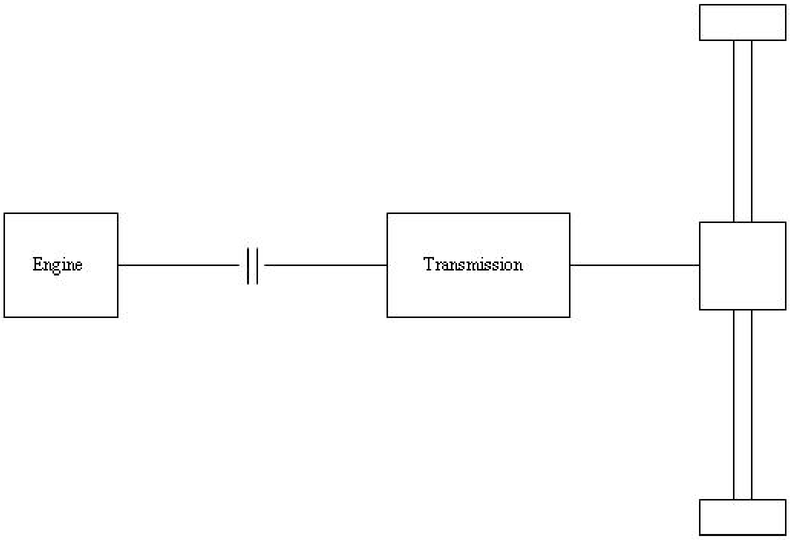

Figure 6. Schematic illustration of a conventional ICE vehicle. (Modified from Ludwig Bölkow Systemtechnik 2002)

In general, when using a fuel cell system to power a vehicle, the electrical power created by the chemical reaction between hydrogen and oxygen must be converted into mechanical energy. The typical FCV system consists of a fuel storage tank, fuel cell stack, power controller unit and a traction motor (figure 7). Some FCVs are designed to use a liquid fuel such as methanol or gasoline, which can be stored on-board in a liquid form. Vehicles using these fuels also need a reformer, a fuel processor that breaks the fuel into hydrogen, CO_2 and water. Although this process generates carbon dioxide, it produces much less of it than the amount generated by conventional gasoline powered vehicles (Ludwig Bölkow Systemtechnik 2002, 80). FCV equipped with a fuel reformer can also be called FPFC, fuel processor/fuel cell system (Ludwig Bölkow Systemtechnik 2002, 76). Methanol can also be used directly in fuel cells without a need for a fuel reformer.

Similar to electric vehicles, a traction motor is used as a drive train in FCVs instead of combustion engine. The electricity produced by a fuel cell is DC energy, and the voltage depends on the amount fuel cells connected in series at the fuel cell stack. Majority of the present day FC systems are designed for lower voltages i.e. from 12 V to 100 V. Therefore, in most cases, a DC/DC boost converter is often needed to raise the voltage before further processing via a DC/AC inverter (EG&G Technical Services, Inc. 2004).

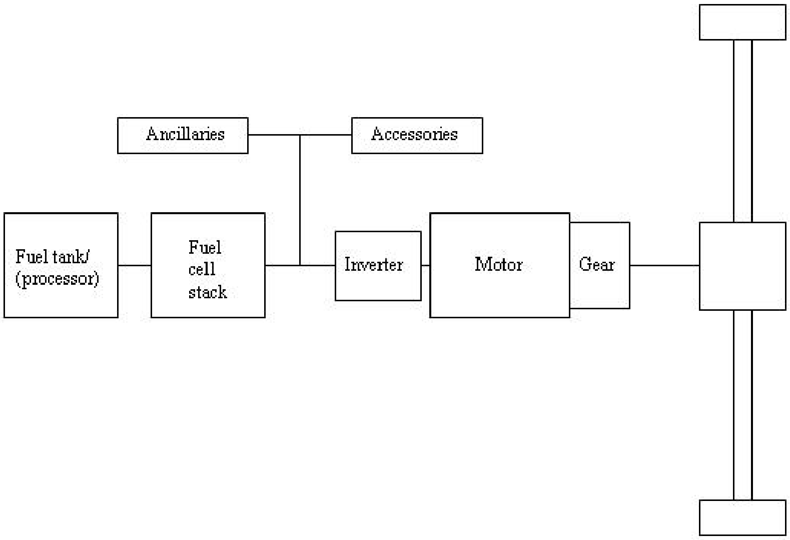

Figure 7. Schematic illustration of a fuel cell vehicle. (Modified from Ludwig Bölkow Systemtechnik 2002)

The power controller unit is needed to manage the operation of the FC stack and the electric motor. The FC stack can be free breathing, which means it takes the oxygen from the surrounding air without any pressurizing, or an additional air compressor can be used to control the oxygen or air flow according to the demand. Hydrogen supplied to the stack is always pressurized to certain overpressure. Finally, the electrical energy is converted to motion by one or several electric motors.

4.3.3 Fuel cell hybrid vehicle (FCHV)

The vehicles can easily be categorized by the type of fuel they use, but this does not necessarily reveal much of the propulsion system of the vehicle. There are numerous different hybrid vehicles that are combinations of various propulsion, fuel and storage techniques.

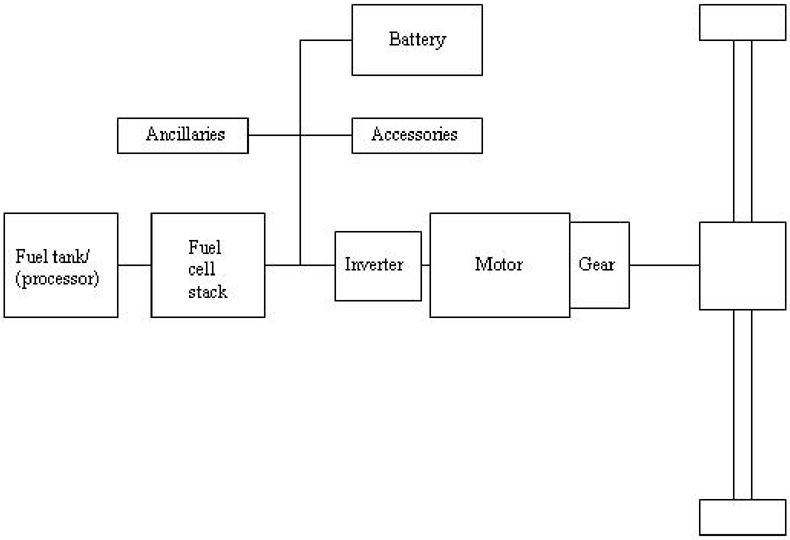

Figure 8. Schematic diagram of a fuel cell hybrid vehicle. (Ludwig Bölkow Systemtechnik 2002)

Fuel cell hybrid vehicles (figure 8) have similar properties than hybrid electric vehicles (HEVs) that are already available for large public. Hybrid electric vehicles typically combine the conventional ICE and an energy storage device, such as battery. The key components of HEV are the energy storage system, power unit and electric motor i.e. the vehicle propulsion system (EG&G Technical Services, Inc. 2004). Toyota Prius is a common example of a hybrid electric vehicle, which is powered by gasoline engine combined with secondary battery. In the case FCHV, fuel cell hydrid vehicle, the ICE is replaced by a FC stack. In both cases the regenerative braking system can be utilized to charge batteries (Kawai 2004, 62). FCHVs are sometimes called as FCHEVs, fuel cell hybrid electric vehicles. The energy storage unit of a hybrid vehicle can be a battery unit, ultracapacitor, fly-wheel or a connection of these options. A vehicle that combines battery, ultracapacitor and FC system to a traction motor can also be called triple hydrid vehicle. (Pede, Iacobazzi, Passerini, Bobbio & Botto 2003)

In the case of FCHVs, the energy storage unit is used to achieve the following advantages (Pede et al. 2003):

- The size of an FC, which is the most expensive component of the system, can be reduced. This reduces the overall drive train cost substantially.
- Drive train flexibility is increased, which leads to improved efficiency due to buffer energy storage which compensates power transients and slow responding FC system.
- Cold start up system heating can be done with onboard energy storage, which enhances the FC performance.
- The braking energy can be recovered and released later in the form of electricity. Otherwise this energy will be lost.

By combining e.g. battery and UC, it is possible to regenerate the braking energy with higher efficiency. The amount of energy saved only by braking energy recovery is cycle dependent and varies between 3.5 and 20 %. (Pede et al. 2003, 280-291)

In all propulsion systems, hybridisation reduces fuel consumption. The benefits of hybridization are larger for ICE vehicles than FCVs because the efficiencies of the fuel cell vehicles are larger in the first place (Ludwig Bölkow Systemtechnik 2002). For certain hybrid vehicles, such as FCHVs, the system configuration can be represented by an index called 'hybridisation rate', that is the ratio between the installed power source and the power required for traction:

$$\text{hybridization degree} = \frac{p_{gen}}{p_{tract}}$$

Hybrids combining ICE and traction motor are expected to be very popular in the near future. By the end of 2003, Toyota had already sold more than 100 000 HEVs in Japan and the US, which according to Toyota accelerates the commercial advent of FCV as well by improving the performance and reducing the cost of hybrid electric drive train (Sperling & Cannon 2004a, 10).

4.3.4 Fuel economy and efficiency of a H_2 vehicle

The direct conversion of fuel into electricity allows fuel cells to achieve substantially higher efficiencies than combustion engines, the efficiency of which is limited by Carnot's Law of Thermodynamics. The efficiency of a FCV can be improved by regenerative braking and utilization of waste heat. The efficiency of FCHV using compressed hydrogen has vehicle efficiency about 2.5 times that of gasoline vehicle manufactured in 2002 (Figure 9).

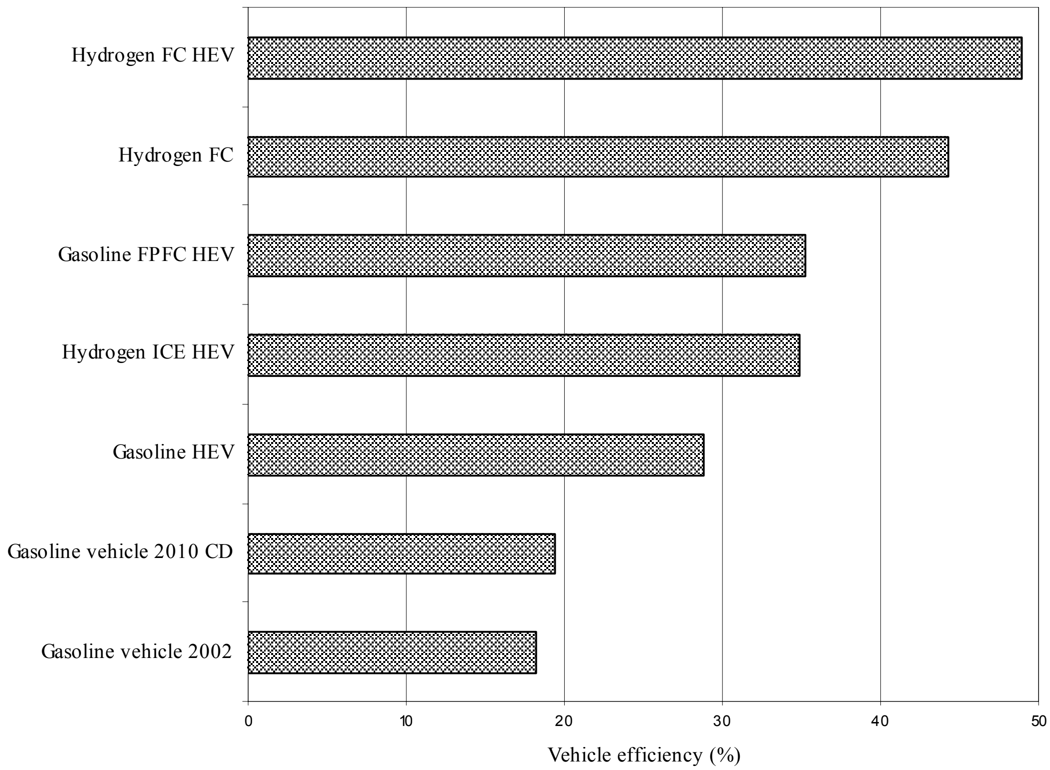

Figure 9. The efficiencies of different types of propulsion systems. Vehicle efficiency is defined as the ratio between the energy delivered to the wheels and total energy content of the fuel input. CD = Conventional drive, FPFC = Fuel processor and fuel cell (Ludwig Bölkow Systemtechnik 2002, 80-82)

According to Bekkeheien et al. (1999) a modern ICE can achieve transformation efficiencies slightly above 30 % under optimal conditions. However, the problem for ICE vehicles is that they are usually designed to operate the most efficiently at higher loads than average driving condition. This leads to low vehicle efficiencies during the average driving, starting from as low as 17 %. (Bekkeheien et al. 1999, 102.) For a FCHV, vehicle efficiency is almost 50 %, but well-to-tank efficiency for hydrogen is low. To achieve a well-to-wheel efficiency of 42 %, which is three times higher than that of current gasoline vehicles, the vehicle efficiency of a FCHV must rise to 60 % (Kawai 2004, 64). Recently on a Japanese survey on hydrogen vehicles the fuel economy of mid-sized FCV was determined as 16.6 km/Nm^3 H_2 (Oi & Wada 2003, 348).

4.3.5 Well-to-wheel emissions of a FCV

Well-to-wheel greenhouse gas emissions of the H_2 vehicle depend on which process is used to produce, to transport and to store the fuel. According to the study made by MacLean and Lave (2003), The GHG emissions of hydrogen FCV lie between 70 – 241 gCO_2equiv./km and the same for mid-sized gasoline powered ICE vehicle was 248 – 333 gCO_2equiv./km (Maclean & Lave 2003). When hydrogen is produced from renewable energy sources at the filling station, well-to-wheel GHG emissions approach zero. Compared to conventional ICE, the CO_2 emissions can be also significantly reduced by using methanol fuelled FCV. According to DaimlerChrysler, NECAR5 which is equipped with methanol fuel reformer produces only a third of the carbon dioxide emissions of a gasoline powered ICE vehicle (Helynen et al. 2004, 233).

4.3.6 H_2 passenger vehicles on the market

Hydrogen fuelled vehicles are entering the vehicle markets with an increasing speed. Every major manufacturer of automobiles has their own response to the FCV challenge. The first fuel cell bus with 90 kW PEMFC was introduced in 1993 By New Flyer Industries Ltd and it had a driving range 400 km (Kivisaari 2003). According to the Online Fuel Cell Information Resource, 18 different hydrogen fuelled passenger vehicles were being introduced before the year 2000. In 2003, the number was increased to 65. (Kivisaari 2003.) The most common hydrogen storage method among these vehicles was compressed gas. Methanol was the next popular fuel for these FCVs. The amount of vehicles storing the H_2 in metal hydrides was less than 10 %. Of the different propulsion types, most common was the combination of FC and battery and only one vehicle was registered to use ICE powered by hydrogen. The best driving range achieved by the vehicles in the list was 970 km made with Jeep Commander fuel cell hybrid vehicle. (Kivisaari 2003)

In 2007, 74 different fuel cell vehicles was registered to this information resource (Fuel Cells 2007), compressed H_2 remaining the most popular storing method. The best driving range achieved with MH storage was 300 km. It was achieved by Toyota FCHV-3 equipped with 90 kW PEMFC (Fuel Cells 2007). Toyota was also the manufacturer to introduce the world's first FCV into the commercial markets in 2002, simultaneously with Honda (Kawai 2004, 62). In Finland, there are no officially registered hydrogen vehicles (Malin, J., AKE, phone conversation 14th March 2007), but there are some registered for the testing purposes.

4.3.7 Hydrogen filling stations

According to the Online Fuel Cell Information Resource (2007), the first H_2 vehicle refuelling station was established in Munich, Germany in 1989, to support the test vehicles developed by BMW. The first commercial H_2 fuelling station opened in Reykjavik in 2003. On February 2007, there were 138 operational hydrogen fuelling stations worldwide and 64 were planned to be opened in the near future. The number of fuelling stations has been increasing steadily, since in 2003, the same register held only 73 refuelling stations providing hydrogen. (Kivisaari 2003.) A development of hydrogen fuelled vehicles provide three main benefits for the society: independence from imported oil, reduced CO_2 emissions, and cleaner local air to the highly populated areas. These are obviously the reasons why 22 % of the operational and 28 % of the planned H_2 fuelling stations is located in California, U.S. (Fuel cells, 2007).

4.3.8 Some future prospects for hydrogen vehicle commercialization

According to Kawai (2004, 64) main concerns before the FCHV commercialization are safety and storage issues. Also cost and servicing during the vehicle use and the environmental impacts from recycling and disposal of the vehicle needs further investigation. Low temperature operation is also one of the main technical challenges facing the FCHV, since the exhaust water can freeze at low temperatures and paralyze the operation of the fuel cell. (Kawai 2004, 64.) However, Sperling and Cannon (2004b, 236) lists four main benefits which make hydrogen fuel cell vehicles worth developing despite the technical challenges they are facing today. First of all, they are environmentally superior to vehicles burning fossil fuels and high torque characteristics of electric motor provide rapid acceleration. Other benefits include quiet operation and potentially low maintenance requirements. (Sperling & Cannon 2004b, 236.) Some scenarios projecting the future of FCVs take the FC car utilizations one step further, making them from power consuming devices a small scale moving power plants. The cars are in some scenarios planned to provide electricity to the grid when parked (Lahidji et al. 1999, 20) or even provide electricity to microwave ovens on the picnic (Kurani et al. 2004, 50).

5 STAND-ALONE HYDROGEN REFUELLING FACILITY

5.1 Small scale RE systems

In addition to utilization of RE sources, building a hydrogen economy is also related to distributed energy production, which is expected to increase its share in global energy production. Mattila et al. (2001, 116) estimated that 40% of the annually installed electricity capacity could be small scale, i.e. <10 MW, by 2010. In the viewpoint of energy policy, distributed generation possesses numerous benefits, such as minimized energy transport losses, system reliability improvements and increased adaptability to power demand variations (Helynen et al. 2004, 294). One of the biggest advantages is its suitability for RE utilization, since renewable energy can be utilized in some form at any location. To its users, a small scale energy system provides some benefits as well. The community based on independent energy system is not vulnerable to large cut-offs of energy supply. And a system based on renewable energy is always a move towards a sustainable energy infrastructure. The renewable energy system could derive its primary energy from any single RE source, or it could be some combination of different energy collectors, like for example, combined system of PV panels and wind turbines.

5.1.1 Storing the intermittent energy

The energy production units can operate either continuously or intermittently. Of RE sources, biomass and hydro units can operate continuously, whereas wind, solar and wave applications produce a rather intermittent energy output (Zervos, Diakoulaki & Mayer 2002, 180). There can be a lot of fluctuation due to time of day, wind conditions, cloudiness etc. It is rare that the consumption rate meets the production precisely. That is why the produced energy has to be stored during the period of low consumption, to be used later when the production can not meet the consumption.

Energy storage units can be categorized depending on their function as short, medium and long-term storage. From a technical point of view, they can be grouped to categories: mechanical, thermal and electrical. In addition to hydrogen storage, the main available options include batteries, hydraulic or pneumatic accumulators, flywheels and pumped storage. (Zervos et al. 2002, 181-182.)

According to the research made by Vosen and Keller (1999), if a certain stand-alone RE system necessitates energy storage of one months energy demand due to intermittent nature of the energy sources, the concept will be become costly by using conventional battery technology only. A hybrid storage system that consists of combination of conventional storage batteries and hydrogen technology can dramatically reduce the cost of energy storage over single storage systems. (Vosen & Keller 1999, 1139-1156.) Numerous other researchers have also noticed the potential of hydrogen as a storage medium. Agbossou, Kolhe, Hamelin & Bose (2003b, 17) determine that hydrogen produced by electrolysis is a promising alternative to long-term renewable energy storage. It also suits to seasonal storage applications better than commonly used battery storage, because of its high energy density. Agbossou et al. (2003b, 17) state also that a typical self-sufficient renewable energy system must include short-term energy storage, like battery unit, and an additional long-term storage. Batteries for short-term storage have benefits like high charging-discharging efficiency and a capacity of taking care of the energy peaks coming from the energy source. Concerning the long-term storage, batteries have deficiencies like low energy density, self discharge and leakage. (Agbossou et al. 2003b, 17.)

5.1.2 Energy flows and equipment in a stand alone RE systems

A system based on hydrogen storage and renewable energy sources, such as wind and PV panels, is an example of an energy system with minimal impacts on the environment. Additional components of such a system include the electrolyser, fuel cell stack, voltage converters, DC and AC loads, energy flow control unit, inverter, battery unit and H_2 storage.

The example system (figure 10) converts the energy from the sunlight and wind into electrical power, which drives the electrolyser to produce hydrogen and oxygen. The water consumed by electrolysis is converted to gases, and again back to water in a fuel cell. Hydrogen is stored in pressurized tanks in this case, but also other methods such as metal hydride storage can be used. With a system equipped with metal hydride storage, according to Vanhanen, Lund and Tolonen (1997), a round-trip efficiency of 30% has already been achieved. Fuel cells in such a system can be either stationary or they can be located e.g. on-board a vehicle.

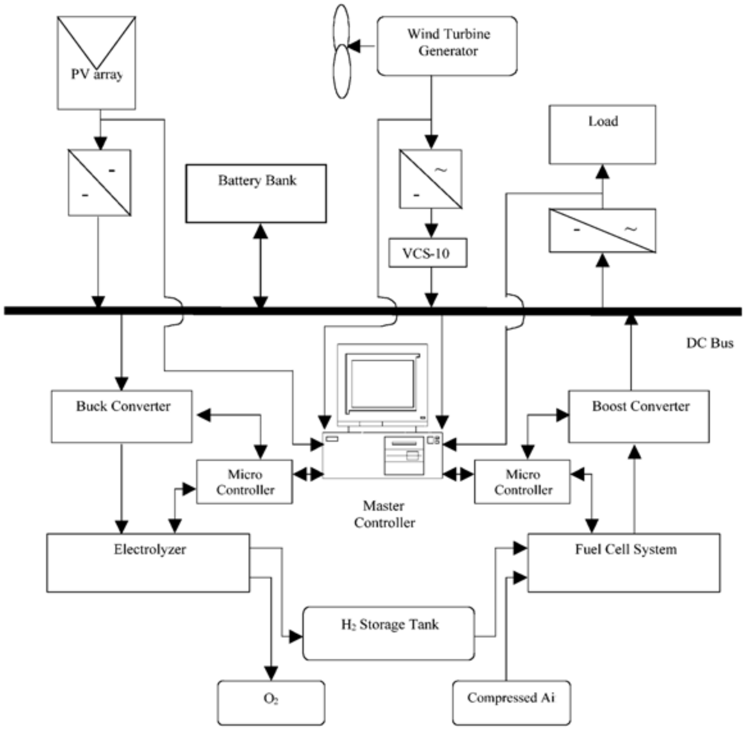

Figure 10. Schematic picture of a renewable energy system based on hydrogen storage. (Agbossou, Kolhe, Hamelin & Bose 2004, 634)

The oxygen as an electrolysis by-product offers a potential for the system improvement. It is been tested (Agbossou, Kolhe, Hamelin, Bernier & Bose 2003a) that the modification of the electrolyser for O_2 storage, purification, drying and re-utilization as oxidant for the PEMFC improves the operation of the system. It has been noticed that feeding enriched oxidant instead of air to the PEMFC improves the current density and the efficiency of the PEMFC. By improving the system performance the oxygen recovery also helps to reduce the size of the PEMFC stack. (Agbossou et al. 2003a.)

The RE system components, consisting WTGs and PV arrays, have substantially different voltage-current characteristics. To achieve autonomous operation of the system, the components can be integrated through different power conditioning devices on a DC bus. An autonomous system requires a lot from the control unit managing the energy flows among the different components of the system. For example, the control system successfully tested by Agbossou et al. (2004, 634) consisted of a master controller for the overall energy management, and secondary micro controllers, which manage the energy flow through power conditioning devices. In addition, to achieve optimal utilization of multi-renewable energy sources, sophisticated algorithms capable of forecasting load requirements and renewable power production can be applied (Zervos et al. 2002, 181).

5.2 Hydrogen refuelling station and a test vehicle in Jyväskylä University

The system under research within this project consists of energy production equipment, hydrogen processing unit, metal hydride storage, PEM and alkaline fuel cells and hydrogen fuelled test vehicle. The energy production equipment was installed on the roof of Jyväskylä University physical chemistry building at Ylistönmäki campus area. The hydrogen production equipment was located in the laboratory two stories below the PV panels and wind turbine. The vehicle was kept in the car park and refuelling was planned to happen by changing the empty portable metal hydride container to a full one recharged in the laboratory. The complete analysis of this system is not included to this thesis, but the whole facility is been introduced in this chapter for clearance.

5.2.1 System description and the energy production unit

The energy producing system included five PV panels and one horizontal-axis wind turbine (figure 11). The maximum power capacity of each PV panel was 100 W and a wind turbine had a capacity of 500 W, making total maximum capacity of 1 kW.

Figure 11. PV panel array (100 W each), wind turbine (500 W) and a weather station at the roof of the physical chemistry building of the University of Jyväskylä, Finland. (photo: RE program)

The energy produced by these two units was connected through a control unit to a battery pack, which consisted of two integrated and series connected 12 V and 75 Ah batteries, thus having an overall storing capacity of 150 Ah (24 V). The system included also an external load for times when the production of energy exceeded the consumption and the when the batteries were full. This simple resistor converted the excess energy to heat energy, which was released in the laboratory room. Figure 12 illustrates the schematic diagram of the whole hydrogen production system from primary energy source to its end-use in a test vehicle.

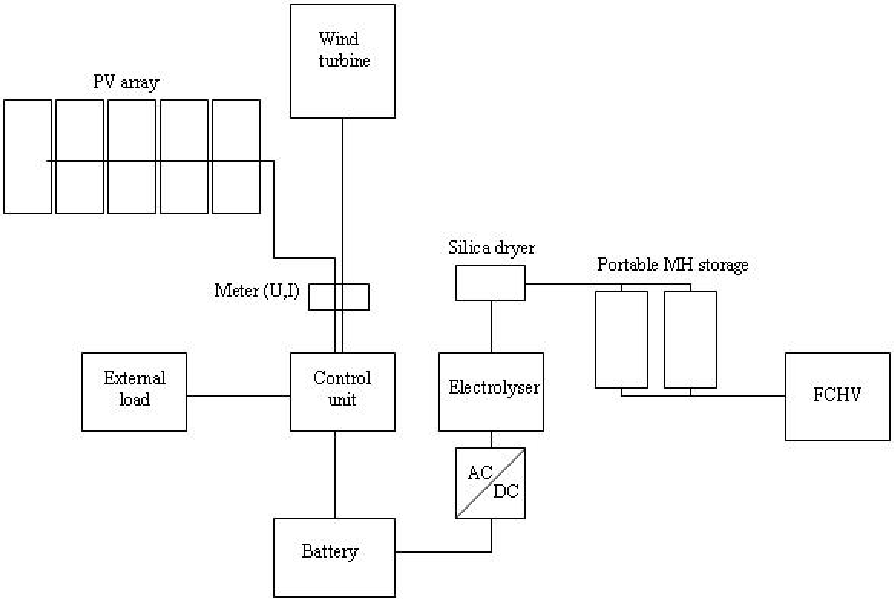

Figure 12. Schematic representation of a Jyväskylä University hydrogen vehicle and refuelling station test scheme. Hydrogen is produced by using RE sources and stored in portable MH containers. Later, hydrogen is utilized as a fuel for a fuel cell hybrid vehicle.

System components and some of their technical specifications are listed in the table 5. PV panels, wind turbine and the external load were connected to the control unit. The function of a control unit was to switch the generated energy to the load when there was no consumption of energy and the battery unit was full. In addition to the components listed in the table, the system consisted of different hose and fitting assemblies, which are explained in more detail later within the tests made with each component. The next chapters will introduce each part of the system more specifically.

Table 5. Components of the Jyväskylä University hydrogen vehicle test scheme.

Component	Specifications, manufacturer etc.
PV Panels	5 x 100W (maximum), NAPS systems OY, Finland
Wind Turbine	500 W, Horizontal axis, made in Russia
Control Unit	Made in Russia
Resistor, load	
Battery unit	150 Ah, 24 V, Optima Batteries, made in U.S.A.
Inverter	DC/AC, 30S-12E, 300 W (continuous), output 230V
Electrolyser	HG500, 350 W, Labgas Instruments
Hydride containers	2 pcs, HC-MH1200, Hydrocell Ltd, Finland
PEM fuel cell	500W, HCORE-500-2, H Power Corporation,
Alkaline fuel cell	HC400, Oy Hydrocell Ltd, Finland
Test vehicle	FCHV Fantasia, EL3 Ltd Finland

5.2.2 Inverter and the electrolyser HG500

The energy produced from RE sources was used in electrolyser to produce hydrogen. As a side product of the electrolysis, oxygen is also generated. In this project, the oxygen was not collected to be fed to the PEMFC, because it would have required more complex arrangements to the hose assembly. Because operating voltage of the electrolyser was 200 - 250 VAC and the battery voltage 24 VDC, an inverter was needed between the batteries and the hydrogen generator (figure 13). Technical information of the DC/AC sin wave inverter is found in table 6.

The electrolyser used in this study was HG500 manufactured by Labgas Instrument Company. The electrolyser has two internal PEM cells in which the electrolysis takes place. All the metal salts are poisonous to the membranes and therefore there is strict requirement for the inlet water. If the inlet water quality specifications are followed, the cell does not need any scheduled service during its normal operational life. The expected normal operational lifetime of the unit is not told in the manual. However, contaminants affect adversely to the membranes performance and therefore to the electrolyser lifetime as well. In this study, the inlet water used was deionised water produced in the chemistry department of the University.

Figure 13. Electrolyser HG500, DC/AC inverter and the battery unit. (Photo: Pasi Paananen)

Table 6. Sin wave inverter 30S-12E technical information

Parameter		Value
Power Out,	Continuous	300 W
	20 minutes	350 W
	Peak	500 W
Output voltage		230 V ± 3%
Output frequency		50 Hz
Output wave type		Sin wave
Harmonic distortion		< 4 %
Input voltage		10 V – 15 V
Efficiency		> 90 %
Current consumption (no load)		0.7 A
Warning limit, low input voltage		10.7 V
Cut-off limit, low input voltage		10 V
Measures (L x W x H) (mm)		285 x 120 x 62
Weight		1.75 kg

Inside the generator there is an internal water reservoir, which is able to maintain normal operation for 15 minutes if the inlet water supply is restricted. The unit is equipped with an internal water quality check that protects the generator cell. Other internal detectors of the unit measure system leakage, excess flow rate and pressure. According to the manual, the general service time interval is recommended to be 4000 hours of normal operation. In addition to that, it is recommended to change the water inside the unit periodically, for example, once a month. That is because the highly aggressive deionised water might dissolve to itself foreign matter from the construction material in the long run. The relevant technical information concerning the hydrogen production unit is represented in the table 7.

Table 7. Technical information of the electrolyser used in this study. (Source: Labgas instrument company, operating manual, hydrogen generators)

Parameter	Value
Maximum flow rate	500 cc/min
Nominal pressure	6 bar (gauge)
Temperature	+5 - +40 °C
Relative Humidity	20 % - 70 %
Dimensions (W x D x H)	315 x 320 x 450 mm
Weight	18 kg
Voltage (in)	200 - 250 V
Current	1.5 A
Fuse	3.15 A (slow)
Power	350 W
Water quality requirements	Deionised, conductivity max. 16 MΩ
Water inlet overpressure	from 0.07 to 0.2 bar

Due to the nature of the process, the hydrogen produced with the unit, is saturated with water vapour. The generator unit (figure 13) has also an internal moisture exchange tube, which transfers the water vapour from the gas to the atmosphere. This will bring the hydrogen dew point to +15 °C. To enhance the moisture removal of the hydrogen gas, the unit was equipped with an external silica dryer unit placed between the generator and the hydride container.

According to the HG500 instruction manual, hydrogen production rate can be approximated with following statement: One ampere of electricity per membrane produces 7.8 ml of hydrogen per minute. Since this model includes two membranes, the production rate can be estimated with the formula:

$$\text{Rate of production} = 2 \times I(amps) \times 7.8cc/\min$$

The electrolyser can be fitted with an analogue data out connection, to retrieve cell voltage and amperes readings through a D9 connector. In case of the analogue data reading, the input impedance of a measuring device should be 1 MΩ minimum. The analogue out is a so-called set-and-forget device that works with no settings once installed.

5.2.3 Hydrogen containers HC-MH1200

After the silica dryer, hydrogen is stored into portable metal hydride containers. Within this project, there was two similar containers each having a capacity to store 1200 litres of hydrogen (Oy Hydrocell Ltd 2005). This is why the model name HC-MH1200. Later, during the tests, the serial numbers of the containers were used to distinguish the hydrides from each other. The individual serial numbers for the containers were 014-0302 and 013-0202. Between the electrolyser, dryer and the container, there was no pressure alternating fitting between them. The pressure inside the generator shifted directly into the container. In the chapter 4.1 the functioning of metal hydride as hydrogen storage medium was explained in more detail. In the containers used within this study, aluminium (Al), Mangesium (Mg) and Mangane (Mn) are used as a storing material. The material inside the container is in powder form and the container volume is 3.6 litres. The amount of self-leakage from the sealed container is undetectable, according to the importer, and the containers can store hydrogen for years without any weakening of hydrogen capacity. (Oy Hydrocell Ltd 2006a.) Because only hydrogen is stored to the hydride and not air, the gas withdrawn from the container is very pure. It can reach purity level of 99.9999% (Oy Hydrocell Ltd 2006a). The containers are equipped with a pressure gauge, quick lock male connector and an emergency valve, which opens itself when the pressure rises to approximately 30 bars (Rantanen J. email 03 April 2006). This will drop the increased container pressure by allowing the hydrogen flow out from the bottom of the container. Because the pressure is strongly depended on the temperature, this means that the highest operating temperature for this container would be 65 - 70 °C (Rantanen J. email 03 April 2006). An important point in predicting the remaining capacity of the container is that the pressure of the container does not drop linearly when the container is emptying. The pressure of the empty container almost equals to that of full one, assuming the temperature does not change (Rantanen J email 03 April 2006). In practice, when the hydrogen is consumed, the pressure drops inside the container. This is due to the endothermic character of the reaction rather than lowering of the capacity. When the hydrogen is drawn from container, the hydride cools down (Rantanen J, email 3 April 2006). The relationship between the temperature and pressure, according to the container manufacturing company, is represented in the attachment 1.

As mentioned earlier, container can hold 1200 litres of hydrogen. This corresponds electrical capacity of 220 Ah at 12 VDC. The weight of the container is 11 kg, which means the storing capacity of 0.98 % H_2 by volume. The diameter of the container is 14.6 cm and the height is 34 cm. There is also a heat transfer system inside the container which allows the altering of pressure by cooling or heating the container if needed. (Oy Hydrocell Ltd 2006a.) Due to the facts mentioned above, hydrogen can not be drawn from the container rapidly without external heating, because cooling of the hydride means lowering down the rate of the gas outflow (Rantanen J, email 13th April 2006). This makes a metal hydride far different storage method compared to the conventional compressed gas storage. This could cause some difficulties technically, but according to the safety issues the result is the opposite. The nominal hydrogen outflow rate from this type of container is 400 l/min (Rantanen J. email 13th April 2006).

5.2.4 Hydrogen fuelled test vehicle

The fuel generated by the electrolysis was to be used in a test vehicle. The name of a prototype vehicle was Fantasia (figure 14) and it was manufactured by Oy EL 3 Ltd, Tampere, Finland. The idea to develop such a test vehicle came from Arto Salonen and originally it was designed to be used in cities for short distances. At the first place, also disabled drivers were taken into account in its design (Liukkonen, 2002). Later, there were some modifications made to the vehicle, that no person with a wheelchair could drive it. According to Liukkonen (2002), the development work of Fantasia started in 1999 and its “role model” had been BMW Isetta, a three-wheeled light duty vehicle. The vehicle was registered as an L5 class vehicle in 2002. (Liukkonen, 2002).

Figure 14. Test vehicle Fantasia ready to hit the test track in Jyväskylä during the spring 2006.

Test vehicle derives its driving power from two American Briggs and Stratton 6 kW electric motors, making the total driving power 12 kW. Vehicle power measured in horse powers is 16.3 hp. On this rear driven car the power to the motors is drawn from two series connected 75Ah batteries and a fuel cell, which makes the car a fuel cell hybrid vehicle. The car can be driven also without a fuel cell using only battery power. When the fuel cell is in use it charges the batteries during driving, and the batteries can act as an electricity storage buffer during peak demand. The electrical system voltage is 24 V and the main batteries are connected to the control unit. In addition to the driving batteries there is a separate battery (figure 15) for the necessary equipment, such as winkers and headlights. This battery is 12 V and has a storing capacity of 42 Ah. There is a chain transmission between the traction motor and the rear shaft. Theoretical top speed with the existing transmission wheels and current tyres is 72 km/h with the winter tyres and 80 km/h with summer tyres. (Salonen A., personal meeting, 20th July 2005).

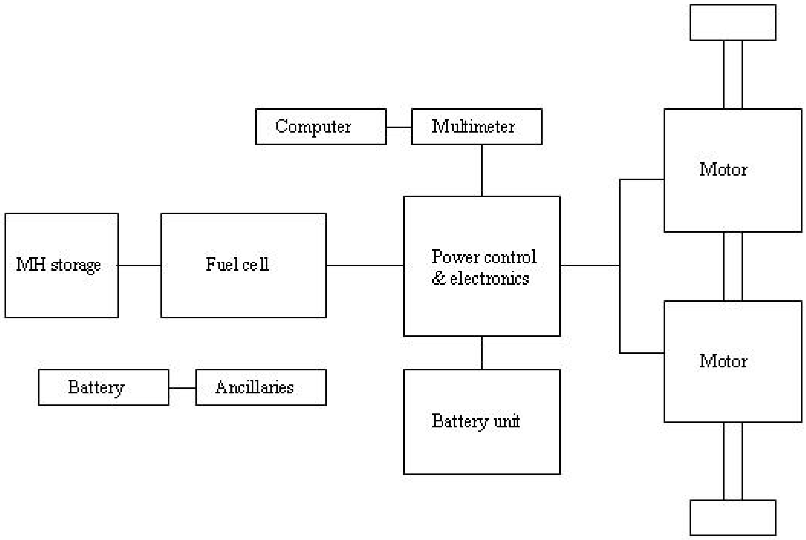

Figure 15. Schematic illustration of the test vehicle Fantasia.

A control unit is used to control the clutch, accelerator pedal, motors and other integrated driving equipment. Some measuring equipment like temperature, voltage and current sensors are integrated to the control unit. The main frame of the car is made of steel and its bottom is made of plywood. The upper part of the frame is made by adhesive technique of ABS plastic. According to Liukkonen (2002), the design and manufacturing of the frame elements is made at Kymenlaakso University of Applied Sciences and the windows and the seat were made at HAMK University of Applied Sciences in the city of Hämeenlinna, Finland. Part of the manufacturing process like the assembly and the electrical installations were made in educational center Tavastia in Hämeenlinna during the spring 2001. This is also the very place where the first test drives were made. (Liukkonen, 2002.) The spiral suspension system of the vehicle has McPherson type heads in front and longitudinal triangle-shaped heads at the rear. The vehicle has hydraulic disk brakes at every wheel. The outer measures of the car are following: Length 2350 mm, width 1470 mm, height 1690 mm. The distance between shafts is 1650 mm and the shaft length is 1450 mm.

An external measuring device, a multimeter (figure 16), could be connected to the control unit of the vehicle. With the multimeter, data collection to the lap-top computer is possible during driving. The unit shows driving speed, voltage, current, power and temperatures of different parts of the motor and the control unit.

Figure 16. The external multimeter used as a data collection device during the test drive.

The device gives out a CSV type data, which can be easily moved to Excel format for further analysis. The device draws its power from the 25-pin connector at the control unit of the vehicle, but it also has a 9V battery power source for periods with no power from the control unit. The signal sent by the multimeter can be analogue or digital, and the device can be connected to the computer with a serial cord, which is necessary according to the data collection since the device itself has no mass memory. The data which the multimeter is sending out is 19200 baud 8N1 data, which can be received by a computer with a 15-pin D2 connector. The data type follows the standard RS-232 and the time interval of the data pulse is one second. Received data can be collected through a data transfer program e.g. Hyperterm.

5.2.5 PEM fuel cell HCORE-500-2

HCORE-500-2 fuel cell (figure 17) is one of the fuel cells to be used within this project. The hydrogen stored in a metal hydride container is aimed to lead to a fuel cell to generate electrical power to the vehicle. This means that the electrical power generated on-board either charges the battery unit of the vehicle or will be converted directly to motive power by the traction motors.

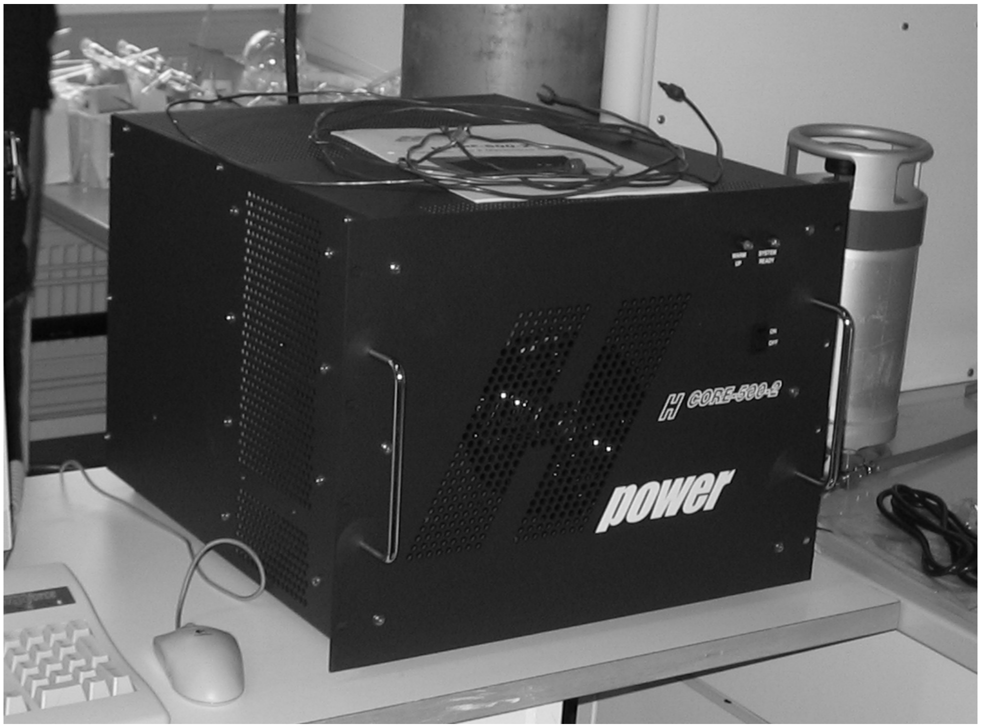

Figure 17. HCORE-500-2 polymer electrolyte fuel cell. (photo: Pasi Paananen)

According to the HCORE-500-2 manual, the fuel cell is designed for both outdoor and indoor use, but outdoor use requires a proper enclosure. The fuel cell takes the oxygen needed to the reactions from the air. Therefore it is necessary to allow free flow of air into and out of the unit cover. Because the fuel cell has a limited tolerance for impurities, according to the manual instructions, the hydrogen used should be at least 99.95 % pure. One crucial point about the hydrogen inlet is that the pressure of hydrogen source must not exceed 4.1 bar (60 psig). According to the manual, the operating hydrogen supply pressure must be 3.1 ± 1.0 bar (45 ± 15 psig). The HCORE-500-2 has an air inlet filter for the mechanical pump. Additional technical information of the fuel cell is represented in the table 8.

This fuel cell is capable of providing power up to 500 W with a nominal voltage of 48 VDC as long as there is an uninterrupted supply of hydrogen. The maximum hydrogen flow rate should not exceed 488 l/hour. When continuously delivering its rated output, the fuel cell can produce a maximum of 8.36 litres of water in a 24-hour period. The fuel cell has several functional inputs and outputs which are explained in the next paragraph. The connection details are given in brackets.

Table 8. The technical information HCORE-500-2 (source: Hcore500 installation, use and maintenance guide)

Parameter	Value
Input Voltage, battery charging	120 VAC ± 10 %
Input Current	1 Amp
Output Voltage	48 VDC Unregulated (39 VDC – 65 VDC)
Output Power	500 Watts (1/3 hp)
Operation Temperature	from –5 °C to +40 °C

Hydrogen input (male 1/8" NPT fitting) needs a pressure regulator in the line between the metal hydride and the FC to maintain the pressure between the limit values. There is no integrated pressure regulator in the fuel cell. Water discharge line (Hose assembly 3/4" ID) locates at the bottom of the rear panel of the cell and produces water at a rate depending on the operational level of the fuel cell. During continuous operation the fuel cell will purge small amount of hydrogen and water vapour through the hydrogen purge line (Hose assembly ¼" ID PVC tubing with barb 1/8" male NPT). Hydrogen over pressure vent line (male 1/8" NPT fitting) is required in the event the hydrogen supply exceeds maximum specified pressure. The enclosure full of small holes acts as an airflow vent which emits heat as a normal by-product of power production. The recommended empty distance around the vents of the unit is about 10 cm. The load can be connected to the two 6-32 lug terminals of the unit. As a data port, there is a 9-pin BD-9 connector for RS-232 serial communications.

Storage temperature for the FC is between 5 °C and 50 °C. To assure the safe operation of the FC, the unit has an internal hydrogen sensor that shuts down the system when unsafe level of hydrogen is detected. The unit has also an internal battery unit which has to be charged with external power before the fuel cell is placed into service or after a long storage time.

The pressure regulators used in this study were made in Germany by Rectus Group. There are two different types of regulators, both having an inlet maximum pressure of 15 bar. The outlet nominal pressure for the other is 3 bar and for the other 4 bar.

5.2.6 Alkaline fuel cell HC-400

The electrolyte inside the alkaline fuel cells HC-400 is KOH solution with small amount of polymer added to it. The HC-400 consists of four integrated HC-100 fuel cells. First of all, two HC-100 fuel cell batteries are connected to parallel, and then two of these parallel connected couples are connected in series to form one compact fuel cell battery HC-400. In this project two of these were tested.

Figure 18. A fuel cell battery HC-400 (photo: RE program)

The alkaline fuel cells differed from each other by the colour of their coating material. The other AFC had a clear transparent coating plastic, and the other one had darkened transparent plastic coating. In each HC-100 fuel cell there is internal low pressure metal hydride storage unit, which can store 40 dm^3 of hydrogen. This corresponds about 80 Wh of electrical power (Oy Hydrocell Ltd., 2005). HC-400 unit can thus contain 160 litres of hydrogen without an external hydrogen container. The unit is also equipped with 0,8/14V voltage control system which prevents the cell from an overload. There is also a small water container, which allows the fuel cell to work properly even if the amount of water produced from the chemical reactions is not at equilibrium with the amount of water evaporated from the cell.

The fuel cell takes the oxygen needed for the chemical reactions form the air through the vent hole equipped with a fan. The fuel cell can be connected to a metal hydride container to increase the capacity. In case the external hydrogen storage is connected to the fuel cell, a pressure regulator is needed to lower the pressure to proper level for the operation.

The humidity level of the cell is important factor concerning the operation of the unit. Therefore the frequent observation of the display scale of the water container is necessary to avoid drying or flooding of the cell. According to the manufacturer, the fuel cell battery can be stored for years without losing the capacity. (Oy Hydrocell Ltd., 2005)

The electrolyte inside the cell is in gel form, which brings some benefits to functionality and safety of the cell. The hydrogen stored in the internal storage is stored at low pressure, about two bars over atmospheric pressure, which is approximately the same as used in passenger car tyres. (Oy Hydrocell Ltd 2005.) Technical information of the HC-400 fuel cell is given in the table 9.

Table 9. Technical information of the HC-400 alkaline fuel cell (Oy Hydrocell Ltd., 2006b).

Parameter	Value
Output voltages for different configurations (V)	14, 28, 42 or 56
Output current	8 A (12 V)
Internal capacity	28 Ah (12 V)
Weight	6 kg
Length	440 mm

5.2.7 Measuring equipment and fittings

The hydrogen assembly included also various measuring equipment and fittings. Most of the hoses used in the assembly were 6.3 mm Hutchinson LP gas hose (20 bar, -30°C / + 70°C EN 559 2004). The measuring equipment assembly built for fuel cell testing included hydrogen mass flow meter and a pressure meter (figure 19). The same assembly can also be used alternatively between the electrolyser and the metal hydride storage.

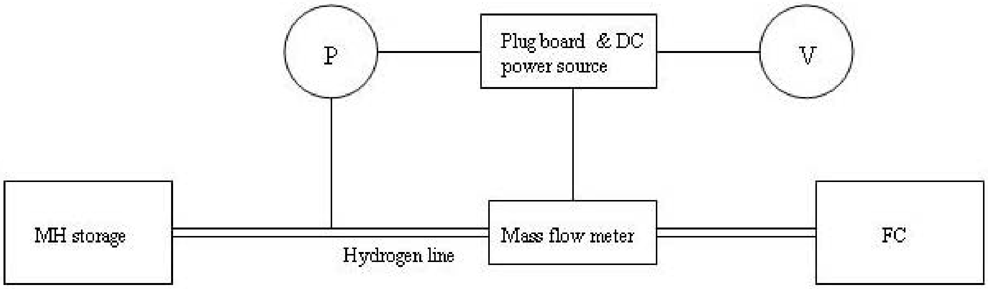

Figure 19. Schematic picture of the hose and metering assembly.

Output voltage of the pressure meter was dependent on the pressure changes in the hose, and thus an external voltmeter and a calibration curve was used to collect the pressure values during the tests. The pressure meter, which was valid for the pressures between 0 and 10 bars, needed an external power source as well. The relation between the voltage display and the pressure (figure 20) was determined by using the parameters given by a pressure meter manufacturer (10 bar = 5 V) and those found in own experiments (1 bar = 0.2362 V). An assumption was that the relation between pressure and voltage measured is linear up to 10 bars.

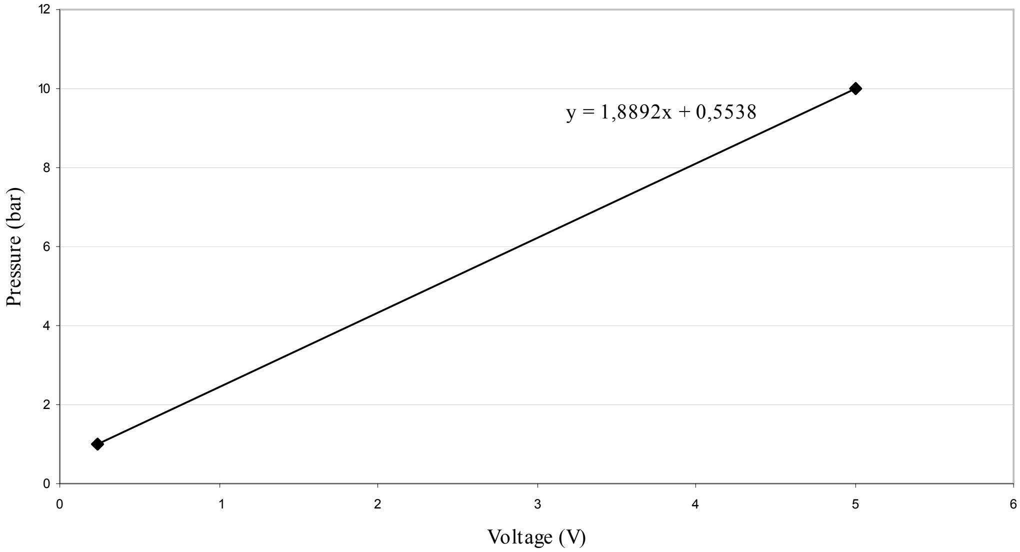

Figure 20. External pressure meter: The relation between the voltage and the pressure. Parameters defining the relation are: 1 bar = 0.2362 V and 10 bar = 5 V.

Plug board was built in the university by David Agar for the purposes of this project to connect the flow meter, pressure meter and their power sources together with external volt meter (figure 21). The external leak detector was included to the equipment to detect possible leaks. The display of the detector showed H_2 concentration in air with an accuracy of 1 ppm. The detector alarmed when the concentration of H_2 in air exceeded 1000 ppm.

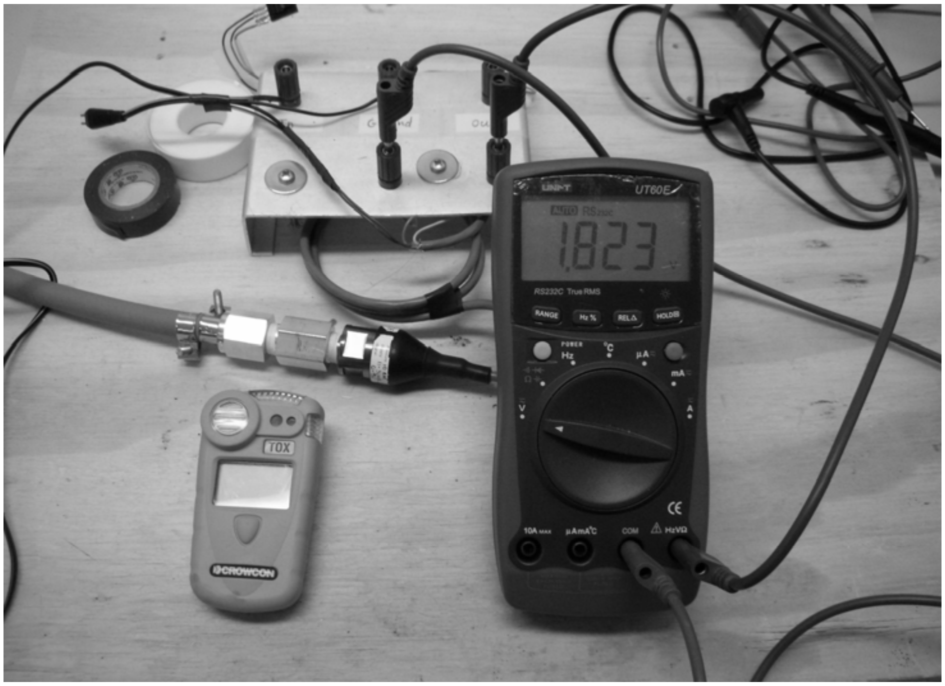

Figure 21. Some measuring equipment in the testing laboratory: portable leak detector, external pressure meter, plug board and a voltmeter.

6 RESEARH TARGETS

6.1 Background of this thesis project

As mentioned before, the aim of the whole project was to build and test a stand-alone energy system based on renewable energy sources, which produces hydrogen to be used as a fuel for a H_2 vehicle. The system located in the University facilities would act as a demonstration project for students and provide long-term experience on the system performance. This thesis is not a complete analysis of the whole system, but it concentrates on the initial testing and characterization of the components it includes. The components under research here are the ones belonging to fuel production, storage, utilization and to the test vehicle.

In Finland, the development work concerning alternative fuels is mainly connected to biofuels such as biodiesel, biogas and ethanol. To date, a concept of hydrogen vehicle including local refuelling station has gained little attention. Even though hydrogen vehicles are probably not penetrating the market within the nearest decade, the practical research work is necessary to start as soon as possible. Globally, there has been research on H_2 vehicles for decades, but the cold climate makes Finland a special testing ground. New fuel and a refuelling station have to prove their functionality through the cold and dark period of the year as well.

In May 2005, the first meeting at the University of Jyväskylä concerning this project induced a four-page list of different topics to put under research. Some points of them would require years of study just by answering to a question like long term driving experience on low temperatures at Finnish winter condition. Some questions were simpler, like determining the temperature pressure relation data for a single metal hydride container. Some were questions which appeared to be simple but which turned out to be impossible to measure or they would take too much money to buy measuring equipment. One of these so-called simple questions was to determine the purity of the hydrogen gas when produced by electrolysis and determining the purity again when releasing the stored hydrogen from the hydride container. So it was obvious since the beginning that there was to be a huge amount of topics left for the future.

Component characterization is an important part of this kind of project, because when each component is tested individually and separated from the system, it is possible to see the weak parts of this long energy chain. One can optimize the whole system by improving first the weakest part, then the second weakest etc. In this project, because it is a new project and all the premises have to be built by researchers and technical staff of the university, the importance of primary testing is emphasized. Individual component testing also provides a basis for the system testing as a whole in the future.

6.2 Target descriptions

This project consisted of four research problems:

1. Driving range of the vehicle
2. Metal hydride container and pressure regulator characterization
3. Hydrogen assembly leak tests
4. Electrolyser testing and characterization.

In this work, the primary aim was to find out how long distance one can drive with a certain amount of self-made and pollution-free hydrogen fuel. Because the vehicle uses fuel cells and battery unit as a power source, the contribution of the fuel cells and a battery unit separately was to be found out. First the driving range was measured by using the loaded batteries only and then the influence of one container and a fuel cell was to be found out. Within these tests, the knowledge about battery voltage behaviour during driving and hybridization degree of the vehicle was included to the analysis.

It is known, that hydrogen pressure inside the metal hydride varies a lot with the outside temperature and also during filling and discharging of the container. Within this work, the affect of temperature changes in the hydrogen pressure inside a container was examined and compared to the graph sent by metal hydride supplier (attachment 1). The temperature and pressure characteristics of the containers are important considering the driving conditions on-board a vehicle during the wintertime. The freezing of the metal hydride container can in practice prohibit the hydrogen flow to the fuel cell. By observing the pressure changes inside the container, an estimate of safe outside temperatures for the operation can be made. A component between the hydride and the FC, pressure regulator, has an important function to keep the pressure steady and correct for the FC despite the fluctuation of the pressure inside the container. In this work, it was also examined how the inlet pressure, which means the container pressure, affects the pressure after the regulator. It is a hypothesis, that pressure regulator is keeping the pressure constant and between the known limits. To put in another words, the aim of this test was to find out if there is any possible condition when the hydride container could not supply a proper pressure to the PEMFC.

The third research problem was dealing with hydrogen leaks. Due to the buoyancy of the fuel, hydrogen hose assembly must be carefully sealed. During this work, three separate leak tests were done to gain knowledge about the hydrogen leakage behaviour. The leakage rate in a sealed hose assembly was examined by using different types of connectors. Also knowledge about leak rate increase due to higher pressures in the hose assembly was one of the targets for examination.

The last of the four research problem was about fuel production. Hydrogen production is one of the key elements concerning the whole refuelling facility. Within this work, it was examined, what is the production rate of hydrogen and does it stay constant during the production. Also the reliability of the process in general, including the inverter and the batteries connected to it, was observed.

All these questions mentioned above are factors concerning the functioning of a test vehicle in practice. In the next chapter the methods and the testing procedures are described in detail.

7 RESEARCH METHODS

Working with this thesis project began already on 31st March 2005 by installing the solar PV panels, wind turbine and the equipment belonging to them to the roof of the physical chemistry building. The setting up of the test facility was done by laboratory engineer Arjo Heinsola, me and my fellow student Pasi Paananen. The tests made later to the components of this project are explained in this chapter, by describing also the surrounding conditions and measuring equipment which were used, as accurately as possible. The results of each test are explained separately in the chapter 8. The tests are explained in the same order as they were mentioned in the target description section.

7.1 Drive tests and fuel cells

7.1.1 Drive tests with batteries

The aim of the drive tests was first to measure the driving distance with fully loaded batteries only, and then later on, when the fuel cells were successfully tested inside the laboratory, measure the contribution of fuel cells to the driving range. Within the battery tests also the behaviour of the battery voltage during the driving was monitored. There were two separate drive tests made with the batteries only. Batteries were loaded with grid power for convenience. The dates to the tests were 2nd May and 10th May 2006. The tests are numbered as drive test no 1 and no 2 for clearance. In addition to this primary research, a secondary unanalyzed research material collected before the start of this project is used to determine the power consumption of the vehicle at different driving speeds.

Drive test no 1 on 2nd May 2006 was made to measure the driving range of the vehicle with the fully charged batteries and with no fuel cell. The test was made at Ylistö test track near Jyväskylä University campus area. The test track was 1.1 km long paved road around the local science park buildings and there were no huge slopes along the track. The test was done by two people, the other one driving the vehicle and the other helping with the necessary arrangements during the test. A fellow student Mikko Lommi helped with the necessary arrangements during the test e.g. timekeeping and driving the other car used for towing. H_2 car batteries were charged full before the test and the car was towed to testing environment. Car was driven along the test track until the batteries were dead. At every lap the voltage of both batteries and lap time were recorded manually. The external multimeter was connected to the lap top computer to collect the CSV data during the drive. The overall distance was measured by calibrated bicycle trip-meter and the car was towed back to car park. The weather conditions during the test no 1 were sunny, dry and moderately warm, approximately +15 °C.

Test drive no 2 was done on 9th May 2006. The aim of this test was to make the knowledge about the driving range only with the batteries more reliable. The test track and the arrangements for the test was made exactly the same as in the first drive test, except this time the data was not collected with the external multimeter, because it obviously gave false data to the computer.

In addition to these battery test drives, the car was tested outside the test track. The aim of these tests was to collect information about the car performance in winter condition on public roads. Also rough knowledge about its driving range and top speed was the topics of interest. Unfortunately, it was not possible to connect the speed sensor of the multimeter, so the top speed figures must be based on a rough estimate. For every test drive a driving diary form (attachment 2) was used to describe the battery and weather conditions and to make other notes as well. There were two types of tyres used in these driving tests. The other ones were for the winter use and they were equipped with studs. The summer tyres were a bit longer in diameter than the winter tyres.

7.1.2 PEM fuel cell primary tests

The testing of the fuel cells began in the laboratory. The reason for this was to gather information of the functioning of the FCs to understand better the results of tests performed inside the car and during driving. Also the safe and leak proof operation was to be confirmed before taking a hydrogen fuelled FC to the closed vehicle. After the first tests, their contribution to the overall driving range could be evaluated.

The experiments started with the PEM fuel cell on 31st March 2006 and continued on 5th April 2006. The aims of these tests were to ensure the safe operation of the fuel cell and to ensure the correct functioning of the FC. At first the container 014-0302 was used at room temperature to supply the hydrogen to the FC. The charge level of the internal battery of the fuel cell was monitored during the testing.

On the other testing day, the container was put to a kettle full of hot water (about 50 °C). The temperature of the water, pressure reading from the external pressure meter and the pressure gauge of the container was monitored during this primary test. On these tests the hydrogen line with the pressure meter was used. This time the container used was 013-0202 and its initial pressure reading before heating was 4.3 bars at room temperature.

7.1.3 Alkaline fuel cell primary tests

The primary tests for the alkaline fuel cells were done at the laboratory on 7th April 2006. The aims of the tests were similar to the aims of PEM testing. Crowcon leak detector was used to detect possible leaks in the system. Due to the nature of the test as an initial procedure, the mass flow meter and the pressure meter was left away from the assembly, and the hydrogen was fed to the fuel cell straight with only a pressure regulator and 0.7 m hose between the container and the alkaline FC. Both alkaline FCs were tested with the same container. Both the hose assembly and the FC internal hydrogen line were monitored for possible leaks. The power produced was measured with external voltmeter. The functioning of all four individual cells in each FC unit was separately monitored and tested.

7.2 Metal hydride containers and the pressure regulator

7.2.1 Temperature – pressure test for HC-MH1200

The container test was done to the container 013-0202 and the test took place on 3rd May 2006 in the testing laboratory. The aim of this test was to gain information on the temperature and pressure relation in the container loaded with hydrogen. The pressure regulator was not used between the container and the pressure meter during this test.

The test was done in a fume chamber to avoid any possible hydrogen leaks to accumulate to the upper parts of the laboratory room. The testing set-up (figure 22) consisted of electric hot plate, metallic container for the water bath, pressure meter and its power source, voltmeter and a thermometer. Empty cucumber can was used as the container to the water bath. The hydrogen container was put to the water bath and the water was cooled down to freezing point with snow and ice. Snow and ice was added until the temperature had lowered to 0°C. At this point the hose with the external pressure meter was not connected. The cooling period took one hour and the pressure reading in the container gauge had dropped from 5 bars to 1.6 bars. Now the external pressure meter was connected to the container and the heating of water bath began. The external pressure meter could not be connected before the cooling, because in that case the initial pressure of the container (5 bars) would have remained inside the hose because of the one way connector. The temperature of the water bath was slowly raised from 0 to 59 °C and the readings of the temperature and container pressure were recorded manually. The external pressure meter needed the voltmeter to get the pressure readings recorded. The voltage readings were converted later to bars according to the equation shown in figure 20.

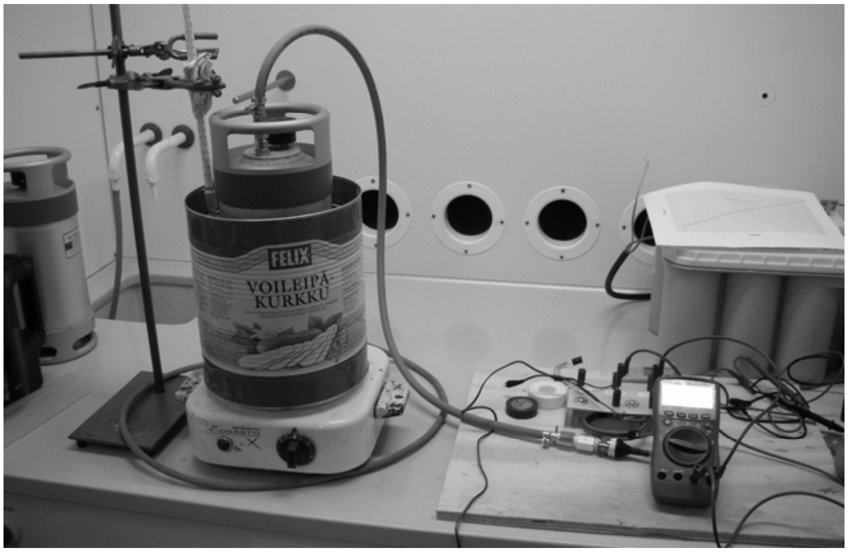

Figure 22. The setup for temperature and pressure test on HC-MH1200.

During the test, leak detector was used intensively to detect any possible H_2 leaks. Heating of the water to its maximum temperature took 3.5 hours making the average rate of temperature increase about 0.28 °C/min. The heating was done slowly in order to let the heat conduct into the inner parts of the container from the water bath. The test was continued after the heating was stopped to collect the readings from the cooling period as well. The cooling process was accelerated by adding cold water to the water bath and the system was monitored for two hours after the maximum temperature point. The hose with external pressure meter was left connected to the container after removing it from water bath.

7.2.2 Pressure regulator characterization

According to the H-CORE-500-2 manual, the inlet H_2 pressure of the fuel cell should be between 2.1 and 4.1 bars. The aim in characterizing the pressure regulator was to see if the outlet pressure from the H_2 container could be kept within these limits in any pressure value inside the container. The test was done on 11th April 2006 at the fume chamber of the laboratory. The method used was similar to the method used at the container temperature and pressure relationship test described above. The container 013-0202 was used as a H_2 storage unit. The pressure regulator used in this test was Rectus 3bar/15bar and it was connected by a quick lock connector to the container.

The testing set-up consisted of an electric hot plate, metallic container for the water bath, pressure meter and its power source, voltmeter and a thermometer. The test proceeded by first cooling down the system with ice cubes to +1 °C, and then warming up slowly the water bath with the electric hot plate and monitoring the pressure gauge of the container and the external pressure meter, which showed a pressure value after the regulator component. Possible leaks were checked from time to time with the portable Crowcon H_2 detector. The hotplate used was 415 W Ikamag RH with the plate diameter of 10 cm, which is very close to the diameter value of the container. The conduction of heat generated by the hotplate straight to the bottom of the container through the metallic bottom of the water bath was prevented with a porous metallic wire rack. The rack was placed at the bottom of the water bath, and the container laid on it in a way that there was a 2 cm layer of water between the hydrogen container bottom and the water bath bottom. This contributed the even distribution of heat from the water to the container unit.

The heating period of this test took for four hours and after the heating had stopped the system was monitored as it was for one hour to observe the effect of cooling and to record the maximum pressure point at the hose. The heating had to be stopped because the water started to boil beneath the bottom of the container and the amount of uneven heat distribution would have been increasing strongly. The maximum temperature reached during the test was +63 °C and the average rate of heating was 0.23 °C/min. The cooling period of this test took half an hour and the pressure reading in the container gauge had dropped from 3.9 bars to 1.2 bars during that time.

7.3 Leak tests for the hydrogen line

The leak tests were done during the spring 2006 in the same fume chamber as used in other characterization tests in the testing laboratory. The aims of these tests were to ensure the safe and reliable proceeding of further tests on the system. The knowledge about the rate of self leakage of a sealed hose assembly and the leakage behaviour during the increased pressure of the system was the points of interest. On all the tests, the pressure generated by other tests was utilized as initial pressure for the leak test assembly.

First leak test to the hydrogen line was done on 31st March 2006 by using the MH container 014-0302 as a source of hydrogen. The test was done to the hydrogen test board with mass flow meter and a pressure meter described earlier in the chapter 5.2.7. The test was a preceding test to the PEM fuel cell testing to be arranged later. The hydrogen leak detector was used again to detect possible leaks. To tighten up the connections between different measuring equipment and hoses, pipeline tape was used on the threads of the connectors. Before this test there had been couple of quick tests for the H_2 leakage with the results of rapid leaks. These quick leak tests resulted the purchasing of new connectors of a different type. On this and the following test the new connectors were used. The connectors were supplied by Oy HVF ltd Helsinki, Finland.

The second leak test was done between 11th April and 2nd May 2006. After the characterization test for pressure regulator, the hose was disconnected from the H_2 container and left pressurized. The pressure change inside the hose was monitored for about three weeks with pressure meter. There was no mass flow meter connected to this set-up. There was only a single 6.3 mm hose with the pressure regulator equipped with one-way quick lock connector on the other end and the pressure meter on the other.

Third self leakage test for a sealed hose was done between 4th May and 19th May 2006. The aim of this one was to test the influence of different type of connector to the self leakage rate. This time the test assembly consisted of the same 6.3 mm hose and the same pressure meter, but the other end of the hose was sealed by a different connector. The connector used here was a simple quick lock connector without a pressure regulating function. This test was done after the container temperature and pressure characterization test and the set-up was monitored for a time period of two weeks. Like in the previous leak test, the timekeeping and monitoring of the pressure drop started when the hose was disconnected from the container.

7.4 Electrolyser test

The initial test for the electrolyser HG500 was done in two days between 8th and 9th February 2006. The aim of this test was to gain information on how reliably the electrolyser works, is there any leak when filling the container, and how fast and to what pressure the storage unit could be filled. The power to the electrolysis was derived from the battery unit which was charged full by the wind turbine and the PV panels. The battery unit was not charged during the test.

Before the generation process was started the water container was filled with de-ionized water. The H_2 outlet of the generator was connected to the dryer unit, which was in turn connected to the hydride container with a rubber hose and a quick lock connector. There were no pressure regulator between the generator and the container. Between the battery and the generator, inverter was used to alter the voltage appropriate for the electrolyser. During the process there was no data collection via the electrolyser data port due to lack of equipment. The battery voltage was measured with external voltmeter. The data, H_2 generation rate and the gas outlet pressure, shown in the display screen of the generator, was collected manually. The consumption of water was not observed during this initial test. The generator was started on afternoon and during the night it was left to idle mode. After an idle period of 15 hours the generation was continued again in the next morning.

On the second day morning the inverter went out of order and the test was carried on with grid power to get the maximum possible pressure to the container. The storage unit was not connected to the generator until the flow of hydrogen had started and the extra air from the hose assembly had escaped. The container was connected to generator at 15:38 pm. The container used in this test was the one with serial number 014-0302. The initial voltage of the battery unit was 26.1 V and after the initiation of H_2 production the generator outlet pressure started to climb from 0.5 bars. At the point when the container was connected to the system the initial pressure reading in the container gauge was 1.2 bars. The inlet water was led to the generator without any pumping system. The water container was at about one meter height above the unit and the water flowed freely to the unit via transparent plastic pipe. The generator outlet pressure reached its maximum at 13:03 pm on a second testing day. After three hours from that the H_2 generation was stopped. The readings from the container gauge were monitored only from time to time to see if the gauge readings followed the pressure readings in the generator display.

8 RESULTS

8.1 Drive test results

The drive tests were the first research problem mentioned in the list of targets, and therefore they can be considered as the main results as well. The drive tests were made to the battery only powered test vehicle on 2nd May 2006 and 10th May 2006. The tests are numbered as drive test number 1 and 2 for clearance, and the results of both are represented separately.

The aim of the drive test number 1 was to measure the driving range with full batteries and without any hydrogen power. The test resulted with a maximum driving range of 7.62 km (figure 23). The battery voltage level decreased rapidly during the seventh lap. The driving battery voltage dropped from initial value of 25.72 V finally to 16.62V. The car exhausted first time during the lap number 7 when the battery voltage was 21.3 V. Battery replenished its voltage quickly during a short brake of few minutes and the test could go on again. The fifth exhaustion was the final one and the car was removed from the track by towing it back to the car park.

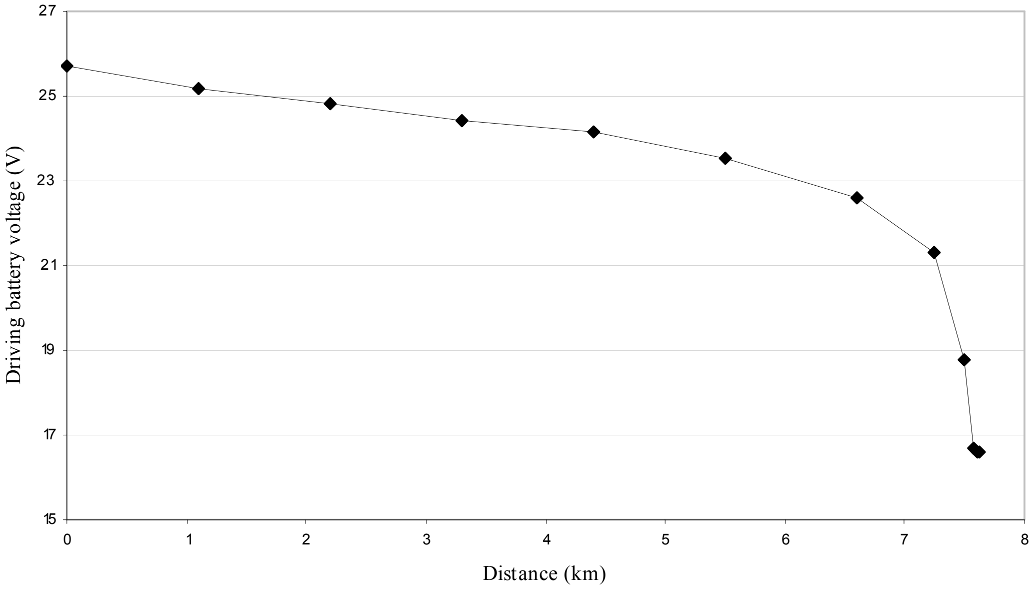

Figure 23. Drive test number one. Driving battery pole voltage in the function of distance driven. The test was continued until the batteries were completely exhausted.

The average lap speed, which was calculated from lap distance and time, was 26.1 km/h at the highest. The momentary top speed could not be recorded because the speed sensor for the external multimeter was not connected due to technical problems. The battery voltage was measured to improve knowledge about the relationship of the battery charge level and the battery pole voltage. According to the battery manufacturer, there is no reliable relation between the voltage and the charge level of the batteries (Suhonen, phone conversation, 10th April 2006)

This test resulted that the vehicle can function at its full power up to certain point very close to the battery exhaustion. At the end of the sixth lap there were no crucial signs of exhaustion and after that the exhaustion happens quickly. For these driving ranges the lights control battery seems to have hardly enough capacity, because the headlights were switched on during the lap number three and there was a significant decrease in the pole voltage of the battery. The voltage of the battery for ancillaries dropped from initial value of 11.57 to 10.16 V within approximately four laps.

The multimeter gave inappropriate figures from the beginning of the test. For example, the power value shown by the meter was 42.43 kW when the gear was put on reverse and the car was not moving. This is obviously impossible with the two motors of 6 kW each and without any movement. The voltage of approximately 58 V indicates wrong calibration values inside the multimeter, real voltage being less than half from that. The CSV data of the whole test drive was collected on computer, but it is left unanalyzed due to its misleading nature. The data collected did not follow the determination of electrical power ($P = U \times I$). Recalibration of the multimeter could not be done due to lack of technical assistance.

Power consumption of the vehicle at different driving speeds was determined from the data measured before this project with the same multimeter and test vehicle. The data on which the determination is based was yet unanalyzed CSV data received from a developer of the multimeter. Five different speed values were chosen for analysis and the correspondent power values were collected for closer analysis. At a certain speed, there were lots of variations in power consumption due to acceleration, deceleration and upward or downward slopes. From the data chosen for analysis, an arithmetic average and standard deviation was calculated. Once this has been done, all individual values differing more than an amount of standard deviation from the average was left out from the final analysis. From the remaining values a new arithmetic average was calculated (figure 24). The exception was power values measured at driving speed 25 km/h. There were only four values measured, of which two values were left out from analysis, since their most obviously were values measured during strong deceleration period. The average of the two remaining figures (5.15 kW) was included. The resulted values can be used to estimate the vehicle performance and driving range with fuel cells.

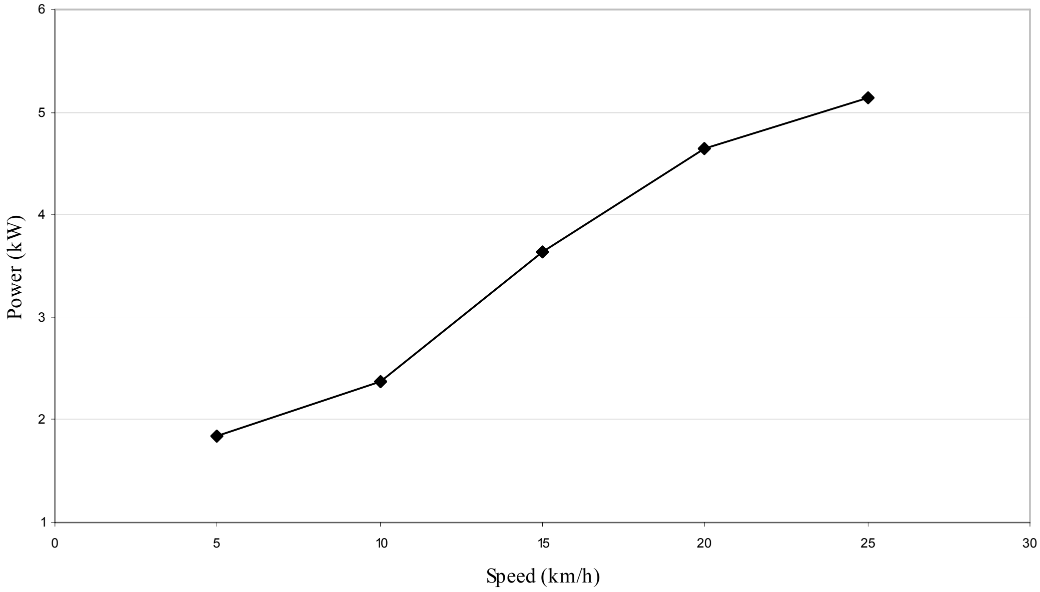

Figure 24. Fantasia power consumption at different speed values. (Based on data measured by R. Gyllenberg 2006)

For the second drive test, the overall distance resulted 2.2 km. The test ended after the second lap with the breakdown of the shaft welds near the right rear wheel of the vehicle. The driving range limit due to the battery exhaustion remained thus a secret. The 6 mm bolt jointing the two parts of the rear shaft together had been cracked for several times before these tests and after that the bolt had been changed to harder steel bolt. The bolt was at its place at this time, but the welds near it were damaged.

This time the top average speed during the lap was 25.7 km/h, which is similar to the results of the first test drive. Before the interruption, the rapid decrease in battery voltage is seen during the first lap. The voltage dropped in driving battery unit from initial value of 27.02 V to 25.74 V during one lap. In the battery for ancillaries, the voltage dropped from initial value of 12.71 V to 12.51 V. After the test car was towed back to car park by another vehicle. The voltage drop during the first lap was much greater than in the first test, which is probably due to overloading of the battery unit before the test. The overloading was due to malfunctions in the battery charger before.

General driving performance of the vehicle was observed on several testing trips. On 19th November the car was driven for about 500 metres in a temperature of -2 °C. During that very short driving period the pressure of the hydride container had dropped from 3 bars to 2.5 bars. The container was kept inside the car but there were no fuel cell to produce power. This indicates the affect of temperature on the H_2 pressure, which has to be taken into account in further tests and development work.

The similar effect was noticed on the 4 km test trip made on 18th December 2006 on the public roads near the university car park, which was the holding place for the test vehicle during the project. The container kept inside the car had initial pressure of 2 bars inside the parking lot and it soon dropped to 1.5 bars when the car was took outside. The weather on that day was cold and sunny winter day with the temperature of –13 °C. The aim of that trip was to test the general driving properties of the vehicle on public roads covered with snow and ice. The visibility from the car and the drive control devices were good and well functioning, but the convenience of driving was distracted by the noise and smell of burnt material coming from the electric motors. The tyres used were with studs but the rear of the car was so light, that acceleration on the icy crossings was difficult. On the snowy local roads, the muscle work of the researchers was needed on some occasions to get the car back to move. On that very trip, the top speed was also measured by driving at same speed with another car behind the test vehicle. The measured top speed was 35 km/h on a flat road. This short trip also revealed the short distance between the bottom of the car and the road. Due to the short diameter of winter tyres, there were difficulties to drive over the small piles of snow and ice on parking lots and crossings. During the end of the trip there was a clear decrease in the battery charge level which was seen as lowered maximum speed.

8.2 PEMFC test results

PEMFC was tested with the hydrogen container and a pressure regulator at the room temperature. No electrical power was measured. Later, the container was put into hot water and so the pressure inside the container increased to about 13 bars. From the equation, which is represented later in the chapter 8.5, this corresponds to the hose pressure of 2.9 bars. This value clearly fulfils the PEMFC inlet pressure requirements. No electrical power was measured on this time either. Few days later, the experiments with PEMFC ended with no results because of component damage inside the fuel cell.

For the FC performance is crucial part of this whole scheme, some estimates were made based on the PEMFC efficiency, power consumption of the vehicle and the available hydrogen storage capacity. The power consumption analysis of the vehicle showed, that the PEMFC of 500 W alone is too small to provide power to the test vehicle even at very low driving speeds. The power requirement for 5 km/h driving speed was 1.85 kW (figure 24). By using the equation represented in the chapter 4.3.3, the hybridisation degree of the vehicle becomes 0.27. By calculating the same degree to other selected driving speeds (table 10), it is noticed that the vehicle performance approaches the performance of an electric car (hybridisation degree = 0). The fuel cell will only provide minor share of the power requirements and act as a battery charger during driving rather than a provider of motive power.

Table 10. Hybridization degree (= p_{gen}/p_{tract}) of the test vehicle Fantasia with PEMFC at selected driving speeds. Hybridisation degree for battery electric vehicle = 0 and for FCV without electricity storage on-board = 1.

Driving Speed (km/h)	Hybridisation degree
5	0.27
10	0.21
15	0.14
20	0.11
25	0.10

The contribution of PEMFC to the overall driving range was estimated in the situation where the average speed of the vehicle would be 25 km/h. This corresponds to the average speed values during the practical drive tests and the power consumption of the vehicle is approximated to be 5.15 kW, according to the analysis represented in the previous chapter. One HC-MH1200 container holds 1200 litres (at STP) of hydrogen, which equals the mass of 0.107856 kg H_2. The energy content of this amount of fuel is 4.25 kWh by using HHV of pure hydrogen. According to the manufacturer, the efficiency of this fuel cell is 37 %, which means that the amount of energy converted by the PEMFC and which is available for the traction motor during driving is therefore 1.57 kWh. This corresponds a driving distance of 7.6 km, provided that the fuel cell and MH storage together with the battery unit are fast enough to produce the required amount of power to the traction motor.

8.3 AFC test results

Alkaline FC tests ended with the notice, that the AFCs can not be used inside the vehicle because of obvious H_2 leaks from the internal hydrogen pipelines of the cells. Both AFC were tested by connecting them to a metal hydride container at room temperature. The hose assembly proved leak proof, but the internal hydrogen lines connecting the four separate HC-100 cells were leaking in both alkaline fuel cells used. No electrical power was measured during tests. Fuel cells were sent back to the supplier for repair. Closer analysis for the AFC performance on-board the vehicle was not done, because the conversion efficiency of AFCs was not known and it was obvious that it would not have been suitable for providing even the same motive power to the vehicle than PEMFC.

8.4 Metal hydride container test results

The main result of the container test was the temperature – pressure relationship equation that is characteristic to the HC-MH1200 metal hydride container with certain amount of hydrogen inside (figure 25). The maximum pressure gained inside the container by heating was 17.25 bars according to the reading of the container gauge. The reading of the pressure meter showed 8.4 bars at that point. The mismatch of these figures depends on too narrow measuring range of the external pressure meter. According to the manufacturer, voltage of 5 V would be the maximum point, correspondent to the pressure of 10 bars. This experiment showed that the limit for the external pressure meter was 4.18 V and 8.45 bars. The analysis is therefore based on the pressure readings on the container gauge only.

As the test proceeded, the external pressure meter gave increasingly lower pressure figures than the container gauge. At the point when the water bath temperature was 43 °C, the difference of the two pressure readings was 2.84 bars. This gives a reason to put the linearity of the external pressure meters calibration curve under question.

By determining the equation of a polynomial trendline to the pressure data from the container gauge, the estimated pressure of the container at certain surface temperatures can be calculated. The equation can be represented as

$$p_c = 0{,}0029T^2 + 0{,}0772T + 1{,}6$$

where, T = H_2 container surface temperature

p_c = H_2 container pressure

This equation is most valid for the case when the container pressure at room temperature is 5 bars. Before the heating, when the value of 5 bars was measured, the surrounding temperature was 20.5 °C. During the heating, when the temperature of the water bath was correspondent to the temperature before the test, the container gauge showed a value of 4.3 bars. This indicates that due to continuous heating, the inner parts of the container were cooler than the water bath. The other proof of this phenomenon is that the container pressure reached its maximum 17.25 bars 12 minutes after the heating had stopped.

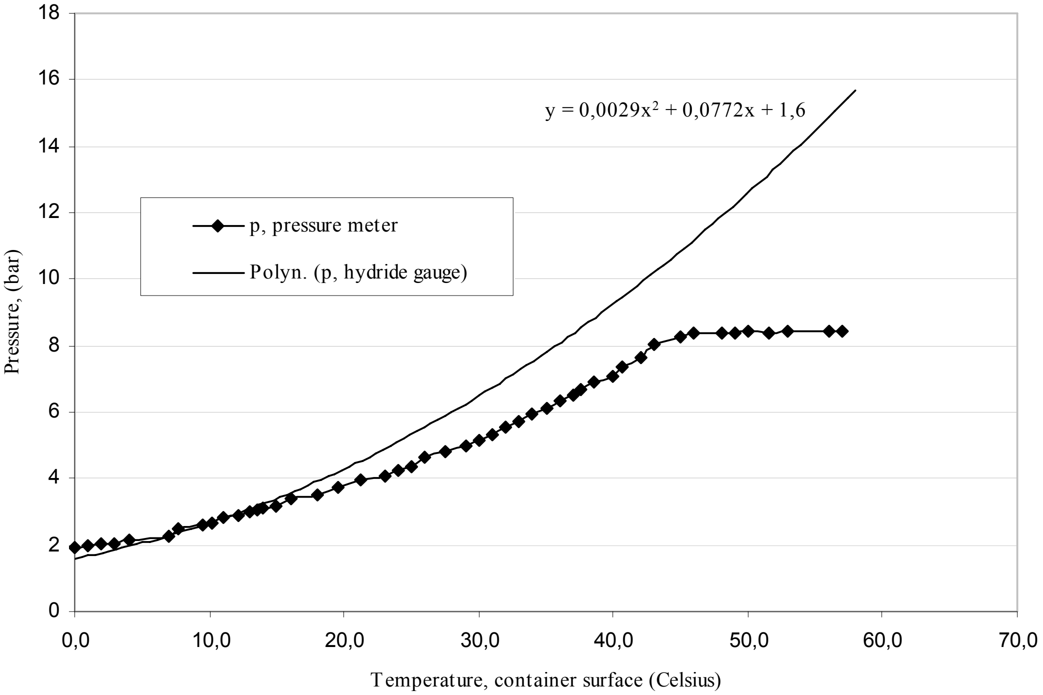

Figure 25. Metal hydride container pressure in the function of container surface temperature. The external pressure meter levels off between 8 and 9 bars and the readings have scientific value only up to that point.

The rate of temperature change during the test was 1.5 °C/min at the highest and it averaged below 0.5 °C/min (figure 26). Continuous line in the graph represents a moving average of the calculated temperature change rate values.

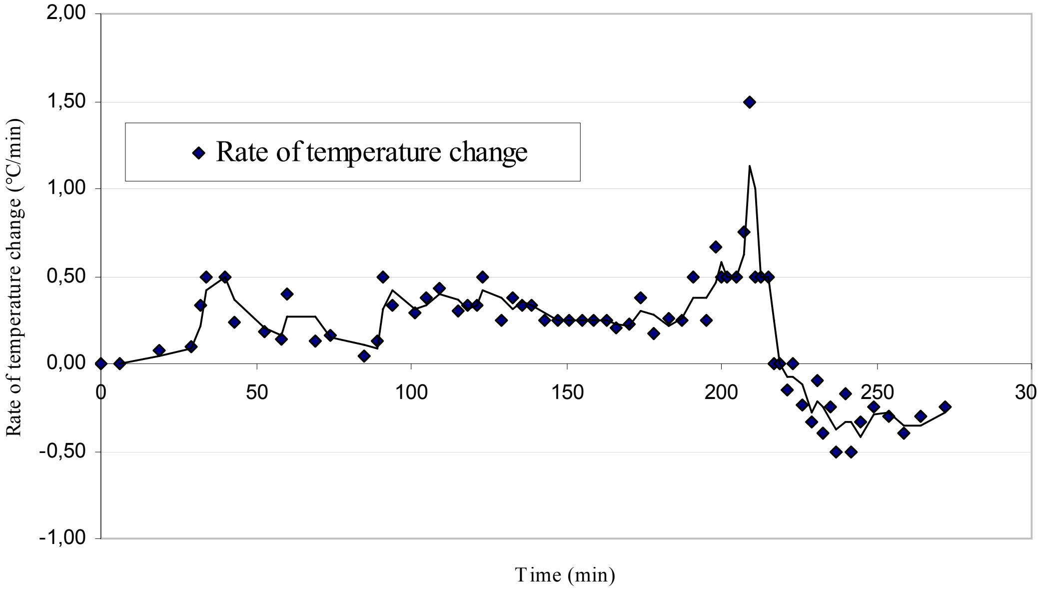

Figure 26. The temperature change rate during the hydrogen container test. On horizontal axis are minutes and the vertical axis describes temperature change rate in °C/min. The heating was stopped at T = 213 min.

The fact that the external pressure meter exceeded its measuring limits during the test was confirmed when the container was cooled back to room temperature. The meter started to show changes in the pressure values again when the container was lifted out of the water bath and the temperature started to decrease.

This experiment could also be used as the reliability test of the external pressure meter and its calibration curve. The reason for reading differences could be the voltage supplied to the meter by its power source. There were no detectable leaks during the experiments, which can indeed be considered as a positive test result. The pressure and temperature –dependence of the container is represented for clearance in a logarithmic scale, where the data measured follows the exponential trendline (figure 27).

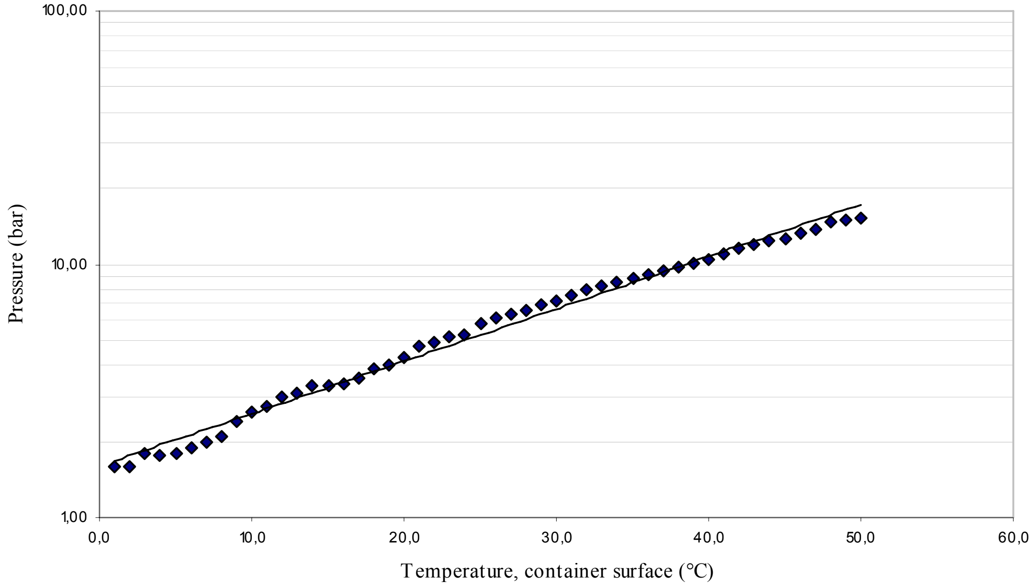

Figure 27. The temperature and pressure relation of the HC-MH1200 container represented on a logarithmic scale.

8.5 Pressure regulator characterization results

In addition to the container temperature and pressure characterization, the functionality and reliability of the pressure regulator was tested. The experiment showed that metal hydride container equipped with pressure reducer 3/15 bar is able to supply hydrogen at proper pressure to the fuel cell at surrounding temperatures between 10 °C and 63 °C. As mentioned before, the H_2 pressure reaching the PEMFC must be between 2.1 and 4.1 bars. The hydride pressure at room temperature (22 °C) was 3.9 bars before the test. Within the test it was noticed that at temperatures lower than 10 °C the pressure of the container is lower than the minimum limit set for the FC inlet pressure. The maximum limit of 4.1 bars for the hose pressure was not achieved during the heating. This results, that in all conditions the hose pressure would not exceed the maximum limit set for the FC inlet pressure. The climbing of the container pressure, which is the inlet pressure to the pressure regulator, together with the hose pressure is represented in the figure 28. The maximum hose pressure, reached at the water bath temperature of 63 °C, was 3.4 bars. The crossing point of the two lines indicates the point where the pressure regulator starts to lower down the inlet pressure.

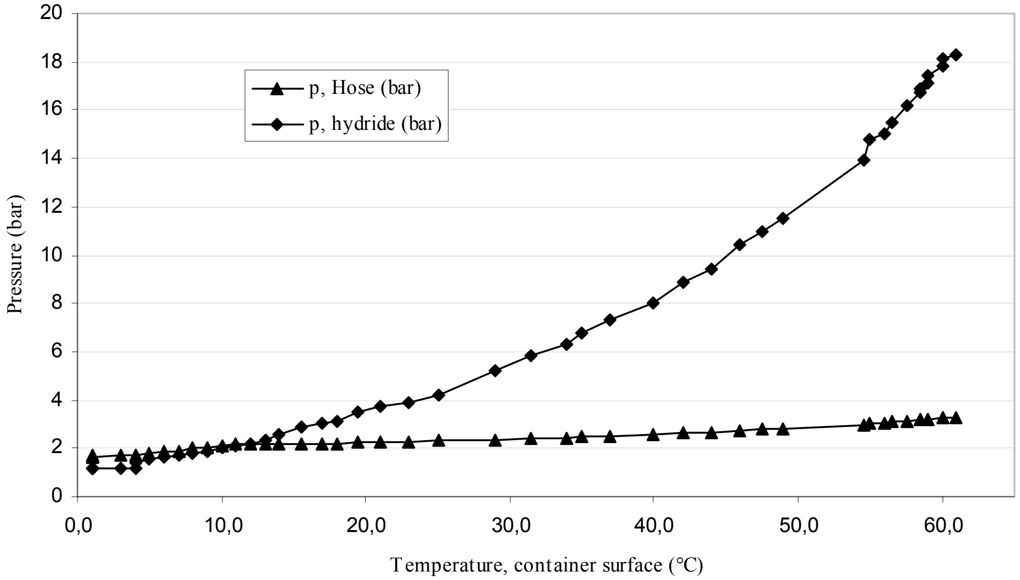

Figure 28. Container pressure and the hose pressure represented in the function of container surface temperature. The test resulted that the temperature inside the hose cannot rise too high for the PEMFC even in the case of high surrounding temperatures.

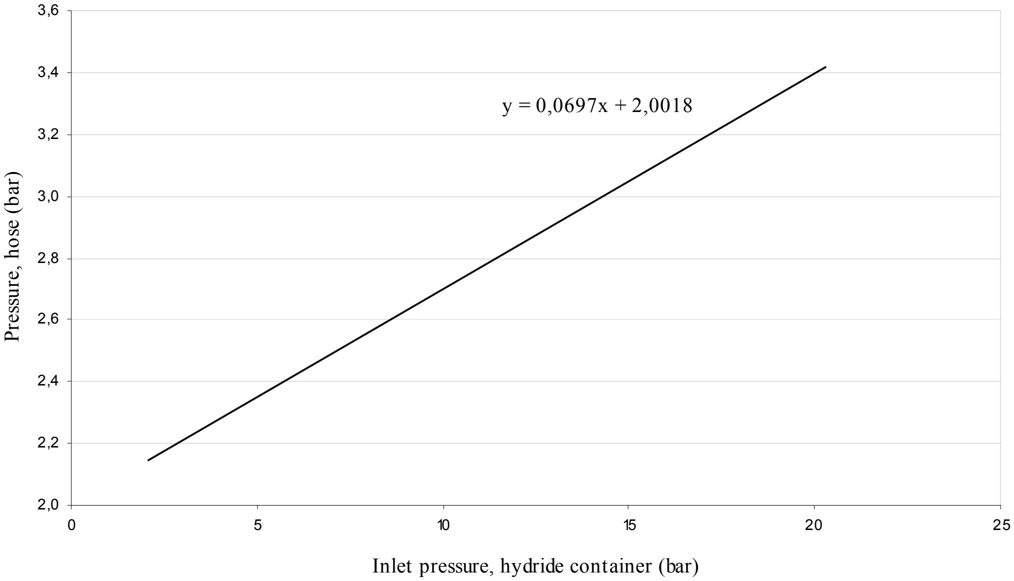

Figure 29. Hose pressure after the pressure regulator represented in the function of inlet pressure (= container pressure).

The pressure inside the hose (figure 29) in the function of the inlet pressure (= container pressure) can be calculated by the equation:

$$p_{hose} = 0{,}0697\, p_{inlet} + 2{,}0018$$

$$p_{inlet} \geq 2{,}1 \text{ bar}$$

The equation is valid only when the inlet pressure is above 2.1 bars. In the situation of this experiment, it means the container surface temperature of 10 °C. As mentioned in the chapter 5.2.3 the emergency valve of the container will open at the pressure of 30 bars. If this inlet pressure value is substituted to the equation represented above, the maximum possible value of pressure inside the hose is

$$p_{hose} = 0{,}0697 \times 30bar + 2{,}0018 = 4{,}09bar$$

8.6 Leak test results

At the first leak test done on 31st March 2006, the pressure inside the test system dropped from 2.29 bars to 1.84 bars in 15 minutes (figure 30). The test was done before the PEMFC testing to ensure the safe operation of hydrogen test board with a mass flow meter and a pressure meter.

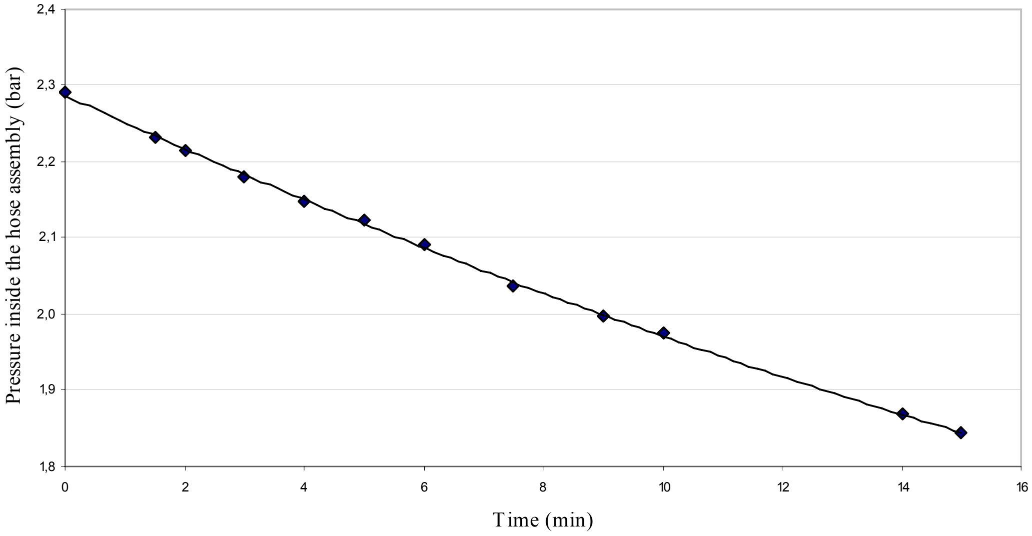

Figure 30. 1st leak test. The pressure inside a sealed hose assembly in the function of time. The rapid rate of leakage was due to the connections between the mass flow meter and a hose.

The initiating rate of pressure drop was -39.0 mbar / min. This figure dropped to -24.6 mbar / min during this short test as the overall H_2 pressure decreased. The test was quitted after 15 minutes due to rapid leakage. With the portable H_2 detector, the leak was located to the connections between the mass flow meter and the hose. The concentration of H_2 in the air near the leak point exceeded occasionally 1000 ppm, which is the concentration limit to the alarm sound of the portable detector.

The second leak test was done after the pressure regulator characterization test between 11th April and 2nd May 2006. The pressure dropped from 3.4 bar to 1.1 bar in 20.9 days (figure 31). In the beginning, rate of pressure drop was -0.84 mbar / min. This was significantly lower than within the first leak test.

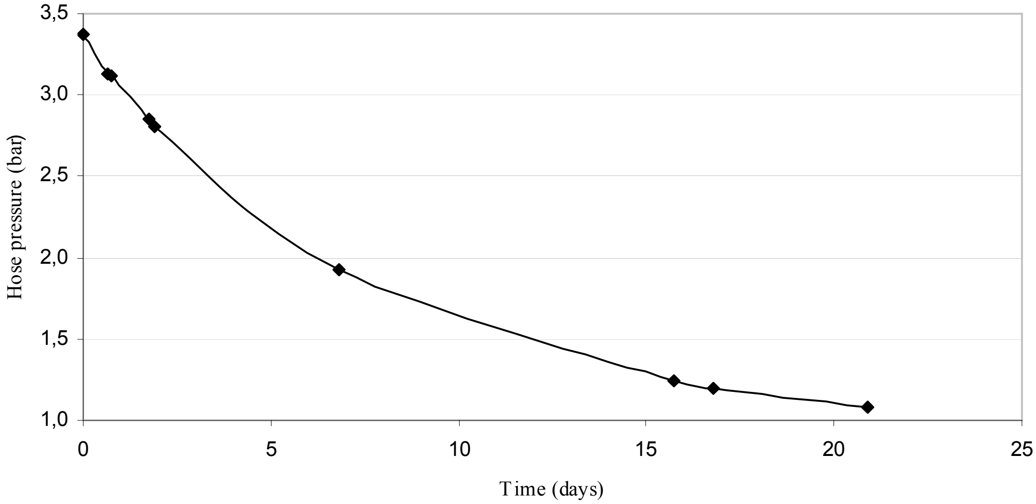

Figure 31. 2nd leak test, pressure inside the hose assembly in the function of time.

The portable H_2 detector could not detect any leaks anywhere in the H_2 hose assembly. For the leak detector, minimum detectable concentration is 1 ppm. According to the safety issues the test was successful, because the amount of leakage was almost negligible. One reason for the tightness of the system was that there was no mass flow meter connected to the setup. On the other hand, the situation is the same in practice when the setup is located in a moving vehicle. The data collected from this test gives us an estimate how much the hose line is leaking without H_2 consumption in FC for a certain pressure. The test showed also a rapid growth in the pressure drop rate when the system pressure increased.

Third leak test, where the influence of different type of connector to the self leakage was targeted, was done between 4th May and 19th May 2006. The pressure of a sealed hose containing hydrogen dropped from 3.99 bars to 1.02 bars in 15 days (figure 32). The initiating pressure drop rate was calculated to be -0.029 mbar / min. The reason for this small drop rate could be that the first readings were recorded only two hours and 10 minutes after the start, and the accuracy of the equipment is not exact enough for calculating the leak rate for such short period of time. This value has therefore been left out from the analysis. The drop rate was -0.39 mbar / min at a system pressure of 3.5 bars (figure 33).

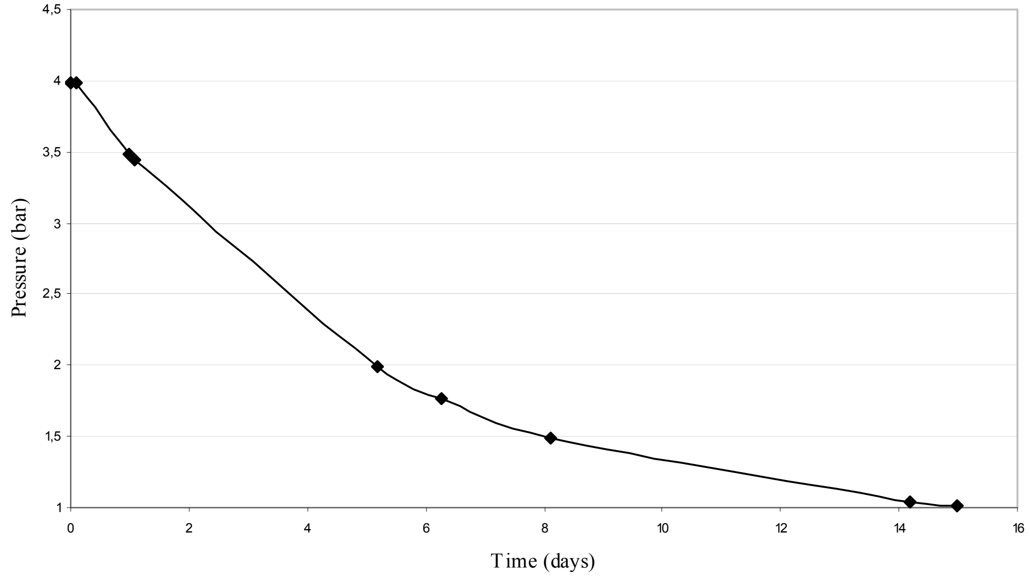

Figure 32. 3rd leak test, pressure inside the hose assembly in the function of time.

The figure shows that the leakage occurs more rapidly as the pressure inside the hose assembly increases. The rate of pressure loss was behaving similarly as in the 2nd leak test, and according to the safety matters the result can be considered again positive.

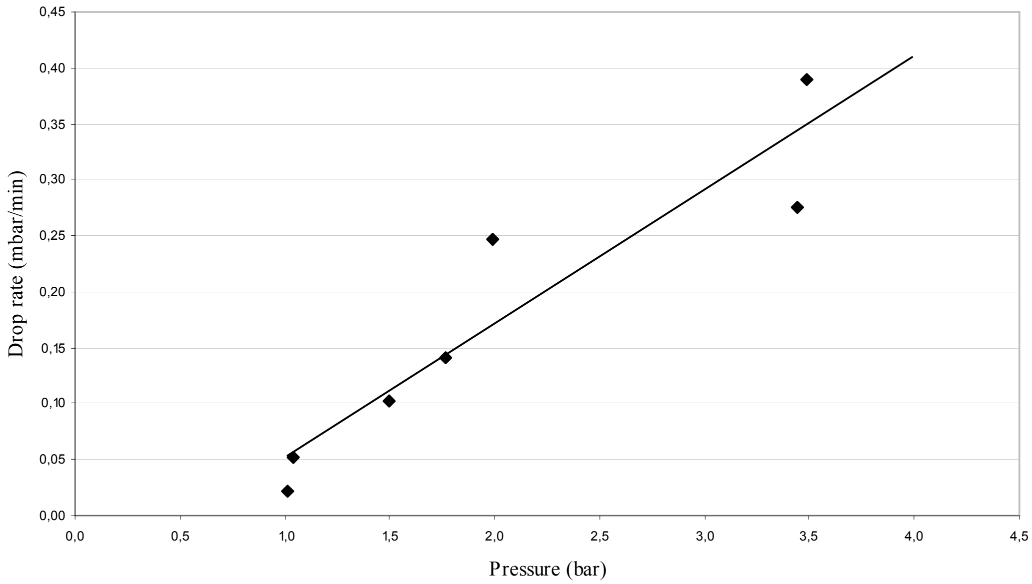

Figure 33. The pressure drop rate in the function of system pressure measured within a third leak test.

8.7 Electrolyser test results

The electrolyser test resulted that the rate of hydrogen production remained nearly constant until the pressure inside the container reached a pressure of 6.5 bars (figure 34). The production rate was approximately 530 cm^3 / min up to that pressure limit after which the rate dropped dramatically. The nominal pressure seen in the generator manual is six bars. At the beginning, the decrease in the rate of production was very rapid, almost vertical, but after couple of hours of operation the production rate started to decrease more slowly. The value of production rate seemed to approach a constant value (figure 35). The system seemed to move towards a balanced situation of system pressure of 6.7 bars and the production rate of approximately 200 cm^3 / min.

The maximum pressure that could be generated at room temperature to the metal hydride container 014-0302 was 6.7 bars according to the generator display. The container was filled in two time periods separated by the night time. Excluding the night time idle mode period the filling of the container took for 11 hours and 2 minutes. The overall time was 26 hours and 12 minutes.

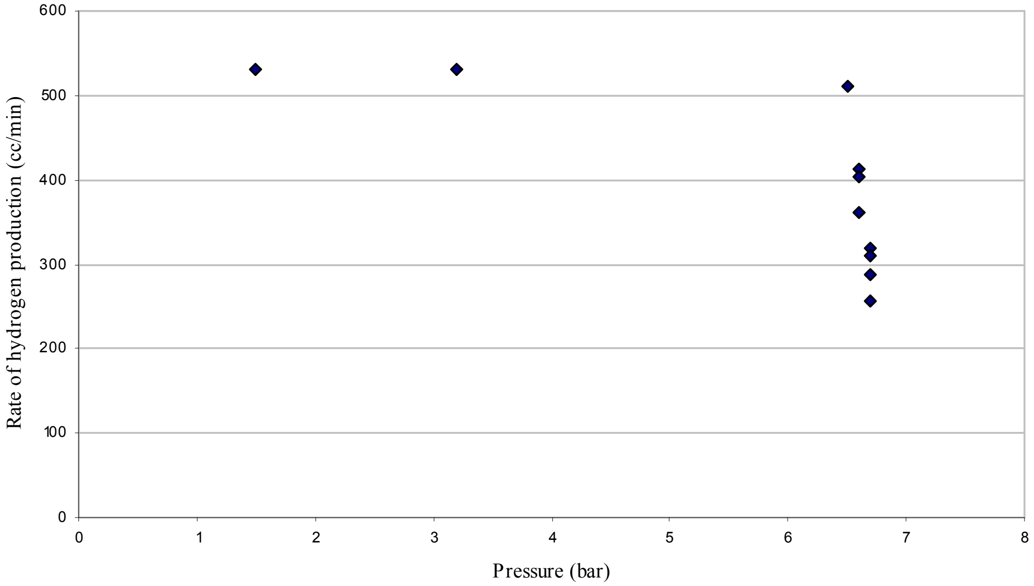

Figure 34. The rate of hydrogen production via electrolysis in the function of electrolyser pressure.

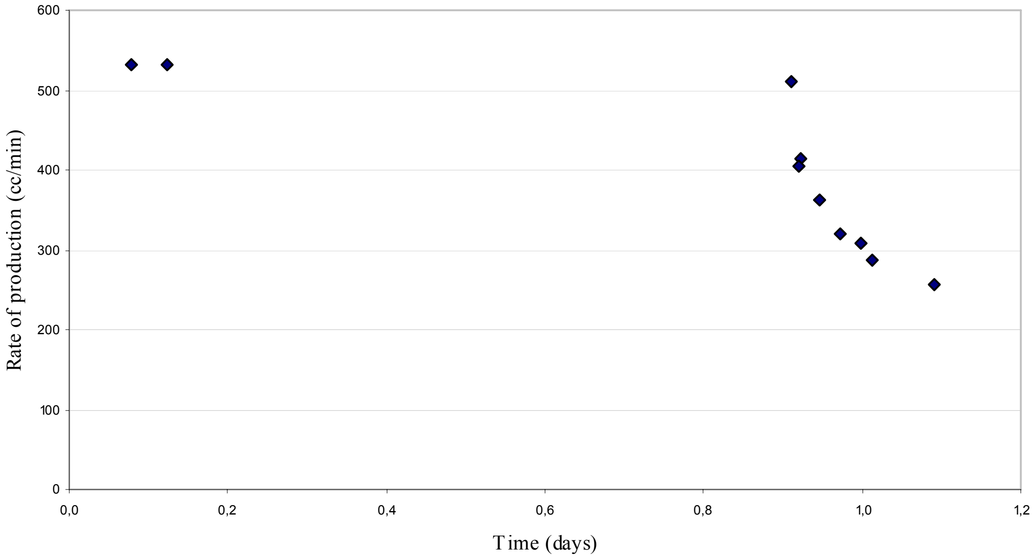

Figure 35. The hydrogen production rate in the function of time. The empty period between T = 0.2 and T = 0.9 represents a period when the electrolyser was in idle mode during the night between the two testing days.

There were no hydrogen leaks detected at any time during the test. The pressure in the system was increasing rapidly up to about 6 bars and after that the increasing rate of the pressure was strongly decreasing (figure 36). The maximum rate of pressure increase achieved during the test was 46.5 mbar / min. This was recorded straight after the process was started again after the night break. After 13:43 pm on a second testing day (T = 1.00 d) there was no observable pressure increase in the system. At the beginning of the production process there was more fluctuation in the climbing rate of the pressure, mainly due to continuous alarms and malfunctions in the electrolyser operation. The process of binding H_2 chemically to the hydride is an exothermic process, which means that during the filling the temperature of the container is rising and this in turn causes the increase of the pressure. The pressure inside the generator and the storage system had decreased during the night time idle period from 3.1 bars to 2.5 bars. The rate of pressure loss during the night was -0.66 mbar / min.

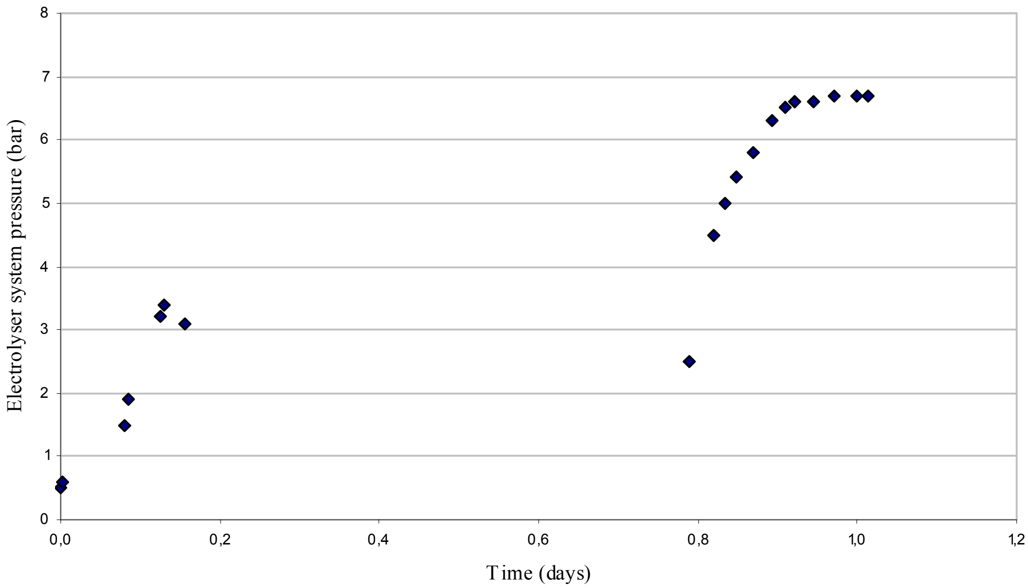

Figure 36. Pressure of the generator and the storage system represented in the function of time. Pressure readings were collected manually from the generator display. The empty period between T = 0.2 and T = 0.8 represents a period when the electrolyser was in idle mode during the night between the two testing days.

The decrease in the battery voltage seemed to be continuous until the system was changed to grid power. This is natural, because there is also a power demand on the idle mode as well. During the time the battery unit was used, the voltage had dropped from

26.1 V to 22.3 V. The voltage drop during the whole test was significant and estimates the upcoming exhaustion of the battery unit. A sign from the possible battery exhaust was also the high decrease rate of the voltage just before the change to the grid power. The decrease rate of -19 mV / min was recorded 20 minutes before the power source change.

The power consumption of the electrolysis could not be measured due to the lack of equipment. Also the inverter proved to be inappropriate to deliver the power to the electrolyser, because it damaged during the test.

9 DISCUSSION

These experiments on the H_2 vehicle and the hydrogen refuelling facility gave a lot of knowledge about the different components and their synchronization with each other. The initial tests proved some of the components useful and well functioning and some of the tests revealed results that were not foreseen in the planning stage of hydrogen car project. Despite of that, this work raised numerous more questions and topics to put under research, which is valuable concerning the nature of the H_2 car project as one of the long term demonstration projects in the University of Jyväskylä RE -programme. Also this work provides some valuable background knowledge on the progress of this whole project.

Driving tests and FC efficiency calculations lead to a conclusion that the driving range with batteries could be doubled with one full HC-MH1200 storage unit. The overall driving range with PEM fuel cell and one container is therefore about 15 km. This is enough for a shopping trip on the city streets without any sightseeing. According to this research, two containers would provide a driving range of about 22 km, which is the total maximum with the current equipment. If the result is compared to the driving range of Jeep Commander fuel cell hybrid vehicle mentioned in the chapter 4.3.6, the accomplished driving range of Fantasia test vehicle is only 2.3 % of this. Even though the driving range achieved with Fantasia was relatively short, it must be kept in mind that the concept under research here is the combination of a refuelling facility and a vehicle. In addition, both the fuel production and the vehicle are non-polluting, local and grid independent. The consumption of fossil material comes to the picture only when the manufacturing process of the equipment itself is considered. The relatively short driving range achieved by this project was partly due to the technical problems in the rear shaft welds of the vehicle. It is possible that the shaft had been partly damaged during the drive tests causing a significant power loss. Driving tests revealed that this vehicle is for the summer weather only when it is to be driven on public roads. There is lot of improvements to be done also for the convenience of the driving. Also the maintenance of the electric motors and the rear shaft was difficult, because there was no service door in the back of the vehicle. For further research, I strongly recommend to modify the vehicle for this part to improve the maintenance convenience.

Fuel cells, both AFC and PEMFC, did not operate as they should and this proved the vulnerability of this project concerning the practical utilization of the vehicle. The vehicle hybridization degree approached a value of 0.1 which means that fuel cell with higher capacity are needed for the future tests. The calculations revealed that the PEMFC used in this project is not sufficient to provide power to the motors even at the speed of 5 km/h. In case that HCORE-500-2 is to be used in the future, there is a need for DC/DC converter between the control unit of the car and the PEMFC, because the output voltage of the fuel cell is 48 VDC.

Metal hydride container tests resulted with the relationship curve of the pressure and the container surface temperature. This was similar to the relationship curve sent by the container manufacturer (attachment 1). The experiments foresaw the difficulties in supplying the H_2 at proper pressure to the FC during driving on winter temperatures. According to the tests, the container surface temperatures below 10 °C can cause difficulties in the PEMFC performance. The endothermic process, which releases the H_2 from the metal hydride, makes the situation even worse by cooling down the container material. The answer for this problem would be the waste heat generated by the PEMFC, which can be utilized during driving. However, the temperatures below 10 °C inside the vehicle is not pleasant concerning the driver, so the heating issue has to solved anyway if the vehicle is to be used in winter period.

Pressure regulator worked properly fulfilling the requirements completely. Though the outlet pressure of this regulator was not constant, it did not exceed the required limits ($2.1 \leq p \leq 4.1$ bars) for PEMFC, provided that the inlet pressure led to it was sufficient. The highest pressure that was measured inside the hose assembly was 3.4 bars, which is 0.7 bars lower than the maximum pressure limit set for the FC hydrogen inlet. It is also not likely that the temperature around the container would climb any higher than 63 °C in practice. This indeed is very close to the maximum operation temperature of the container (65 - 70 °C) announced by the manufacturer. In was calculated in the chapter 8.5 that even the maximum possible pressure (4.09 bars), that could be achieved before the emergency valve of the HC-MH1200 container would open, can not cause too high pressure in the hydrogen line. In conclusion, high temperatures are not a problem to the H_2 production and utilization, but the situation is different at low temperatures. Because the nature of the metal hydride container is that it can hold large amounts of hydrogen without an overpressure more than 1 or 2 bars, the difficulties in getting the hydrogen pressure to proper level at winter conditions are likely. That is not the feature of pressure regulator, but of the container itself.

The leak tests were performed for the safety and reliability reasons and they proved to be useful. The tests showed that safe and leak proof system can be built, but it requires carefully chosen connectors and fittings. Even a sealed hose assembly kept its pressure for maximum of three weeks. The leak rate equations can be used to calculate the H_2 consumption of the FCs and also to estimate losses in hydrogen supply during future experiments. The figure representing the leakage rate in the function of pressure is not absolutely reliable due to scarcity of the data measured. The data was collected manually and therefore more intense pressure readings were not possible. In future tests, this leak monitoring would require automated and computer aided data collection. The results of these tests are guiding, and reveal rapid growth of hydrogen leakage when the system pressure increases. During the leak test number three it was noticed that the pressurized hydrogen will escape from the hose completely at the end. The connector type does not seem to affect the leak rate critically. The quick lock connector used in this test seemed to leak a bit more rapidly than the connector with a pressure regulator (in 15 days to 1.02 bars; in 20.9 days to 1.1 bars).

Hydrogen production was also one of the topics under research. The fuel manufacturing worked properly and the container could be filled easily with hydrogen without any detectable leaks. The battery unit turned out to be too small to produce enough power for the electrolysis only with the storage capacity it had. The main function of the battery unit was therefore to act as buffer storage for the energy coming from the RE-sources. The inverter used in these experiments proved to be unreliable to supply the suitable voltage for the generator. The same can also be noticed from the manual instructions of the inverter and the hydrogen generator. The nominal power consumption of the generator is 350 W and the continuous output of the inverter is 300 W. The deviance of this research on the generator was that the power consumption for the volumetric unit of H_2 could not be measured. This was due to the lack of measuring equipment. Anyway, this production experiment resulted with useful notices about the process, such as the maximum pressure and the rate of the H_2 production in the function of pressure. The production test revealed some observations which require more research and which were not noticed before the test. The behaviour of the metal hydride during filling is one of the topics which needs more research. As mentioned in the chapter 4.1.3, in MH storage hydrogen is chemically bonded to a metallic lattice. This means that the process has a certain speed to happen. Therefore, the pressure drop during the idle period of the test was probably due to small and continuous leakage or slowness of hydrogen seepage in the metal hydride container. However, when the amount of pressure drop is compared to the leak test results, the decreasing of the system pressure during the idle period could be explained with the leakage only. Other deviance of the electrolyser testing was that it was stopped soon after the pressure reached its maximum and therefore it remained a secret how the production rate would have behaved as the metal hydride reached its maximum storing capacity. As mentioned in the chapter 8.7, the process seemed to approach a balanced situation where the system pressure and the H_2 production rate would have been constants. In practice, this would mean a situation where the production rate and hydrogen seepage in the metallic lattice are equal under the prevailing pressure.

The continuous alarms during the fuel manufacturing process caused some confusion to the test results as well. During the process there were water low- and overload -alarms recorded several times. Water low –alarm was probably due to the long storing time of the generator without using it, which had probably dried its internal water reservoir. This alarm did not occur after the first testing hours. The overload alarm is a safety feature which turns off the production when there is a risk of hydrogen leakage. This in turn could be due to the nature of the metal hydride container, which binds the hydrogen more rapidly than it could be produced, and the generator took this phenomenon as a leak. This causes nothing but the difficulty in assessing the amount of H_2 produced, because there was always a short brake after the overload alarm before the normal operation could continue.

Answers were found to the questions presented as research problems, but the accuracy and quality of the measured data could have been better. This inaccuracy of the data measured was mainly due to lack of equipment and technical assistance and also to the planning of the tests. The fuel cell experiments remained as well insufficient in practical sense due to technical problems, and they will require more research in the following experiments. To move forward with this experimental setup in the future, the purchasing of new components to replace the most unsuitable parts of the system would be the first thing to do. The fixing of car power transmission and the data collection systems would be the most important future improvements.

As a word of criticism, it was found out during this research that the technical problems with the components and the cost of appropriate measuring equipments proved to be too difficult to overcome for the University of Jyväskylä Renewable Energy Programme. Also the collaboration between different faculties proved to be too difficult due to responsibility and bureaucracy issues. The technical and monetary resources must be increased in order to efficiently realize this kind of project in the future. One of the lessons learned from this work would be that careful planning concerning the automated data collection devices and their computer software must be done before further research. However, the real life project of this kind is highly educative concerning the students and researchers in the University.

As mentioned before, the proposals for further research problems would not be hard to make. The driving capacity without a grid power within a week or a month would be interesting to find out. Yet the driving range of the car was estimated within this work, it remained a secret how often a trip like this could be made. To put it in another words, what is the maximum hydrogen production capacity by using only the installed PV and WTG. At least from my point of view, an interesting thing would be to know the efficiencies of all the components of this long chain of energy. Based on this, a well-to-wheel efficiency of this particular vehicle and the refuelling system could be determined. A more detailed research problem could be to determine the power consumption of the electrolyser when an empty container is reloaded.

ACKNOWLEDGEMENTS

Special thanks to David Agar, Mikko Lommi, Ulla Haara, Robert Gyllenberg, Pasi Paananen and supervisor Dr. Mohanlal Kolhe.

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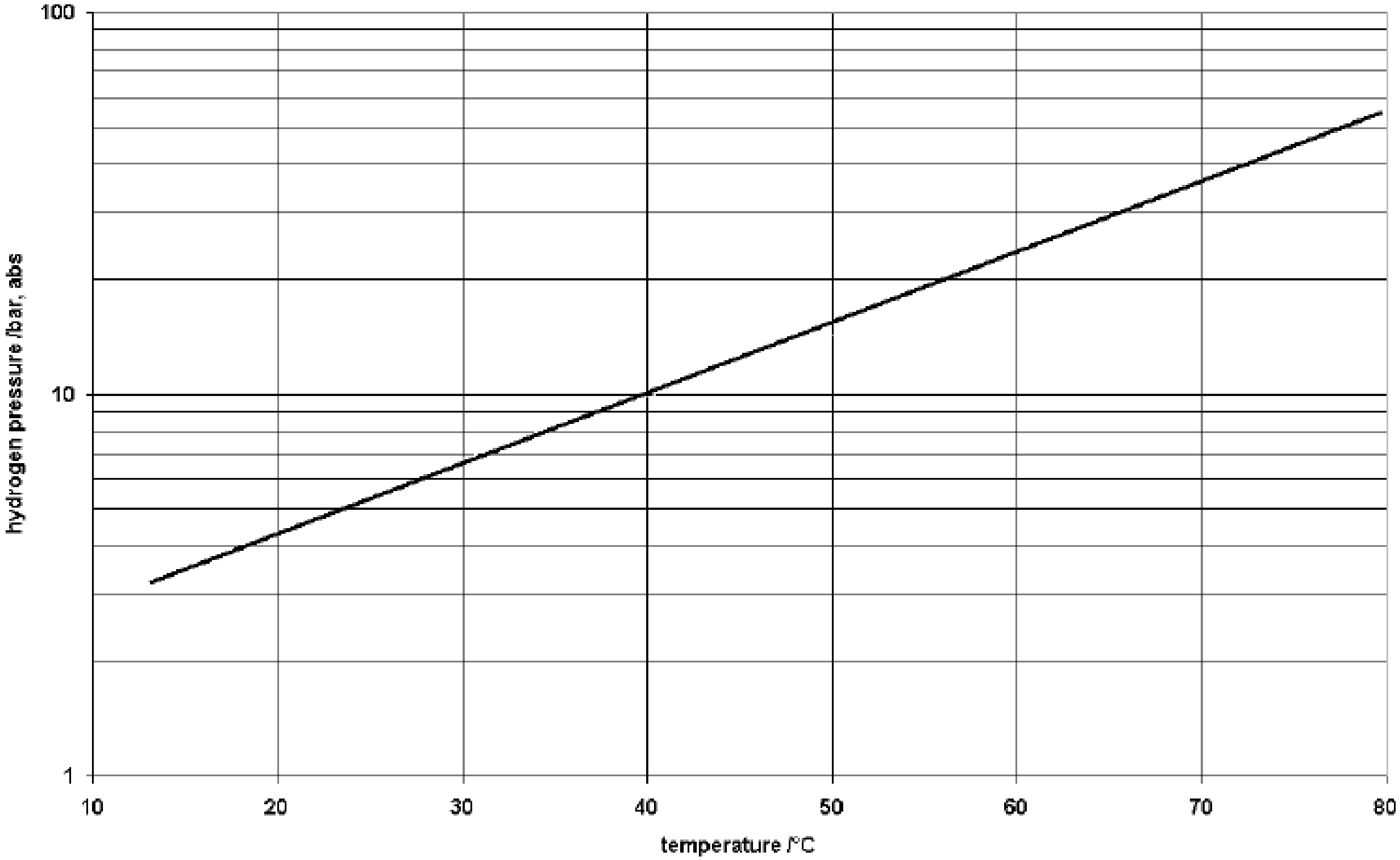
ATTACHMENTS

1) Temperature and pressure curve for HC-MH1200 sent by the manufacturer (Source: Oy Hydrocell Ltd. / Rantanen J. email 3rd April 2006)

2) Driver´s log form for Fantasia test vehicle


100
10
1
hydrogen pressure /bar, abs
10
20
30
40
50
60
70
80
temperature /°C

H2-car Fantasia **DRIVER´S LOG** *form updated:* 18 Nov 2005, AP

Date: ____________________

Time: ____________________

Drivers: ____

Weather conditions: ____

Air temperature: __________ °C

Air moisture content: ____________________

Car wheels: ____________________

Fuel cell type: ____________________
(energy source)

Route: ____

____

Distance (total): ____________________ km

Hydride pressure (starting point): ____________________ bar

Hydride pressure (finishing point): ____________________ bar

Battery level of charge (starting point): __________ (finishing point): __________

Voltmeter:

Top speed: __________ km/h

Current max: __________ A

Voltage max: __________ V

Power max: __________ kW

Other notes: ____

____

____

____

____
Signature