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TECHNO-ECONOMIC FEASIBILITY OF HYDROGEN PRODUCTION VIA POLYMER MEMBRANE ELECTROLYTE ELECTROLYSIS FOR FUTURE POWER-TO-X SYSTEMS

Faculty of Natural Sciences
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ABSTRACT

Mikko Lappalainen: Techno-economic feasibility of hydrogen production via polymer membrane electrolyte electrolysis for future Power-to-X systems
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PEM water electrolysis and Power-to-X systems are seen as potential technologies to achieve GHG emission reductions and limit the global warming to 1.5 °C above pre-industrial levels. Moreover, Power-to-X systems are also seen as a solution to balance electricity grid in the case of higher share renewable energy production which would increase the intermittent energy production. One technology cannot solve all problems but hydrogen has already versatile end-use applications and potential to make significant reductions in CO_2-emissions in energy sector, limiting the global temperature rise.

Thesis first introduces the hydrogen as an energy carrier. Hydrogen properties, main production methods, delivery and end-use applications are examined. PEM water electrolysis fundamentals, technology and operation strategies with the most significant factors impacting the viability are presented. PEM hydrogen production costs are also compared with SMR and alkaline electrolysis production methods. Power-to-X pathways that can be considered potential for PEM electrolysis are introduced and also recent notable projects are presented. In practical part, thesis is divided into a technical evaluation of the pilot scale PEM electrolysis operation and economic calculations of feasible operation frameworks for PEM electrolysis in Finland. First, working principal of the electrolyser, auxiliary equipment and monitoring systems are introduced. For the economic calculations, the calculation parameters to find suitable economic operation framework are presented. Three different electricity price scenarios based on realized hourly prices in Finland 2018 and in Denmark 2018 were used. One of the scenarios used optimistic lower average electricity price with higher volatility modified from the Finland 2018 scenario. Other necessary parameters were gathered from the literature.

Results from the operation of the pilot scale PEM electrolyser showed excellent dynamic properties and stable, independent hydrogen production. Challenges came from low ambient temperatures which impacted the cold start-up time. The economic calculations showed only Speculative 2030 with FCR-N scenario with optimistic assumptions to be feasible without raised hydrogen and oxygen prices. With higher products values, all scenarios became viable. The distribution of yearly costs and incomes for different scenarios showed that main incomes are due to hydrogen sells. Additional revenues from oxygen and heat utilization have significantly smaller but still relevant share of incomes. Also FCR-N operation proved to be beneficial in all scenarios but significance decreased as electrolyser utilization rate increased. Additionally, FCR-n market revenues are limited and auction based therefore future auctions are not guaranteed to offer same revenue for balancing services. Feasible electrolysis operation required high utilization rate, in which case costs were dominated by electricity costs other costs played smaller role.

Keywords: PEM electrolyser, hydrogen, Power-to-X

TIIVISTELMÄ

Mikko Lappalainen: PEM (Polymer Electrolyte Membrane) elektrolyysin teknis-taloudellinen soveltuvuus vedyn tuotantoon tulevaisuuden Power-to-X systeemeitä varten
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PEM elektrolyysi ja Power-to-X systeemit nähdään potentiaalisina teknologioina kasvihuonekaasupäästöjen vähentämiseen ja ilmaston lämpenemisen rajoittamiseen 1.5 °C:seen verrattuna esiteolliseen aikaan. Power-to-X systeemit nähdään myös mahdollisena keinona tasoittaa mahdollisen uusiutuvan energian lisääntymisestä johtuvaa jaksoittaista energiantuotantoa, joka toisi haasteita sähköverkoille. Yksi teknologia ei ratkaise kaikkia ongelmia, mutta vedyllä on jo monipuoliset käyttökohteet ja potentiaalia tehdä merkittäviä vähennyksiä CO_2 päästöihin energiasektorilla ja näin rajoittaa ilmaston lämpenemistä.

Ensin diplomityössä esitellään vety energiankantajana. Teoriaosuudessa esitellään vedyn ominaisuudet, pääasialliset tuotantomenetelmät, jakelu ja käyttökohteet. Kappaleissa taustoitetaan PEM elektrolyysin perusteet, teknologia ja operointi strategiat, sisältäen kannattavuuteen eniten vaikuttavat tekijät. PEM elektrolyysillä tuotetun vedyn kustannuksia myös vertaillaan SMR-teknologian ja alkalielektrolyysin tuotantokustannusten kanssa. Potentiaaliset Power-to-X reitit esitellään yhdessä viimeaikaisten huomattavien projektien kanssa. Käytännöllinen osuus on jaettu tekniseen, pilotti kokoluokan PEM elektrolyysin operoinnin tarkasteluun ja taloudelliseen laskentaan kannattavien toimintaympäristöjen löytämiseksi Suomessa. Tekninen osuus esittelee elektrolyysin toimintaperiaatteen sekä avustavat- ja monitorointilaitteistot. Taloudellisessa laskennassa tuottavan toimintaympäristön löytämiseen vaikuttavat tekijät käydään läpi. Laskennassa käytettiin kolmea eri skenaariota, joissa sähkönhintoina käytettiin Suomen ja Tanskan vuonna 2018 realisoituneita tuntikohtaisia hintoja. Yhdessä skenaariossa käytettiin optimistista alempaa sähkön keskihintaa ja suurempaa volatiliteettia. Muut tarvittavat parametrit saatiin kirjallisuudesta.

Pilottikokoluokan PEM elektrolyysin operoinnin tulokset osoittivat erinomaisia dynaamisia ominaisuuksia ja vakaata, itsenäistä vedyntuotantoa. Haasteita ilmeni alhaisissa käyttöympäristön lämpötiloissa, jotka vaikuttivat kylmäkäynnistys aikaan. Taloudellinen laskenta osoitti, että ainoastaan Speculative 2030 FCR-N skenaario optimistisilla lähtöarvoilla osoittautui kannattavaksi ilman korotettuja vedyn ja hapen myyntihintoja. Korotetuilla vedyn ja hapen arvoilla kaikki skenaariot olivat kannattavia. Vuosittaisten menojen ja tulojen jakauma osoitti, että valtaosa tuloista saadaan vedyn myynnistä. Lisätulot hapesta ja lämmöstä olivat selvästi pienemmät, mutta silti merkittävät. FCR-N operointi osoittautui hyödylliseksi kaikissa tapauksissa, mutta hyöty väheni käyttötuntien kasvaessa. Lisäksi FCR-N markkinat ovat rajalliset ja tarjouskilpailuun perustuvat, joten tulevaisuuden tarjouskilpialut eivät välttämättä tuota samoja tuloja tarjotusta FCR-N kapasiteetista. Kannattava elektrolyysin operointi vaati korkeita vuosittaisia käyttötunteja, jolloin suurin osa menoista syntyi sähköstä ja muut menot olivat pienemmässä roolissa.

Avainsanat: PEM elektrolyysi, vety, Power-to-X

PREFACE

This Master’s thesis is part of the Bioeconomy+ project, which targets to demonstrate the utilization of biobased CO_2 as a raw material for synthetic transportation fuels, biogas upgrading or industrial products. The project is co-financed by the European Regional Development Fund, VTT and a number of partners.

I would like to thank my examiners Janne Kärki and Cyril Bajamundi from VTT and Henrik Tolvanen from Tampere University for help and excellent guidance in this thesis. Also huge thanks goes to all personnel in VTT who gave advices and supported me during the writing process. Finally I wish to thank my family and friends for always being supportive and encouraging.

Jyväskylä, 21.3.2019

Mikko Lappalainen

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LIST OF SYMBOLS AND ABBREVIATIONS

AC	alternating current
BOP	balance of plant
CAPEX	capital expenditure
CCS	carbon capture and storage
CCUS	carbon capture, utilization and storage
DC	direct current
DRI	direct reduced iron
ESD	emergency shutdown
FCEV	fuel cell electric vehicle
FCR-D	Frequency Containment Reserve for Disturbances
FCR-N	Frequency Containment Reserve for Normal operation
GHG	greenhouse gas
HER	hydrogen evolution reaction
HHV	higher heating value
HIA	Hydrogen Implementing Agreement
HRS	hydrogen refueling station
IEA	International Energy Agency
ICPP	Intergovernmental Panel on Climate Change
IRENA	International Renewable Energy Agency
KPI	key performance indicator
LFL	lower flammability limit
LHV	lower heating value
MEA	membrane electrode assembly
OER	oxygen evolution reaction
OPEX	operational investments
PEC	photoelectrochemical
PEM	polymer electrolyte membrane
PTG	power to gas
SNG	synthetic natural gas
SOE	solid oxide electrolysis
SOEC	solid oxide electrolysis cell
STHC	solar high temperature thermochemical
STP	standard temperature and pressure
TOC	total organic carbon
TSO	transmission system operator
UFL	upper flammability limit
VSPA	vacuum pressure swing adsorption
WACC	weighted average cost of capital
WGS	water-gas shift
WTP	water treatment plant

ΔG	Gibb's free energy change	J mol^{-1}
ΔH	enthalpy	kJ mol^{-1}
ΔS	entropy	J mol^{-1} K^{-1}
η_E	electrolysis efficiency	-
η_F	Faraday efficiency	-
C_E	specific energy consumption	kWh/Nm^3 / kWh/kg

F	Faraday’s constant	96,485 C/mol
f_{H_2}	hydrogen production rate	Nm^3/h
I_{cell}	cell current	A
T	temperature	K
U	cell voltage	V
U_{act}	activation overvoltage	V
U_{con}	concentration overvoltage	V
U_{ohm}	overvoltage caused by ohmic losses	V
U_{rev}	theoretical cell voltage	V
U_{tn}	thermoneutral cell voltage	V
z	electrons transferred per reaction	-

1. INTRODUCTION

According to the Intergovernmental Panel on Climate Change (IPCC) special report on the impacts of global warming 1.5 °C published in October 2018 [1], there is a high confidence, that it would require ”rapid and far-reaching transitions” across the global economy to limit warming to 1.5 °C above pre-industrial levels. The Paris Agreement (2015) aims to keep global temperature rise below 2 °C and even limit the rise to 1.5 °C [2]. Currently the agreement has been ratified by 185 parties of 197 parties in the convention [3]. The difference between warming of 1.5 °C and 2 °C impacts several hundred million people causing suffering from water-stress, hunger, poverty, tropical diseases, higher sea level rise and heatwaves. Actions required to stay in 1.5 °C would mean 40 - 50% reduction of CO_2 emissions from 2010 levels by 2030 and reach net zero around 2050.

Energy demand is growing globally and simultaneously CO_2-related emissions have increased. 67.4 % of global greenhouse gas emissions are produced due to energy systems. Renewable power generation technologies such as wind and solar photovoltaic have grown exponentially since the 2000s to reduce CO_2 emissions from energy production. Nature of these technologies is intermittent and difficult to predict. Also the most favorable locations for generating renewable energy are often far away from the consumption centers. These circumstances create a need to store the produced electricity [4].

Power-to-X is a concept where electric energy, and preferably intermittent renewable energy, is utilized to produce various types of energy carriers, such as hydrogen, to store electricity and balance grid during the peak times. Hydrogen’s ability to be converted between chemical and electrical energy while having a high heating value and potential for a long term energy storage is valuable in energy-systems. Hydrogen has also flexibility in producing methods (fossil fuels, renewable sources, thermal), function of scale (from W to GWs) and end use possibilities (industry, transportation, heating) [5]. Water electrolysis can be used to produce hydrogen and oxygen without CO_2 emissions if renewable energy sources for electricity are utilized. Polymer Electrolyte Membrane (PEM) is particularly suitable technology for operating with alternating energy production because of its dynamic range and ability to quickly ramp up and down [6].

Power-to-X concept with PEM electrolysis plant has been demonstrated in projects such as H2Future and Energiepark Mainz. H2Future project uses 6 MW Polymer Electrolyte Membrane (PEM) electrolysis plant at a steelworks in Linz, Austria to validate electrolysis route in steel manufacturing processes and additionally in grid balancing services. The operation of the PEM electrolysis plant is analyzed by administrative and general, safety and environmental, technical and economic performance [7]. A technical and economic evaluation was done to Energiepark Mainz 6 MW PEM electrolysis plant in Germany. The technical analysis included a calculation of the plant efficiency and economic evaluation consisted of analysis of different electricity procurement scenarios [8]. This thesis concentrates on hydrogen production via PEM water electrolysis economic and technical evaluation for Power-to-X systems. Main focus is in Europe and in techno-economic calculations, particularly in Finland. Research questions of the Master's thesis are:

- What Power-to-X pathways are considered potential for PEM electrolysis and what noteworthy projects have been demonstrated recently?
- What are the process limitations for 30 kW small scale PEM electrolyser system operation in Finland?
- Which factors have the most significant impact on economic feasibility of PEM water electrolysis in Finnish conditions?

Theoretical part of the thesis first introduces the hydrogen as an energy carrier. Hydrogen properties, different production methods, delivery and end-use applications are examined. Production methods include short introduction to the current main hydrogen production method, steam methane reforming, the most mature water electrolysis technology, alkaline electrolysis and research level solid oxide electrolysis. PEM water electrolysis fundamentals, technology and operation strategies with the most significant factors impacting the viability are introduced in the next subchapters. Lastly, PEM hydrogen production costs are compared with SMR and alkaline electrolysis production methods.

Power-to-X chapter introduces pathways that can be considered potential for PEM electrolysis. Also, noteworthy and recently demonstrated Power-to-X projects with PEM electrolyser system are presented. Pathways are divided into four categories. Hydrogen decarbonizing gas grid introduces direct hydrogen injection into gas grid and injection after methanation. Hydrogen decarbonizing transport presents hydrogen use in wide range of mobility applications. Hydrogen decarbonizing industry talks about hydrogen end-use in refineries, steel manufacturing and chemical industry. These sectors have also largest capacity PEM electrolysis system demonstration projects which are shortly introduced.

Practical part of the thesis presents the operation of the pilot scale PEM electrolyser. The working principal of the electrolyser, auxiliary equipment and monitoring systems are introduced broadly. The methods to study process limits and system behavior for PEM electrolyser in experimental operation are explained. Lastly, calculation parameters and methods to find suitable technical and economic operation framework for PEM water electrolysis in Finland are presented.

Finally results and discussion chapter presents the acquired results and discusses and completes main aspects from the thesis. First, steady-state and dynamic operation data are presented with practical experimental observations from the process. These experiments and data are used to find process limitations for PEM electrolyser operation in Finland. Calculations to find a feasible operational framework for the PEM electrolysis are mainly made on the basis of literature presented in theory part.

2. HYDROGEN AS AN ENERGY CARRIER

Hydrogen is not an energy source but a flexible energy carrier with a high heating value. Even though hydrogen is abundant in nature, to generate pure hydrogen, energy must be used. It has versatile end-use applications and potential to make significant reductions in CO_2-emissions in energy sector, limiting the global temperature rise [9].

The Figure 1 shows how the hydrogen could be implemented in the existing energy system. Hydrogen could balance the electric network during peak times, storing surplus electric energy into hydrogen gas which can be utilized in diverse ways.

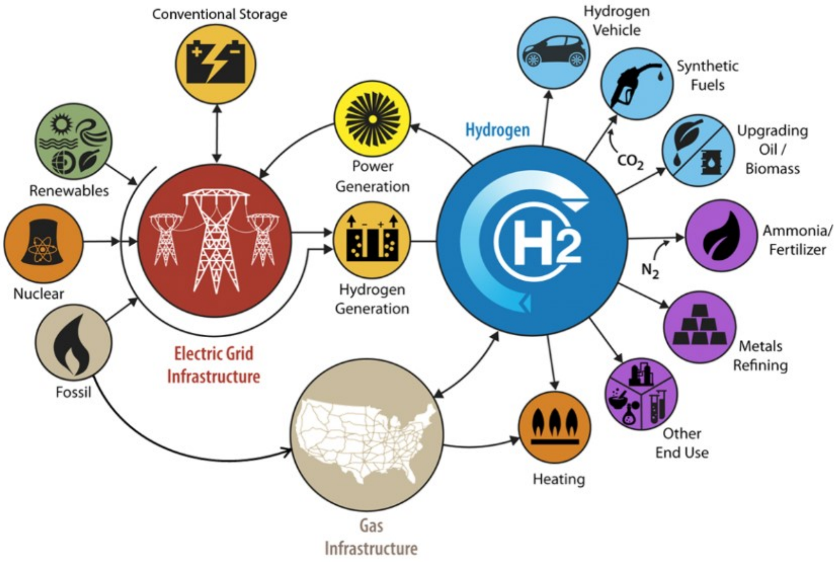

Figure 1. *Hydrogen as a part of energy system [10].*

Hydrogen is a colorless, odorless and tasteless non-toxic gas in standard temperature and pressure (STP, 273.15 K and 1 atm). Selected physical properties of hydrogen are presented in Table 1. Hydrogen has a relatively high energy content: 142 MJ/kg higher heating value (HHV) and 120 MJ/kg lower heating value (LHV). The difference between high and low heating values is molar enthalpy of vaporization of water (44.01 kJ/mol) [11].

Hydrogen is very light gas with density of 0.0899 kg/m^3 under STP conditions. This makes hydrogen storage more complicated and hydrogen is traditionally pressurized or liquefied in order to store reasonable amounts of energy. Figure 2 illustrates hydrogen density dependency on temperature and pressure. Pressure and temperature areas for pressurized, liquefied and cryo compressed storage. For example, if hydrogen is compressed to 700 bar at 293 K temperature, the density increases to 40 kg/m^3. Additionally, many other storage methods are also being investigated. These methods can be categorized in materials-based technologies which include metal hydrides, liquid organic hydrogen carriers (LOHCs) and sorbents (MOFs, Zeolites, Nanotubes) [12].

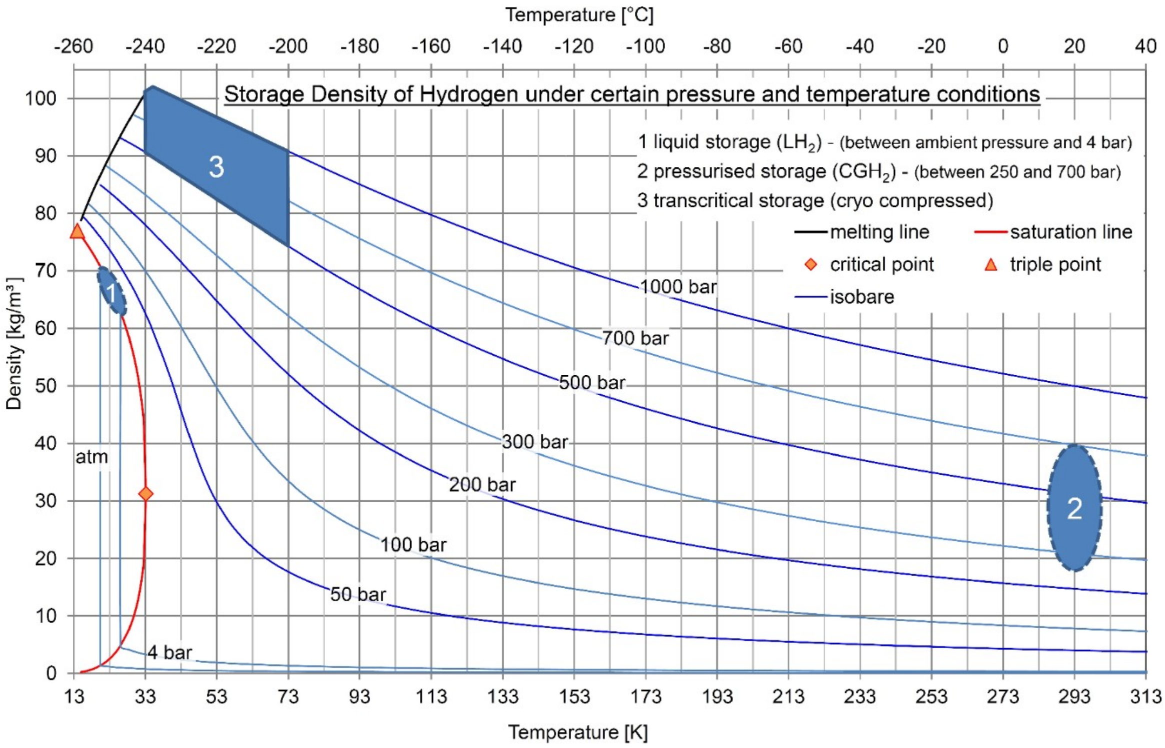

__Figure 2.__ Density of hydrogen under different temperature and pressure conditions [13].

Hydrogen has high diffusivity and can pass through porous materials and metals. Pressurized tanks with special coatings are well suited option for storage in small- and mid-scale applications, due to safety, efficiency and affordable price, as long as cycling rate is high [4].

Table 1. *Hydrogen's selected physical properties [11].*

Heating values	HHV: 142 MJ/kg (39.4 kWh/kg) LHV: 120.0 MJ/kg (33.3 kWh/kg)
Density	0.0899 kg/m^3
Boiling point	20.27 K
Melting point	13.99 K
Lower and upper flammability limits	LFL: 4 % UFL: 75 %
Autoignition temperature	773 - 850 K [14]

Hydrogen has a wide flammability range 4 - 74 % in air but as long as it remains in well ventilated area, there is no risk to reach this limit. However, limits increase with the temperature. Also, hydrogen has a relatively high autoignition temperature 773 - 850 K for stoichiometric hydrogen in air [14]. Autoignition temperature varies in literature as the temperature is dependent on the system factors. Hydrogen's low density makes it safer because it does not collect near the ground, but dissipates quickly in air in case of leakage [11]. In case of pressure and temperature changes, a special property of hydrogen, negative Joule-Thomson coefficient, must be taken into account. Unlike air in normal conditions, hydrogen heats up when it expands. Only below temperature 202 K does hydrogen show a typical Joule-Thomson effect [12].

2.1 Hydrogen production and utilization

About 113.2 millon metric tons of hydrogen was produced in 2017 in the world [15]. Approximate market value was 115.25 billion USD in 2017. This market is expected to grow to 154.74 billion USD in 2022 [16]. Hydrogen can be produced from various sources such as, water, natural gas, oil, biomass and (after gasification) from coal [17]. Figure 3 shows that globally, 95 % of hydrogen is produced from fossil fuels. Nowadays, hydrogen is mainly produced from natural gas via steam methane reforming, although partial oxidation of oil and gasification of coal are also used. Electrolysis (mainly alkaline electrolysis) method has a minor role in hydrogen production.

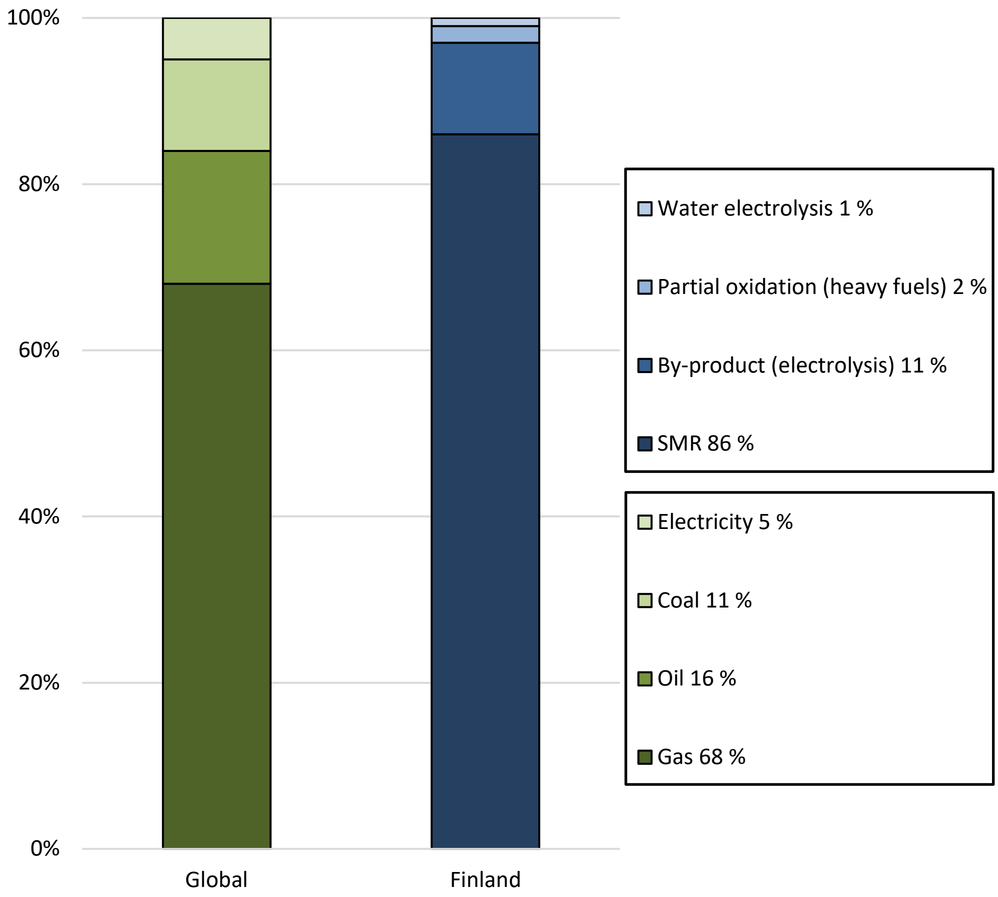

Figure 3. *Left column: estimated global hydrogen production by feedstock [12]*
Right column: estimated hydrogen production by technology in Finland [18].

The estimated total hydrogen prodcution in Finland is 200,000 t/a. Corresponding shares by technology are 86 % steam methane reforming, by-product (electrolysis) 11 %, partial oxidation of heavy fuel 2 % and water electrolysis 1 %. The largest share of the hydrogen is produced at Neste Oil Kilpilahti refinery (around 120,000 t/a). The biggest water electrolysis plant (alkaline) with capacity of 1300 t/a is at Woikoski in Kokkola Industrial park [18, 19].

Estimated use of hydrogen globally and in Finland is presented in Figure 4. Today, the primary uses of hydrogen are in the chemical industry. Ammonia and fertilizer production 51 %, and in oil refining industry 31 %. Noteworthy 11 % of the produced hydrogen is processed into methanol. Other chemicals, processing and steel, glass, welding sectors account for a minor part of the hydrogen consumption [20].

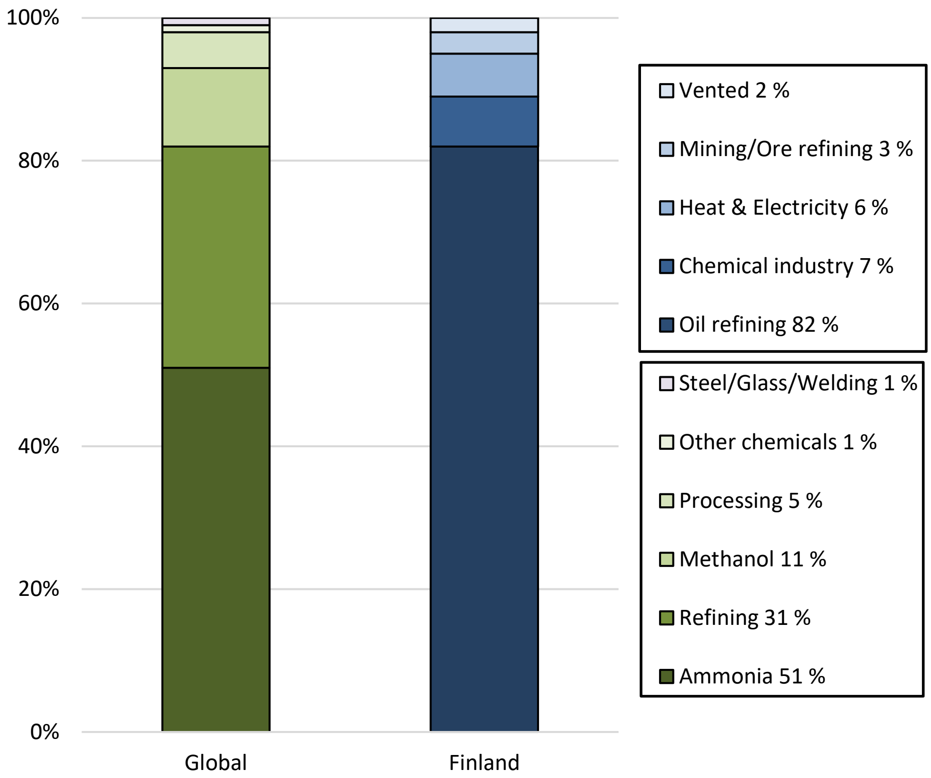

Figure 4. *Hydrogen demand by sector. Left column: globally [20] Right column: in Finland [18].*

Hydrogen consumption in Finland is dominated by oil refining 82 %. Chemical industry, mainly consisting of hydrogen peroxide (H_2O_2) and hydrochloric acid (HCl) production, is second largest sector with 7 % along with heat and electricity sector 6 %. The vented hydrogen 2 % comes from the chlorine/chlorate electrolysis plants.

Hydrogen consumption is expected to rise. Annual demand could increase from about 8 EJ to almost 80 EJ by 2050. This increase would be due to rise in hydrogen use in industrial, residential, transportation and power sectors. The corresponding reduction potential of CO_2 is 6 Gt annually. For this scenario to happen, policy and financial support with notable cost reductions are mandatory. [20, 21]

2.1.1 Alkaline water electrolysis

Alkaline water electrolysis systems are a mature technology scaling up to the MW-range and represent the most of the electrolytic technology at commercial level [22, 23]. A great advantage of this technology is that the alkaline water electrolysers can be manufactured from cheap and abundant materials [24]. In Figure 5 is shown the working principle of an alkaline water electrolysis cell.

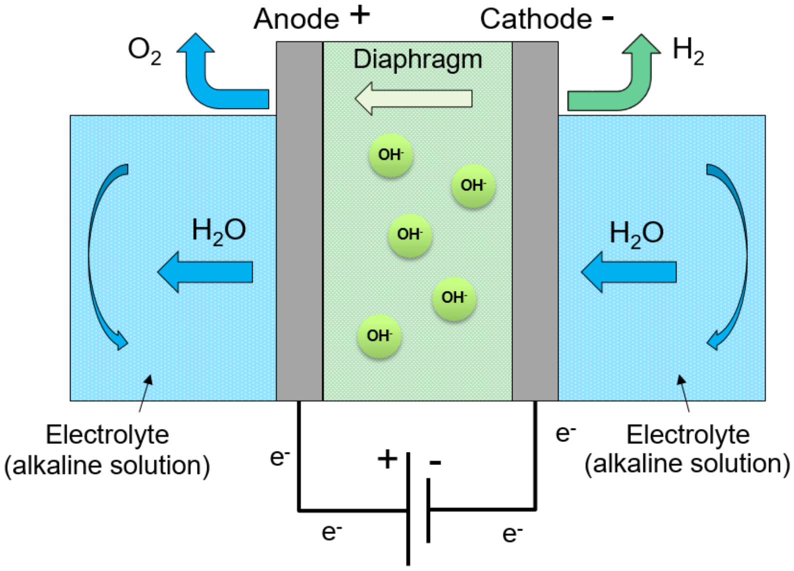

Figure 5. *Schematic of the working principle of an alkaline water electrolysis cell (adapted from [23]).*

The cell is structured from two electrodes separated by diaphragm and immersed in an alkaline aqueous solution made of concentrated potassium hydroxide (KOH). Gas-tight diaphragm transports hydroxide anions (OH^-) between the electrodes but separates product gases oxygen (O_2) and hydrogen (H_2) [24].

When electrical energy from the direct current (DC) source is applied to create sufficient difference of potential in two electrodes, the water is reduced at the cathode and simultaneously oxidation of water occurs at the anode according to reactions R1 and R2, respectively [24].

$$Cathode: \qquad 2H_2O_{(l)} + 2e^- \rightarrow H_{2(g)} + 2OH^-_{(aq)} \qquad \text{(R1)}$$

$$Anode: \qquad 2OH^-_{(aq)} \rightarrow \frac{1}{2}O_{2(g)} + 2e^- \qquad \text{(R2)}$$

The hydrogen gas is generated at the cathode and due to electric field, the yielded OH^- transports across the diaphragm to the anode. On the surface of anode, the OH^- anions recombine to produce O_2. The purity levels of product H_2 and O_2 can be 99.9 vol-% and 99.7 vol-%, respectively. [11, 23]

2.1.2 Solid oxide electrolysis and reversible solid oxide cell

Solid Oxide Electrolysers (SOE) enable water/steam electrolysis at high temperatures (600 °C - 900°C) [23]. This results in higher efficiency compared to other water electrolysis technologies. Currently, technology is at R&D stage. The working principal of solid electrolysis cell (SOEC) is shown in Figure 6.

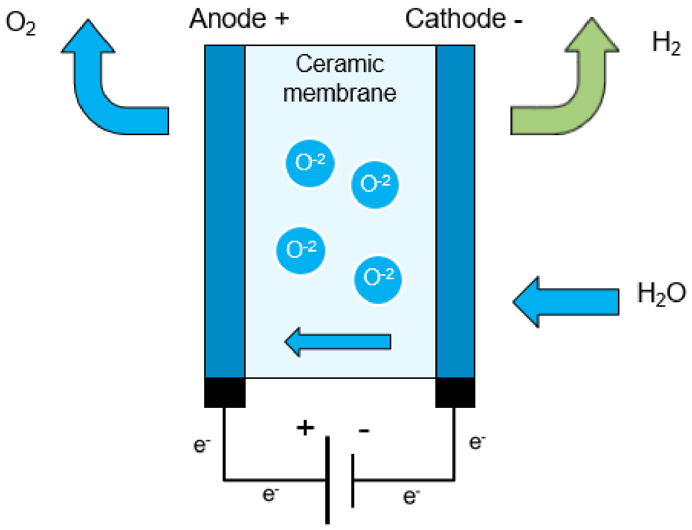

Figure 6. *Schematic of the working principle of a solid oxide electrolysis cell (adapted from [23]).*

Steam is supplied to the cathode, where it is reduced to produce H_2 according to reaction R3. The oxide anions (O^{2-}) produced in the cathode transport through the ceramic membrane (solid electrolyte) to the anode, where they recombine forming O_2 according to reaction R4 [23].

$$H_2O_{(g)} + 2e^- \rightarrow H_{2(g)} + O^{2-} \tag{R3}$$

$$O^{2-} \rightarrow \frac{1}{2}O_{2(g)} + 2e^- \tag{R4}$$

In contrast to alkaline electrolysis, reactions evolve with the electrodes in gas phase. The main problem for the industrial application is the limited long-term stability of SOE cells [23].

According to Santhanam et al. article of Theoretical and experimental study of Reversible Solid Oxide Cell (r-SOC) systems for energy storage (2017) [25] Solid Oxide Cell reactors can theoretically be operated bot as an electrolysis and a fuel cell. This would enable an effective energy storage systems. The article mentions roundtrip efficiencies of 55 % for commercially available systems at 1 bar pressure. At increased 30 bar pressure the roundtrip efficiency increases to 60 %. The theoretical limits for efficiencies are 98 % and 99 % respectively for same systems with studied conditions.

2.1.3 Steam methane reforming

Today, steam methane reforming (SMR) is the main hydrogen production method but its disadvantage is caused of CO_2 emissions [26]. From reaction R6 can be seen that one mol of CO_2 is generated for every mol of produced H_2. Figure 7 displays a simplified block diagram of a SMR system. First methane (CH_4) and steam (H_2O) react over a high-temperature catalyst. Typically a nickel-based catalyst operates at 900 - 1,200 K temperatures and 5 - 25 bar pressures. The endothermic reaction requires external source to provide heat to the system. The heat is usually provided via combusting additional methane and utilizing the available energy in the separated exhaust stream [27].

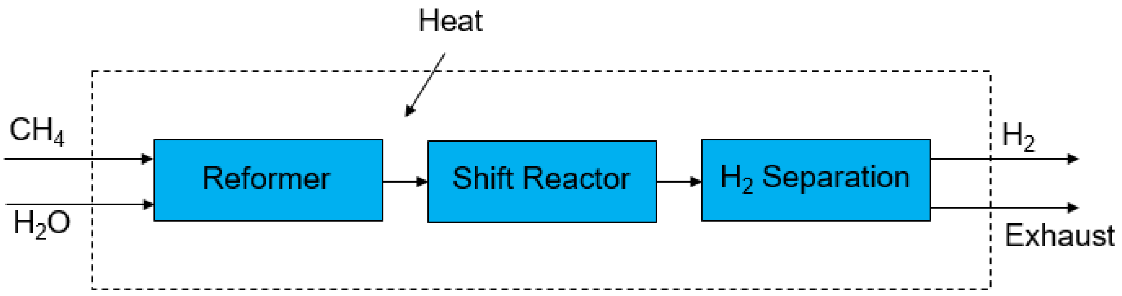

Figure 7. *Simplified diagram of SMR system (adapted from [27]).*

Reaction R5 shows the ideal global reaction for methane reformation. Reaction assumes reactors kinetics to be fast enough to achieve chemical equilibrium in product gas (syngas). Syngas composition depends on steam-to-carbon ratio, so it may contain also significant amounts CH_4, H_2O and CO_2.

$$CH_4 + H_2O_{(g)} \rightarrow CO + 3H_2 \qquad \Delta H_R = +251\ MJ/kmol_{CH_4} \qquad \text{(R5)}$$

After reformation, the syngas is passed through a water-gas shift (WGS) reactor where CO converts to CO_2 and H_2 using the additional H_2O or the available H_2O in the syngas (Reaction R6).

$$CO + H_2O_{(g)} \rightarrow CO_2 + H_2 \qquad \Delta H_R = -41.2\ MJ/kmol_{CO} \qquad \text{(R6)}$$

Although the WGS reaction is exothermic, the net reaction of (R5) and (R6) is endothermic. Last step in SMR is the separation of H_2 from product gas exiting the WGS reactor. Purity for generated H_2 is >99 % [27].

2.2 Hydrogen delivery pathways

As in hydrogen production methods, the hydrogen delivery pathways are distinguished in distributed and centralized categories (Figure 8). In distributed, also called on-site, hydrogen generation the production and delivery take place in the immediate proximity of the end-use site. On-site hydrogen production by water electrolysis requires demineralized water or tap water source with water treatment plant. 1 kg of produced hydrogen requires approximately 9 liters of water. Electricity needed in the process can be obtained from the grid or from separate production. Electricity consumption depends on electrolyser system efficiency but is in range of 50 - 60 kWh/kg of H_2 [12]. Possible further compression may increase electricity demand or a plant air source for pneumatic compression.

Centralized pathway includes large-scale hydrogen production at centralized location and a delivery to the end-use site. The transport from centralized hydrogen generation can be divided further into lorry delivery of compressed gaseous or liquid hydrogen and pipeline transportation of compressed gaseous hydrogen [28]. Currently, and likely in the medium term, these are the leading transportation methods used commercially [12].

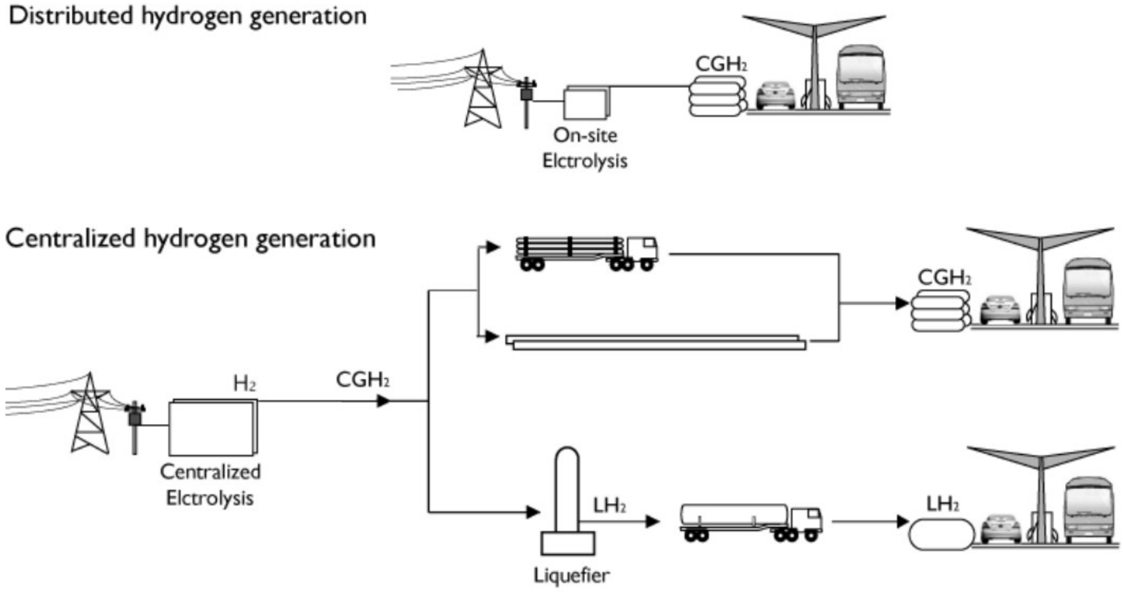

Figure 8. *Typical hydrogen delivery pathways separated in distributed and centralized hydrogen generation (adapted from [29]).*

Every transport option requires specific equipment, involve different operating costs, energy input and transporting volumes. Required transport volume, distance and acceptable costs determine which option is the most suitable in each case. Small to medium amounts of hydrogen are usually transported by lorry in pressurized gaseous form in tubes made of steel or lighter composite. For larger hydrogen volumes and longer distances liquid trucking is preferred. Lower trucking cost compensates for additional liquefaction cost. For very large amounts of hydrogen transportation, a pipeline is most cost-effective. However, initial building costs are major barrier to pipeline delivery infrastructure [12]. Potential solution is the development and use of cheaper pipeline materials, for example fiber reinforced polymer, or adapting natural gas grid network to hydrogen transportation [15]. Injecting hydrogen into natural gas grid is discussed more in chapter Power-to-X.

3. POLYMER ELECTROLYTE MEMBRANE WATER ELECTROLYSIS

In the 1960's General Electric developed the first water electrolyser based on a solid polymer electrolyte, which is referred as Polymer Electrolyte Membrane or Proton Exchange Membrane (PEM) water electrolysis. Some sources use name solid polymer electrolyte (SPE) water electrolysis as well [22].

This chapter first introduces water electrolysis thermodynamics, electrochemistry and system structure. Next, system and stack efficiencies, voltage degradation and system lifetime are discussed. Lastly, hydrogen production cost chapter discuss about parameters affecting hydrogen cost produced via PEM water electrolysis and also includes a production cost comparison between different technologies.

3.1 Thermodynamics

In reaction the global reaction for water electrolysis (R7), water dissociates into molecular hydrogen and oxygen. In standard conditions (T° = 298 K, p° = 1 bar), water is liquid and products (H_2 and O_2) are gaseous.

$$H_2O_{(l)} \rightarrow H_{2(g)} + \frac{1}{2}O_{2(g)} \tag{R7}$$

Enthalpy, entropy and Gibbs free energy change for the reaction are:

$$\Delta H° \left(H_2O_{(l)}\right) = +285.84\ kJ\ mol^{-1}$$

$$\Delta S° \left(H_2O_{(l)}\right) = +163.15\ J\ mol^{-1}K^{-1}$$

$$\Delta G° \left(H_2O_{(l)}\right) = \Delta H° \left(H_2O_{(l)}\right) - T\Delta S° \left(H_2O_{(l)}\right) = +237.22\ kJ\ mol^{-1}$$

Formation of 1.5 mole gaseous species has a favorable entropic contribution but strongly endothermic enthalpy change results in positive Gibbs free energy change. Consequently, the reaction is non-spontaneous [11].

For reaction R7, at pressure 1 bar, state functions are plotted in Figure 9. The total energy ΔH (T), thermal energy ΔQ (T) (= TΔS(T)) and electrical energy demand ΔG (T), are state functions which change with temperature T. The total energy needed to split one mole of water increases marginally with the temperature, varying between 283.5 and 291.6 kJ/mol$_{H2}$ in the temperature range of 0 - 1,000 °C [30]. However, ΔH stays almost constant at the electrolyser's operating temperature.

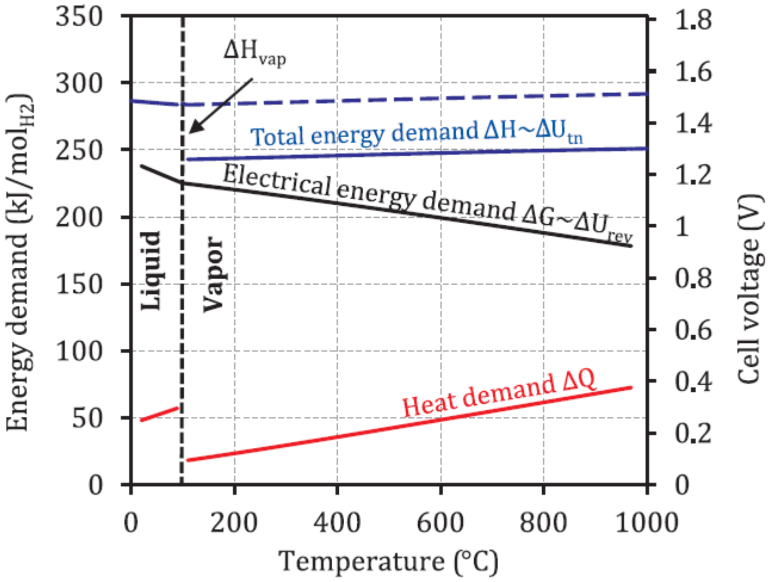

Figure 9. *Total ΔH (T), thermal ΔQ (T) and electrical ΔG (T) energy demand of the water dissociation reaction as function of temperature [30].*

Gibbs free energy change (ΔG), also describing the electrical energy required for the reaction, decreases with the rising temperature. At high temperatures (>2500 K) ΔG becomes negative. This would allow direct thermodissociation of water, but it is not considered practical application because of material issues. The required heat (ΔQ) for the reaction increases with temperature. Discontinuities at 100 °C temperature are due to water vaporization. Dashed trendlines show, how the state functions would behave without water vaporization. This would be the case in electrolysis of pressurized liquid water [11].

As Figure 9 shows, with higher temperature, less electricity is needed and more heat is used. Electricity is more expensive than heat so high-temperature water electrolysis is interesting from the economical point of view. High-temperature water electrolysis operated at 700 - 900 °C requires a heat source available at the site but also offers possible high utilization of internal heat losses. Furthermore, feeding the water in vapor form instead of liquid, some amount of the heat demand is supplied corresponding to the latent heat of vaporization [30].

The reversible cell voltage U_{rev} is the theoretical minimum cell voltage for water electrolysis operation. It is directly proportional to the electrical energy demand ΔG according to equation 1.

$$U_{rev} = \frac{\Delta G}{zF} \tag{1}$$

where z $(= 2)$ represents the total amount of electrons transferred per reaction and F is the Faraday’s constant 96,485 C/mol. As can be seen in Figure 9, when temperature rises, the U_{rev} decreases. U_{rev} ranges between 1.25 - 0.91 V in the temperature range of 0 - 1,000 °C. [30] The thermoneutral cell voltage (U_{tn}) represents the lowest voltage input for water electrolysis to occur in an ideal cell without heat integration (equation 2).

$$U_{tn} = \frac{\Delta H}{zF} \tag{2}$$

Here the overall energy demand ΔH (also heat) is supplied electrically. As Figure 9 shows, U_{tn} is approximately 1.47 - 1.48 V when feeding liquid water in under 100 °C temperature and 1.26 - 1.29 V in the temperature range 100 - 1,000 °C, if steam is supplied. Because of heat losses and thermodynamic irreversibilities, in real electrolyser the thermoneutral cell voltage is a little higher [30].

3.2 Electrochemistry

PEM is a low temperature electrolyser and is operated above U_{tn} cell voltages due to overvoltages or high internal losses. This leads to heating of cells and a need for cooling of the stack. The real cell voltage U can be calculated according to equation 3 [30].

$$U = U_{rev} + U_{ohm} + U_{act} + U_{con} \tag{3}$$

U_{ohm} is the overvoltage caused by the ohmic losses, which consists of resistance to the electrons flow in several cell elements like electrodes, interconnections and current collectors. It is mostly proportional to the current flowing through the cell. The term U_{act} is activation overvoltage and is caused by electronic kinetics. The charge has to overcome an energy barrier when transferring from reactants to the electrodes. This energy is dependent of catalytic properties of the electrode and behaves logarithmically. The term U_{con}, called concentration overvoltage, is caused by mass transport processes. Mass transport limits the process by reduced reactant concentration and simultaneously increasing the product concentration in the space between the electrolyte and electrodes [23].

Figure 10 illustrates the relationship between cell voltage and current, or current density (A/cm^2), at different temperatures and pressures (at 80 °C). Current density is often used to allow comparison between cells with different surface areas [23]. U_{tn} and U_{rev} are also represented in same operation conditions. Both voltages decrease slightly while temperature increases but U_{rev} is more sensitive for the change. Increased operating pressure conversely increases the U_{rev} and U_{tn}. The I-V curves show that when cell voltage is below U_{rev}, electrolysis reaction does not occur. But when cell voltage is between U_{rev} and U_{tn}, heat must be brought to the system for the electrolysis to take place [23].

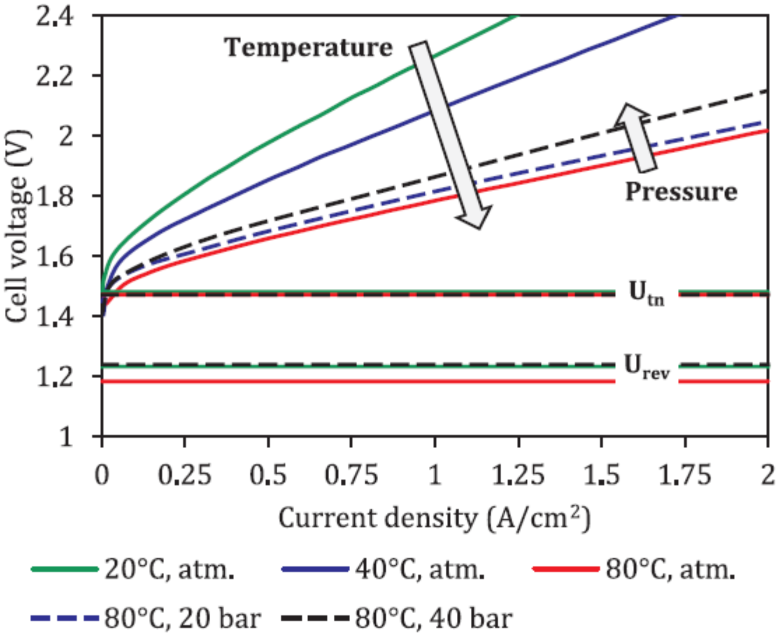

Figure 10. *Characteristic I-U -curve of PEM electrolysis cell with dependency of temperature and pressure [30].*

Near zero current density, the I-V curves behave logarithmically but as current increases, curves become linear. Activation phenomena are dominant in the beginning but as current increases, the ohmic and, in a lower degree, concentration phenomena become also substantial. Cell voltage decreases as temperature increases for any current [23]. However, cell voltage also increases with the pressure increase.

According to Faraday's law, the hydrogen production rate is approximately proportional to the current density. Nevertheless, for real electrolysis cell, the Faraday efficiency (η_F), defined as the ratio of real to theoretical H_2 production rate, must be added into operation. The difference between actual and theoretical production results from parasitic current losses in the gas pipes and cross permeation of gaseous products, which increases with rising temperature and pressure [30]. Maximum Faraday efficiency, usually above 0.95, is commonly achieved when electrolyser operates at rated production conditions [23].

The real hydrogen production rate can be calculated from equation 4:

$$f_{H_2} = \eta_F \frac{N_{cell} I_{cell}}{z F} \frac{22.41}{1000} \cdot 3{,}600 \, , \tag{4}$$

where N_{cell} is the number of cells in the stack and I_{cell} is the cell current in A [23]. The specific energy consumption C_E gives the energy consumed per hydrogen produced according to equation 5.

$$C_E = \frac{\int_0^{\Delta t} N_{cell} \cdot I_{cell} \cdot V_{cell} \, dt}{\int_0^{\Delta t} f_{H_2} \, dt} \tag{5}$$

C_E is usually expressed in units of kWh/Nm^3 or kWh/kg. This does not take in account the auxiliary equipment in the electrolysis system. Another key parameter of electrolyser is its efficiency η_E which expresses the ratio between the HHV or LHV of hydrogen (3.54 kWh/Nm^3 or 2.99 kWh/Nm^3) and the C_E in kWh/Nm^3 according to equation 6. Naturally, the efficiency can be calculated either for the whole electrolysis system or for the stack [30].

$$\eta_E = \frac{HHV\ or\ LHV\ of\ H_2}{C_E} \tag{6}$$

To increase the electrolyser's efficiency, the waste heat caused by overvoltages and internal losses remained in the products, could be recovered and utilized. Parameters such as operating temperature, effectiveness of heat exchangers and product flow rates affect the heat recovery practicality. PEM electrolyser cell temperature is typically between 50 - 80 °C [22], which sets limits for heat recovery temperature. Therefore heat integration is seen a more potential option in high temperature water electrolysis systems [31]. However, PEM electrolyser systems with waste heat recovery are also demonstrated. Power-to-Gas demonstration plant in Ibbenbüren Germany utilized waste heat from PEM electrolyser which increased efficiency 15 % [32]. Integrated heat recovery is also implemented in ITM Power multi-MW system designs 10 - 100 MW [33] and optional in H-TEC Systems products where system efficiency is claimed to increase up to 21 % with heat extraction [34].

3.3 PEM electrolyser system structure

PEM electrolyser system consists of the stack and balance of plant (BoP). The stack consists of cells which include membrane electrodes assembly (MEA), which is typically clamped between two porous current collectors. Two separator plates cover current collectors and separate two adjacent cells. BoP consists of the auxiliary equipment typically including: water treatment plant, gas/water separators, pumps, heat-exchangers, dryers/purifiers, gas storage, monitoring and ventilation systems. Equipment is system specific and the BoP for experimental 30 kW PEM electrolyser system is later explained in more detail.

Heart of the PEM electrolyser is the membrane electrodes assembly (MEA). It is composed of a membrane and two catalytic layers, electrodes, anode and cathode as illustrated in Figure 11. A thin (≈ 100 μm) proton-conducting polymer membrane works as a solid electrolyte and is placed between the electrodes. Its two main functions are to carry ionic charges and separate electrolysis products. That way it prevents hydrogen's and oxygen's spontaneous exothermic recombination back into water [11]. Common material for the membrane is perfluorosulfonate (PFS) polymer due to its mechanical strength, thermal and chemical stability, and good proton conductivity. Drawback are high cost and disposal because of contained fluorine in the structure [22].

The membrane lies between electrodes (anode and cathode) which are placed face to face and connected to an external DC power source. The electrodes commonly are made of noble metals such as iridium or platinum [23]. The following half-cell reactions occur at the anode and cathode:

$$anode: \quad H_2O_{(l)} \rightarrow \frac{1}{2}O_{2(g)} \ +2\,H^+ + 2e^- \tag{R8}$$

$$cathode: 2H^+ + 2e^- \rightarrow H_{2(g)} \tag{R9}$$

At the anode occurs oxygen evolution reaction (OER), where liquid water is introduced to the catalyst and DC current provides electrical work to split water into molecular oxygen, protons and electrons (R8). Solvated protons migrate through the membrane to the cathode side, due to an electric field. At the cathode, hydrogen evolution reaction takes place (HER), where protons are reduced to molecular hydrogen (R9) [11, 35].

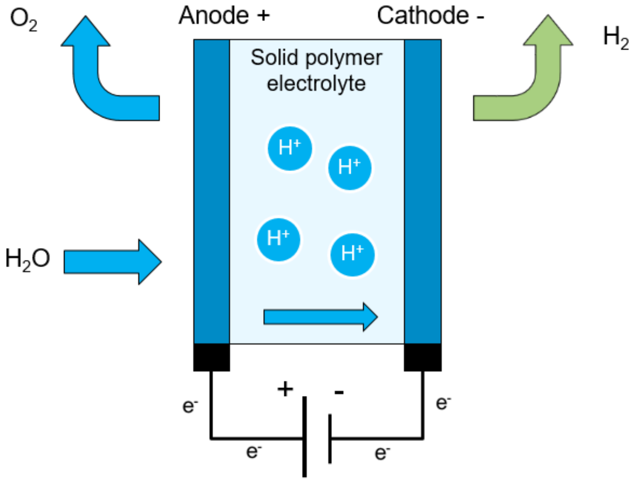

Figure 11. *Schematic cross section of MEA (adapted from [23]).*

Water is typically fed to the anode side of the cell in PEM water electrolysers. Figure 12 shows the parts of the entire PEM cell. The liquid water flows via separator plates' channels and diffuses through the current collector reaching the catalyst layer. At the catalyst, the water is split and the produced gaseous oxygen has to travel back, against the water, through the catalyst layer, current collectors, separator plates and the out of the cell. The electrons flow from the anode side, through the current collectors and separator plates, to the cathode side. The produced protons cross through the membrane to the cathode side. After reaching the catalyst layer, the combine with electrons to form hydrogen. Then the hydrogen must flow through the cathode current collector, separator plates and leave the cell [22].

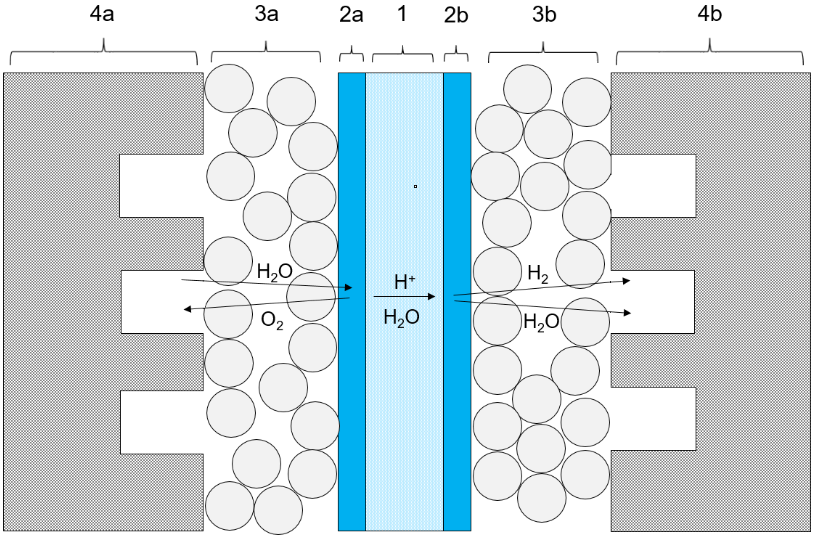

Figure 12. *Cross-sectional view of PEM electrolyser cell. 1 - Polymer Electrolyte Membrane, 2 - electrocatalytic layers (electrodes, a: anode and b: cathode), 3 - porous current collectors (a: anodic and b: cathodic side), 4 - Separator plates (a: anodic and b catodic side) (adapted from [11]).*

Important properties for current collectors (3a and 3b in Figure 12) are: good electrical conductivity, corrosion resistance, good mechanical support for the membrane and effective transport for the water and gases. The last property is emphasized at high current densities where the limiting factor is mass transport. The pore size and structure of the current collectors is key factor for optimization. Large porosity will increase the gas removal and reduce the volume of water trapped in the catalytic layers but will also decrease the efficiency by reducing the electron transport. The material used for about 0.8 - 2 mm thick current collectors must be corrosion resistant and a typical choice is thermal sintered spherically shaped titanium powder with pore size of 5 - 30 µm [22].

Separator plates (4a and 4b in Figure 12) along with current collectors contribute great deal to the stack cost, internal ohmic resistance and mass transport, which are controlling sources of irreversibility at higher current densities. The separator plates also must provide structure to the cell, offer insulation between oxygen and hydrogen and give a conductive route for the electrons and the heat. Nowadays material used for separator plates is titanium, graphite or a coated stainless steel which all are expensive choices [22].

3.4 Efficiency, voltage degradation, and stack lifetime

As stated before in chapter 3.2, the electrolyser stack and system efficiencies can be measured in terms of specific energy consumption (kWh_{el}/kg_{H2} or $kWh_{el}/Nm^3{}_{H2}$). The efficiency of commercial electrolysis systems and stacks are presented in Figure 13 as a ratio of specific energy consumption and LHV value (2.99 kWh/Nm^3) of hydrogen.

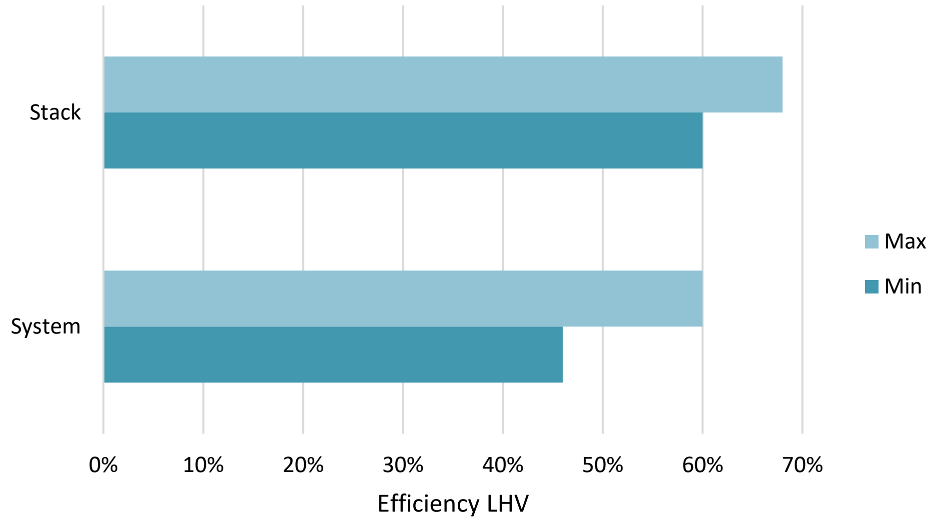

Figure 13. *Current PEM electrolyser stack and system efficiency estimates (LHV) [30].*

Stack efficiency ranges from 60 $\%_{LHV}$ to 68 $\%_{LHV}$. Systems efficiency, including stack, rectifier and utilities but excluding external compression, was in the range of 46-60 $\%_{LHV}$. Buttler et al. (2018) [30] based these values on different size electrolyser systems from different manufacturers. The efficiency points are assumed to be given at full load operation, which is the typical operation design point for commercial electrolysers today. Future system efficiencies were estimated by Bertuccioli et al. in study on Development of Water Electrolysis in the European Union (2014) [36]. The system efficiency was assumed to range between 62 - 75 % in 2030.

Conventional electrolysis system efficiency behavior is illustrated in Figure 14. Inefficiencies from the stack increase along with system load. This leads to higher system efficiency at lower system loads. However, inefficiencies from BoP are significant at minimum loads resulting to low efficiencies. Consequently, most efficient operation range is around 20 - 40 % load.

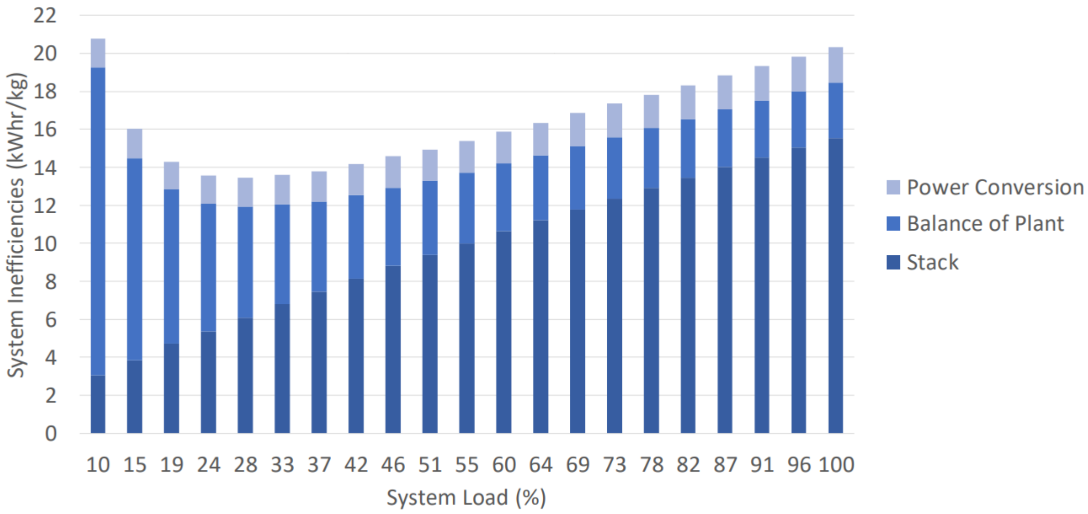

Figure 14. *System inefficiencies as a function of system load [33].*

Electrolysis systems could also be optimized for part-load operation, with ramp-up ability to full load operation, for a short periods of time, taking advance of fluctuating power input. This would require minimization of stand-by power and parasitic loads as pumps and inventers [36].

Voltage degradation is an important parameter for system efficiency. It refers to an increase of the cell overpotentials due to various degradation processes in the catalysts and membrane, as the cell ages. The voltage degradation leads to a reduced efficiency over the lifetime of the stack. For PEM technology, average status of voltage decay can be estimated to be below 4-8 µV/h. Over lifetime of 80,000 h operation, cell voltage increase results to an approximately 5-10 percentage points lower efficiency [30]. Data on long-term degradation for dynamic operation is practically unavailable. Voltage degradation as an efficiency drop is relevant to the stack lifetime since the stack typically does not fail suddenly. Other common reason for cell replacement is water contamination related user error [37].

Acceptable level of the efficiency drop depends on cost effectiveness of the replacement investment and what can be accepted by the operator [36]. Figure 15 shows estimates for PEM stack lifetimes according to different sources.

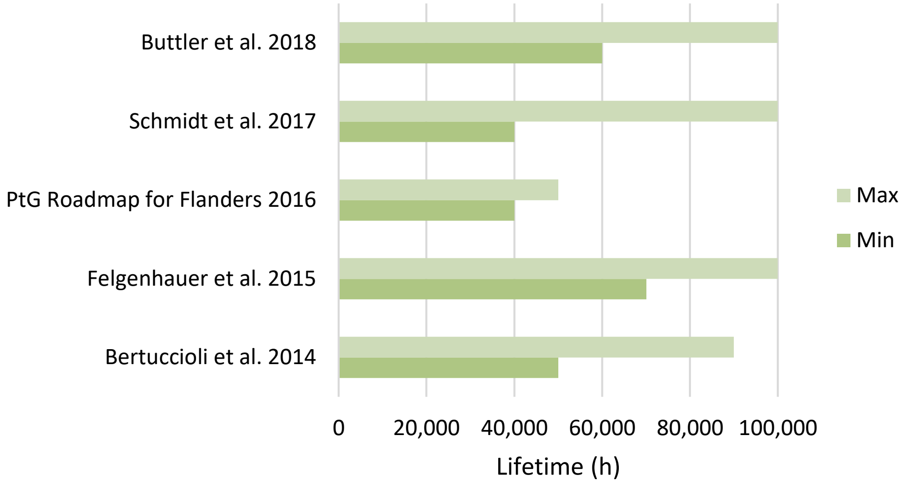

Figure 15. *PEM stack lifetime estimations for MW-scale systems (adapted from [30, 36, 38-40]).*

The minimum lifetime estimate for PEM electrolyser stack was 40,000 h and maximum lifetime was 100,000 h. All the stack lifetimes are for continuous operation and the lifetime under dynamic load may vary. Buttler et al. also mentions the lifetime for BOP to be on average 20 years [30]. Bertuccioli et al. gives 20 - 30 years lifetime estimate for the whole electrolyser system including several stack replacements or overhauls [36].

3.5 Hydrogen production cost

Hydrogen production cost sums up the costs to generate one unit of hydrogen at the production site. This parameter allows economical comparison with other hydrogen production methods. Major contributors to the hydrogen cost in PEM electrolysis are electricity price, capital costs, operating costs, utilization rate and possible secondary revenue sources such as grid balancing services and selling the oxygen. These parameters are introduced in the following chapters. Lastly, a comparison between SMR, alkaline electrolysis and PEM electrolysis is made. These technologies were chosen for comparison due to their mature technology and commercial status in hydrogen production.

3.5.1 Capital expenditures and operational expenses

Capital expenditure (CAPEX) are typically high for PEM electrolysis due to expensive materials in cell stack. Figure 16 shows the future estimates and targets for PEM electrolyser capital costs (€/kW_{el}). The expected cost development trend in the Figure 16 is also in line with the IRENA, which estimated in the technology outlook for the energy transition (2018) [21] the capital costs in 2017 for multi MW system to be around 1,200 €/kW.

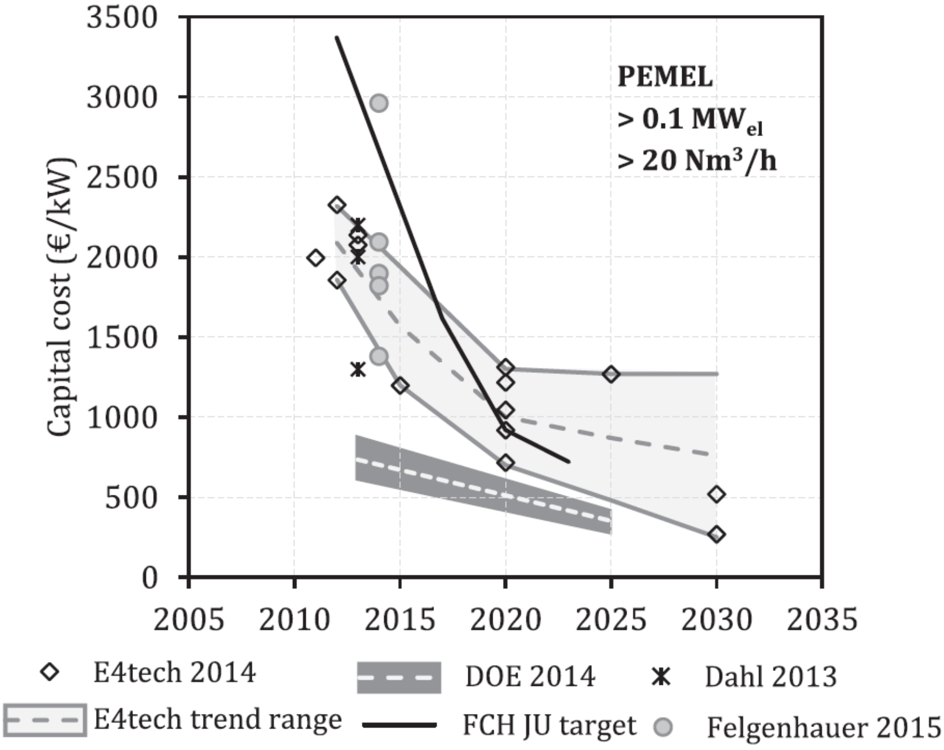

Figure 16. Expected future trend for PEM electrolyser system capital costs (adapted from [30]).

It is worth to note, that electrolysers are still built in small volumes for niche markets, which puts the large expected costs reductions into context. Significant cost reductions are expected to come from technology innovations, improved supply chains and bigger production volumes, which enable more efficient manufacturing techniques [36]. Mass production and supply chain development will follow with increased deployment of electrolyser systems and can allow the use of standardized mass manufacture components for lower costs. However, technology innovations might have the greatest impact on capital costs [38]. The system's current density also affects the capital cost. Increased current density results to reduction of capital cost but also increases the average cell voltage. Consequently, operational costs increase.

The data in Felgenhauer et al. 2015 [40] suggests that PEM electrolysis systems follow economy-of-scale behavior. Investment costs range from around 1,300 €/kW to 3,000 €/kW for electrolyser with hydrogen generation capacity of 21.6 kg/h and 9.0 kg/h respectively. However, the economy-of-scale effect is limited for electrolyser systems due to linear production dependence on the electrolysis cell area [30]. Similar behavior of PEM electrolyser systems' capital costs was found in data from International Energy Agency (IEA) Proost J. et al. Hydrogen Implementing Agreement (HIA) Task 33 [26]. The data showed steep decrease in capital cost (from 4,000 €/kW to 1,200 €/kW) as power input increased from 100 kW to 2,000 kW.

Figure 17 shows that the specific investment development for larger system sizes, up to 100 MW, is in line with data from the Task 33. Cost reductions as a function of system size become less significant in >10 MW systems.

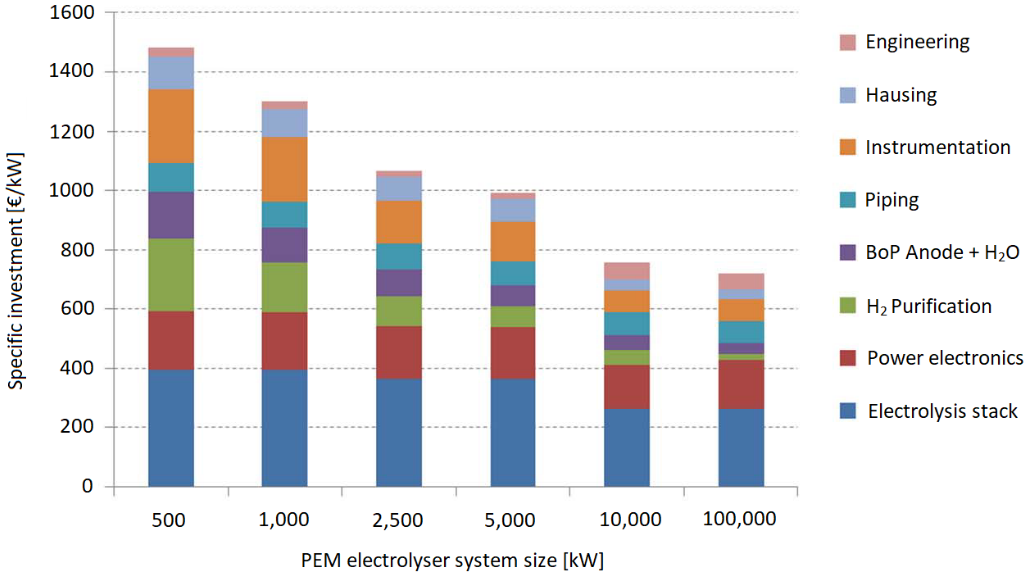

Figure 17. *PEM water electrolysis specific investment as a function of system size [41].*

In order for water electrolysis to become a feasible technology for hydrogen production, it is mandatory to operate at MW-scale range. This is not only in terms of investment costs but also a prerequisite to be able to couple with MW-scale renewable energy sources [26]. Currently, MW-scale Power-to-X applications require multi-stack systems since single-stack units limit to 1 - 3 MW power input. Modular design approach can be used to reach system sizes of up to 100 MW in the future [31]. Multi-stack usage enables larger MW-systems but affects the expected decreasing trendline of specific investment (€/kW) by reducing the cost decrease as more stacks are included into the system [26].

Figure 18 shows a breakdown of the cost for a conventional PEM electrolyser system and stack. About 60 % of the electrolyser's system cost originate from the stack. Balance of the plant and the power electronics share the second largest part of the capital costs with 15 % cuts. Balance of plant cost is expected to reduce as systems are scaled up, because current small volume systems commonly use custom parts [42]. specific

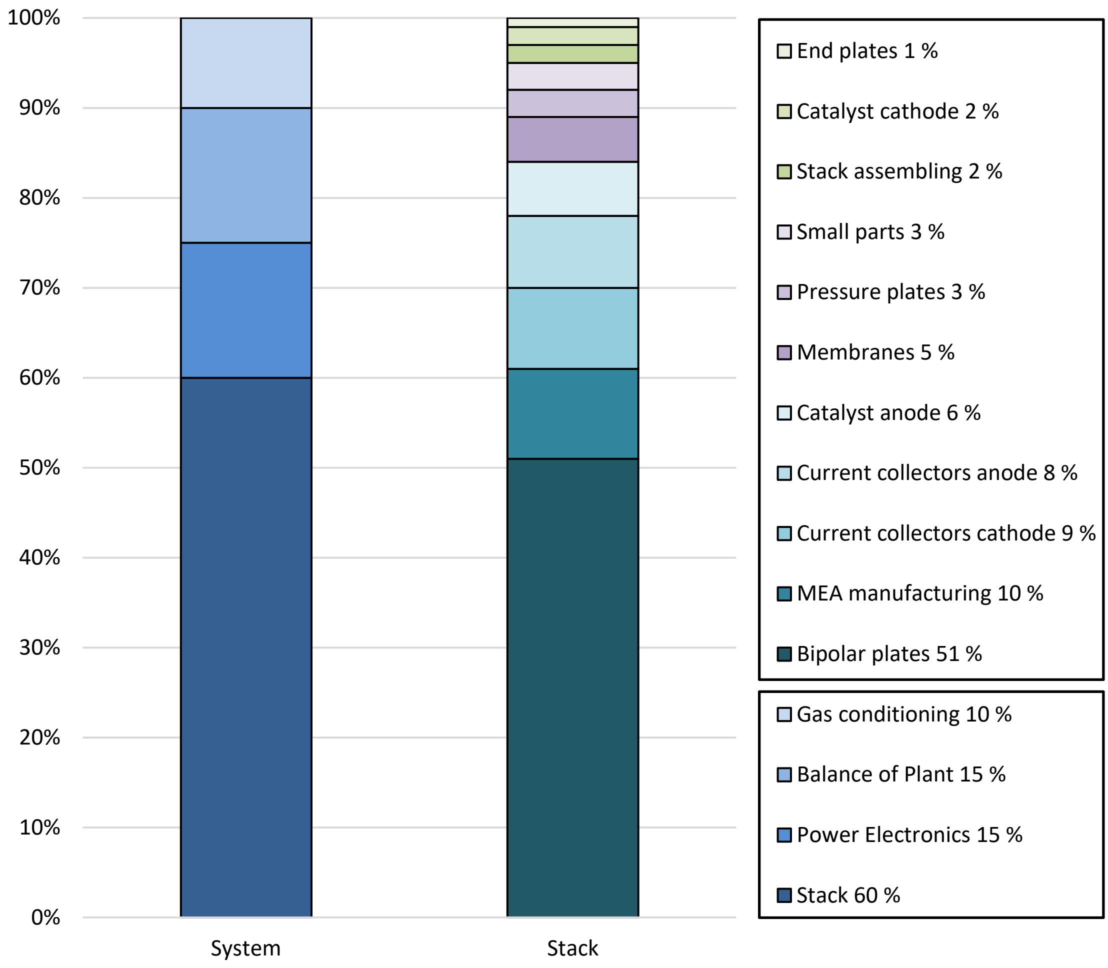

Figure 18. *PEM water electrolyser system and stack cost breakdown [36].*

A further directional breakdown for the cost of the electrolyser stack cost shows bipolar plates 51 % cover major part of the stack capital costs. Materials and geometric requirements make these components expensive to manufacture. Current collectors (cathode 9 %, anode 8 %) and catalysts (cathode 2 %, anode 6 %) have substantial shares of the stack cost. Also MEA manufacturing contributes noteworthy 10 % part to the expenses. Cost shares vary depending on manufacturer's design [36].

Catalyst materials for the OER are expensive and rare noble elements iridium and ruthenium. Similarly HER electrode materials palladium and platinum are expensive and consequently research is done to reduce the usage of these materials [36]. Furthermore, increased current density (>3 A/cm^2) and replacement of titanium in bipolar plates (51 % of the stack cost) with low-cost substrate like steel is researched to reduce capital costs [38].

Here, operational expenses (OPEX) are defined to consist of planned and unplanned maintenance/spare parts and operation/labor [40]. According to Buttler et al. [30] calculations based on Felgenhauer and Hamacher [40] PEM electrolyser OPEX costs are 2 - 3 % of CAPEX excluding electricity. Additionally, the OPEX values show similar behavior as CAPEX values, tending to reduce as the capacity of the electrolysis system increases [30].

Similar OPEX value assumptions was made in Roadmap to Flanders report [39]. For PEM electrolysis, OPEX was distributed in MW-scale and multi-MW scale categories. MW scale values for years 2015 and 2030 were 60 €/kW/a and 48 €/kW/a, respectively. Multi MW-scale category OPEX values for same years were 40 €/kW/a and 32 €/kW/a. Moreover, because of the wear and spare parts, 1/3 of the OPEX was expected to be fixed and 2/3 to be proportional to operating hours per year.

3.5.2 Electricity price

The dominant parameter for hydrogen cost of a PEM electrolyser system, at high utilization rate, is considered to be the electricity price. The total electricity price consists of energy price, network charges and taxes. Average spot price in Finland 2018 was 46.80 €/MWh. Electrolyser operation could be entitled to the lower tax category II, which is 0.87172 c/kWh including VAT 24 % [43]. Network charges depend on service provider and are estimated to be 15 - 18 €/MWh including tax and tax returns.

The volatility of electricity prices is important in optimizing the operation strategies. According to the European Network of Transmission System Operators for Gas (ENTSOG), around 50 % of electricity demand will be covered by renewable energy by 2030. Depending on the scenario, this share will rise between 65 - 80 % by 2040 [44]. Drive for decarbonization of the electricity sector (EU renewable energy target of 32 % for 2030 [45]), while wind and solar technologies are becoming more cost-effective, makes this scenario very possible [46].

In Finland there were 700 wind power plants with combined capacity of 2,044 MW in the end of the 2017. 4.8 TWh of electricity was produced with wind power, which covers 5.6 % of Finland’s total electricity consumption. At the moment (15.11.2018), nine new wind power plant investments, with combined power output of 340.2 MW, have been launched and started construction. All these new projects are executed without governmental support. In addition, there are plans to build approximately 15,500 MW of wind power of which about 2,700 MW are off-shore wind turbines [47]. Combined solar power capacity in 2017 was approximately 70 MW. In Finland, solar power has strategically minor role and it is considered insignificant at least until 2025 if additional government support is not introduced [48].

Against this background, one can assume that growing intermittent power production and volatility in electricity prices will increase. Consequently, a chance that consumption and production will not meet, increases. This results in a need for energy storage and grid balancing, which gives an opportunity for the Power-to-X technologies to grow.

Annual operational hours, which can be expressed as a utilization rate, can have a significant impact on hydrogen production cost. Utilization rate also determines if the costs are dominated by CAPEX or by electricity cost, as shown in Figure 19.

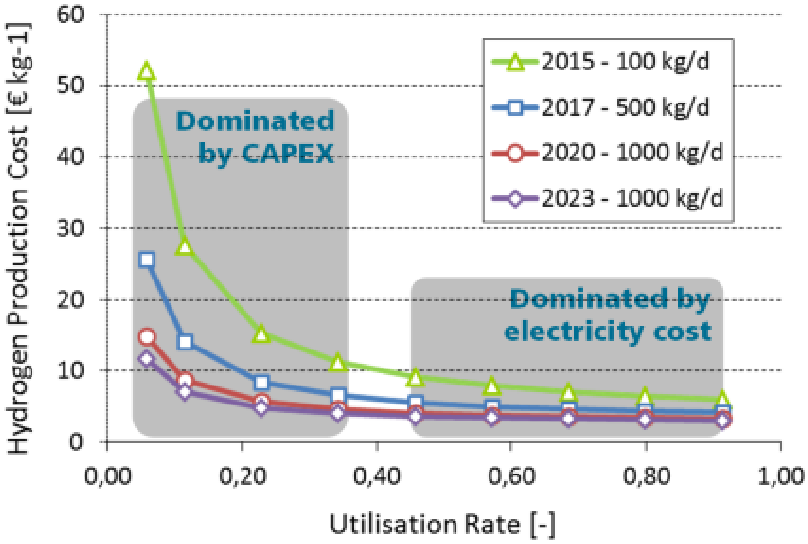

Figure 19. *The influence of the utilization rate on hydrogen production cost [41].*

High volatility in electricity prices allows electrolyser operators to optimize hydrogen production in times of lower electricity cost. However, low utilization rate leads to rapidly growing hydrogen production cost due to CAPEX costs.

Industrial applications typically require constant hydrogen production which leads to a high electrolyser utilization rate. Such applications' hydrogen production cost is dominated by electricity cost, thus high electrolyser efficiency is essential as it determines the electricity consumption [49].

3.5.3 Frequency Control Reserve markets

Strategy to lower the cost of electrolytic hydrogen is to providing grid balancing services to network operator for a payment. Frequency control reserve markets in Finland and in Nordic power system consist of Frequency Containment Reserve (FCR) and Frequency Restoration Reserve (FRR). FCR is divided in normal operation containment reserves (FCR-N) and frequency containment reserves for disturbances (FCR-D). They are used to control frequency and keep it 49.9 - 50.1 Hz.

While operating electrolyser in the FCR-N, it should be noted that offered power reserve must be symmetrical. If the frequency drops below 49.95 Hz, up regulation is needed and electrolyser has to decrease power. Consequently, if the frequency exceeds 50.05 Hz, down regulation is used and the electrolyser power is increased. Reserve must be able to operate up and down regulation as illustrated in Figure 20. Up and down regulation are needed almost equally according to Fingrid's data on FCR-N, procured volumes in hourly market (2018) [50].

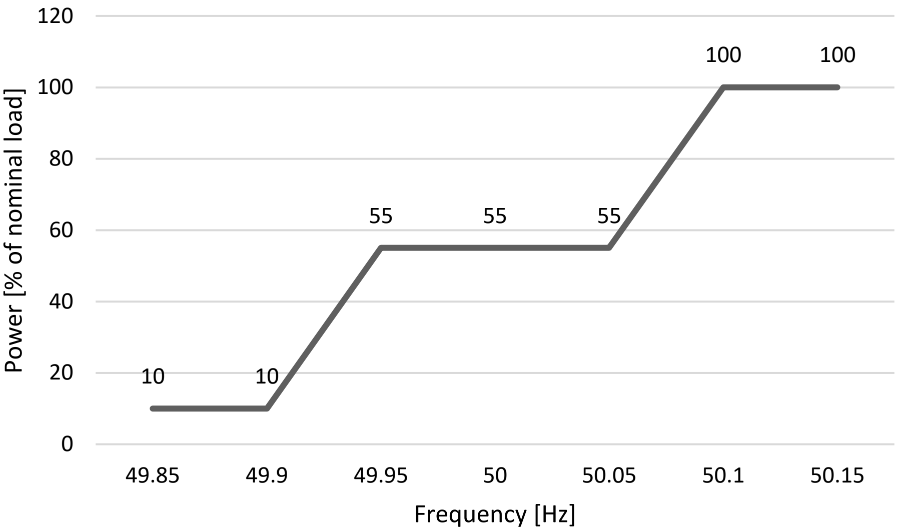

Figure 20. *Electrolyser operating in FCR-N. Load adjustment must have reserve to do up regulation during decreased frequency and down regulation during increased frequency.*

Electrolyser has to operate at part-load which can, according to Figure 14, affect the electrolyser efficiency. Operating at 55 % load offers 45 % up and down regulation of the nominal capacity. Technical requirements and price levels for FCR-N in 2019 are presented in Table 2.

Table 2. *Technical requirements and price levels for frequency containment reserves in Finland [51].*

	Minimum size	**Full activation time**	**How often activated**	**Price level 2019 / 2018**
FCR-N	0.1 MW	3 min	Several times a day	13.5 €/MW, h (yearly market) 12.76 €/MW, h (average hourly market 21.1.2019) 23.6 €/MW, h (average hourly market 2018)

Electrolyser operator may offer its capacity to hourly and/or to yearly market. In yearly market, the fixed price is valid throughout the year. The price is set based on the highest approved bid in yearly bidding competition. In hourly market, the payment is set based on the most expensive bid separately for each hour. Operators that have a yearly agreement can participate in the hourly market if they have fully supplied the agreed amount of reserve already in the yearly market.

3.5.4 Oxygen

In the water electrolysis reaction for every 1 mol of H_2 produced, ½ mol of O_2 is also generated. Oxygen (O_2) is a highly oxidizing, colorless and odorless gas in standard conditions. Oxygen has largest global industrial gas market share of 26 %. Global demand was estimated to be 19.2 billion USD in 2017 [52]. The price for one ton of O_2 varies in literature and depends on form (gaseous/liquid) and end-use application. Thomas, D et al. assumes the selling price for oxygen in Roadmap for Flanders (2016) to be 24.5 €/t [39]. Breyer C. et al. (2015) estimated in Power-to-Gas profitable business calculations [53] oxygen value to be 80 €/t. Higher selling price was assumed by Hurskainen et al. in Industrial oxygen demand in Finland (2017) [54] for liquid oxygen in medical use (250 €/t) and in small scale waterworks use (235 €/t). Significantly lower selling price was assumed for larger scale use, such as pulp mills.

Currently, the two main conventional methods for oxygen production are vacuum pressure swing adsorption (VPSA) and cryogenic air separation. The ambient air, of which oxygen makes up 21 %, is generally used as a feedstock for oxygen separation. Purity rates for technologies are <95 % and >99.5 % respectively. Cryogenic units are more mature technologies but less flexible than VPSA. Thus cryogenic technology is more suitable for large-scale (>100 tons/day) oxygen production and VSPA technology for medium-scale production (20 - 100 tons/day) [54].

In a report on industrial oxygen demand in Finland (2017), the total estimated oxygen consumption in Finland is around 1.3 Mt annually. Sectoral oxygen consumption in Finland is shown in Figure 21. Most of the industrial oxygen is used in steel industry, metal refining and pulp & paper industries. Other smaller scale sectors are chemical, glass and medical oxygen industries [54].

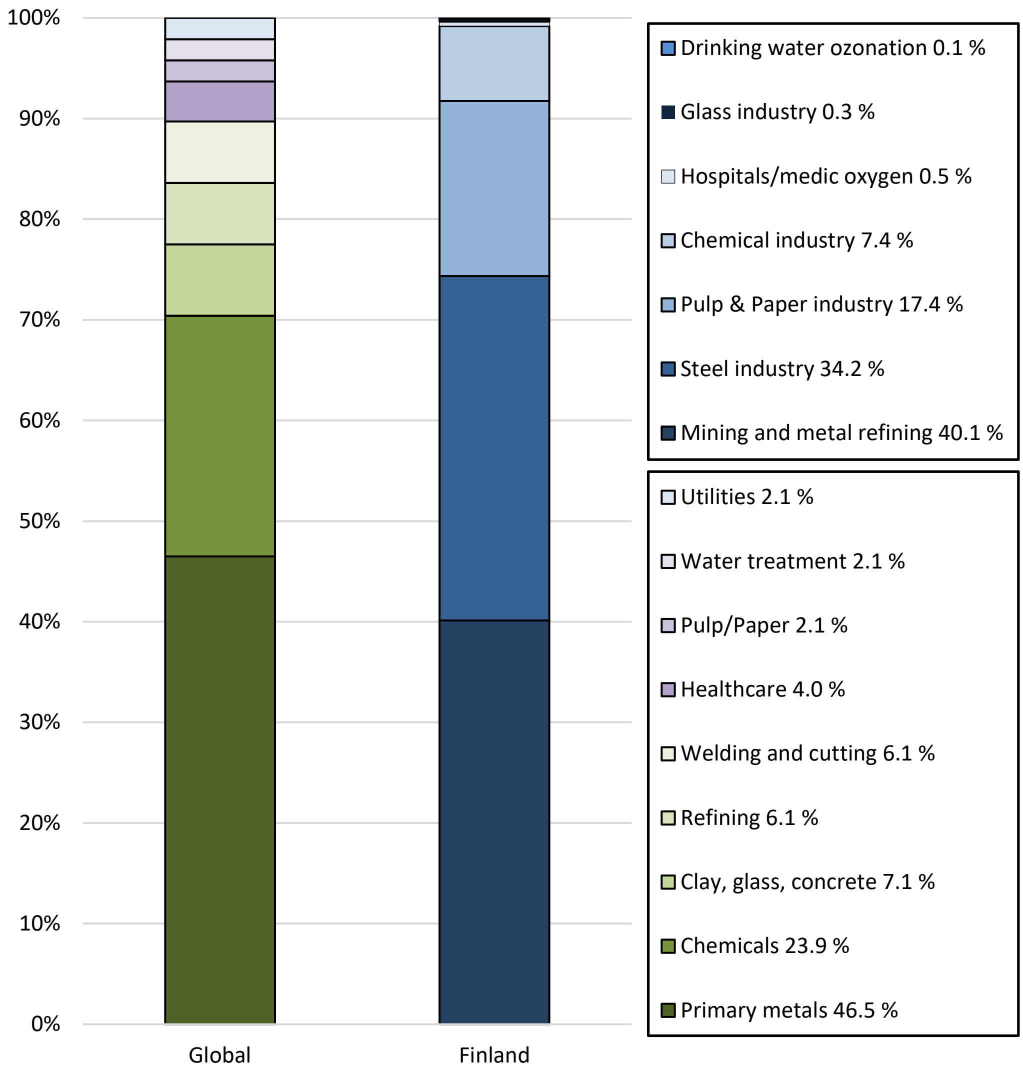

Figure 21. *Oxygen consumption (t/a) by sector globally and in Finland (no data from the oil refining sector) [52, 54].*

Best scenario for PEM water electrolysis by-product oxygen demand would be a consumer with high volume and high purity requirements. However, practically these kind of situations are rare. High purity oxygen is used in laboratories and in semiconductor industry where markets are assumed to be small. Potentially most promising end-users for water electrolysis by-product oxygen in Finland are reported to be pulp mills due to sufficient scale of oxygen demand (8,000 - 15,000 t/a) and current use of delivered liquid oxygen [54].

3.5.5 Hydrogen production cost comparison

SMR technology is very mature technology and the cost of hydrogen production is typically 1-3 \$/kg (0.9-2.7 €/kg) [26]. The cost depends on regional natural gas prices, taxes and CO_2 allowance prices. In case of CO_2 emission allowance cost increase, SMR can be coupled with carbon capture and storage (CCS) technology, which will add costs and can result up to 3.4 €/kg hydrogen production cost with high natural gas cost [55]. As the SMR is currently the main hydrogen production method with share of 68 % globally and 86 % in Finland, the large-scale facilities and infrastructure already exists and therefore SMR is going to remain relevant in the near future.

The main parts of the hydrogen cost for water electrolysis technologies are electricity cost, investment cost and operational expenses such as maintenance, labor, water and other operation resources. Alkaline electrolysis has been in use for decades in different applications, especially in chemical industry, and is more mature water electrolysis technology than PEM electrolysis. Longer history in development and better established volumes in manufacturing have led to lower capital costs and longer lifetime for alkaline electrolysers. Nevertheless, the PEM electrolysis is more flexible in power ramps, starts and stops, which are becoming more important properties with increasing fluctuating renewable energy production (Table 3). This enables PEM electrolyser to be able to provide capacity for frequency containment reserve for extra revenue. PEM technology also has more potential for high pressure operation (up to 200 bar) than alkaline electrolysis. In addition, capital costs for PEM electrolysis have dropped notably in recent years and future estimates show more reductions in costs. [21, 56]

Table 3. *Dynamic properties of alkaline and PEM water electrolysis [36]*

	Alkaline water electrolysis	**PEM water electrolysis**
Start-up time [minutes]	20 - several hours	5-15
Ramp-up time [%/sec]	0.1 - 25	10 - 100
Ramp-down rate [%/sec]	25	10 - 100

Load factor or utilization rate determines affects the hydrogen cost significantly. Revenues come from the hydrogen value and possible oxygen and heat utilization value. Cost reduction strategies for PEM electrolysis such as grid services and operating window optimization can also be implemented in electrolysis operation. Due to lack of flexibility, the alkaline electrolysers are assumed to run as grid-connected and do not receive revenues from frequency containment reserve services.

Hydrogen production costs of alkaline and PEM electrolysis technologies are compared in International Renewable Energy Agency (IRENA) report (2018) [21]. Scenario trendlines for years 2017 and 2025 illustrate the hydrogen cost development as a function of load factor (Figure 22). Scenarios use electricity prices from Denmark 2017 which represents grid with a high share of renewable power generation [21].

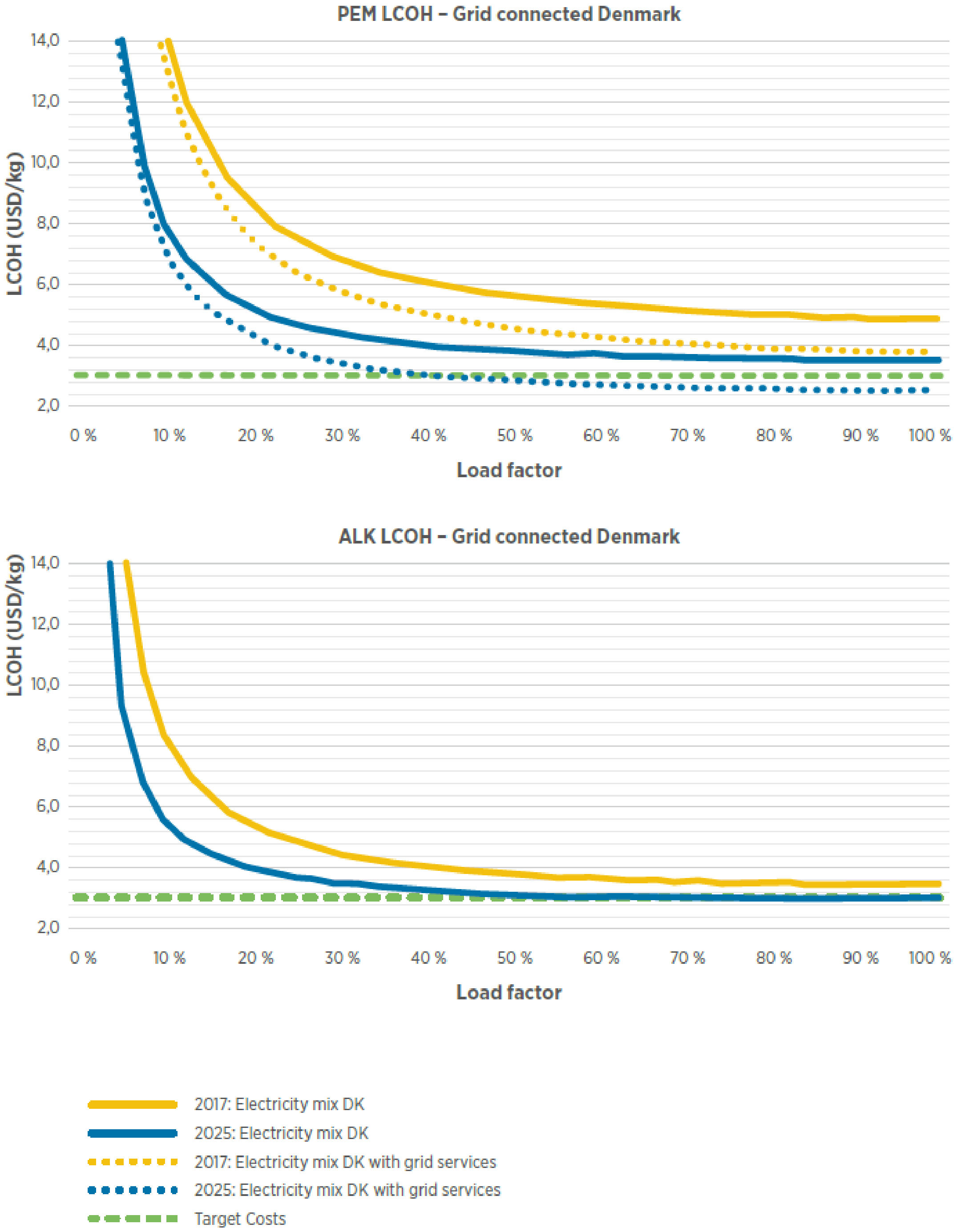

Figure 22. *Levelised cost of hydrogen (LCOH) as a function of load factor for years 2017 and 2025 with electricity Denmark 2017 electricity prices [21].*

Comparison shows that for the alkaline electrolysis, the hydrogen cost drops steeply at 10 % load factor, flattens at 50 % and reaches 3 - 3.5 $/kg (2.6 - 3 €/kg) cost at 100 % utilization rate. PEM electrolysis has additional scenarios with FCR grid services for extra revenue. Hydrogen cost drop is more flat for PEM electrolysis and for 2017 scenario remains trend reaches around 5 $/kg and 4 $/kg (4.4 €/kg and 3.5 €/kg), with grid services, at 100 % load factor. PEM 2025 scenario shows more steep and early drop in the cost and is similar to alkaline electrolysis trend. Without grid services, trend reaches around 3.5 \$/kg (3 €/kg) hydrogen cost and with grid services cost falls under 3 \$/kg (2.6 €/kg) [21].

Table 4 sums up the key parameters for technologies in comparison. Hydrogen cost values are converted from USD to EUR with 30.1.2019 rate (1 USD = 0.8762 EUR). Water electrolysis technologies' hydrogen costs are dependent on various factors. Values are calculated with Denmark 2017 electricity price, near 100 % utilization rate and 20 MW capacity. PEM has additionally hydrogen cost value with grid services which assumes USD 18/MWh FCR bid for every hour it is in operation.

Table 4. *Key parameters for SMR, AEL and PEM hydrogen production technologies ([9] unless otherwise stated).*

Method	**Maturity**	**Capacity**	**System efficiency (LHV)**	**Lifetime**	**Hydrogen cost (2017)**
SMR	Mature	150 - 300 MW	70 - 85	30 years	0.9 - 2.7 €/kg
AEL	Mature	Up to 150 MW	51 - 60 [30]	55,000 - 120,000 h [30]	3.2 - 3.6 €/kg
PEM	Early market	Up to 20 MW*	46 - 60 [30]	60,000-100,000 h [30]	4.8 - 5.0 €/kg 3.6 - 4.0 €/kg (grid services)

*Currently, the largest PEM electrolysis plant with 20 MW capacity is planned to be built in Canada [57].

4. POWER-TO-X

Power-to-X (PtX) technology converts the electric energy into various end-use products and especially stores renewable surplus energy. The focus in this thesis is a pathway combination of "Power-to-Hydrogen", where hydrogen is generated via PEM water electrolysis, and "Hydrogen-to-X", where hydrogen is utilized directly, or converted into different end-use products. This could be a flexible way to store energy, cut carbon emissions and combine power grid with gas grid. Different pathways are illustrated in the Figure 23.

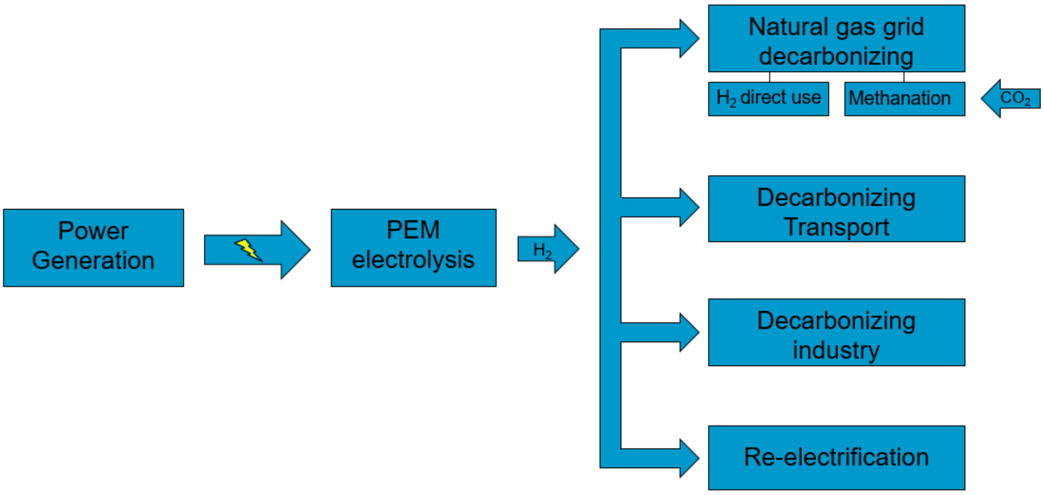

Figure 23. *Different pathways Power-to-X from power generation to various hydrogen end-use applications.*

Drive for Power-to-X technologies come from ambitious targets to achieve environmental targets. According to Hydrogen Roadmap Europe (2019) [58], especially in EU region, Power-to-X concept has potential to grow due to commitment to reduce emissions by 80 - 95 % compared to 1990 levels by 2050. Also several EU-wide research programs with high investments have been established lately. Other noteworthy countries, such as China, Japan, South Korea, Australia and USA are mentioned to pursue towards hydrogen technologies.

Finland's actions to achieve targets in Government Programme and EU for 2030, and proceed consistently towards 80 - 95 % mitigation in GHG emissions by 2050 are outlined in the report on National Energy and Climate Strategy for 2030 (2017) [59]. According to report, the minimum of 250,000 electric (includes electric cars, hybrids and hydrogen fuel cell vehicles) and 50,000 gas-powered (methane) vehicles will be implemented, share of transport biofuels is increased to 30 % and obligation to blend 10 % of bioliquids in light fuel oil, used in machinery and heating, will be introduced. Also, use of the imported oil is to be halved.

In terms of road transport, Finland's national target for 2050 is near-zero emissions. In 2030, all new cars sold in Finland are proposed to be compatible with some type of alternative fuel: electricity, hydrogen, natural gas, biogas or liquid biofuels even in high concentrations. The national program proposes alternative fuel distribution network consisting of public refueling points for natural gas, biogas and additionally there should be around 20 refueling stations for hydrogen by 2030. This network would be built mostly on market terms and by energy companies or other commercial operators. National and/or EU support can be used in the construction phase. [59, 60]

Following chapters introduce different hydrogen end-use sectors and the first research question "What Power-to-X pathways are considered potential for PEM electrolysis and what noteworthy projects have been demonstrated recently?" is answered. Examples of recent global Power-to-X demonstration projects with PEM water electrolysis, paving the way for self-sustaining market, are also presented in chapters and more comprehensive list of PtX projects are presented in Appendix 1.

4.1 Hydrogen decarbonizing natural gas grid

Hydrogen can be implemented into natural gas grid directly or after converting it into synthetic methane using CO_2. The rationales for methanation are: limited or missing hydrogen infrastructure, including hydrogen grid, storage and end-use technologies [61]. Hydrogen can be also directly injected into natural gas grid up to certain levels, depending on several factors described later. Both methods contribute to reducing CO_2 emissions related to natural gas usage.

Natural gas has existing infrastructure in many countries which makes it economically interesting solution for renewable energy storage. Natural gas is also main method for heating buildings in Europe (42 %) [58]. In the short term, hydrogen injection in the gas grid is a low-value, low-investment step to support the early-stage hydrogen production. In the long run, direct hydrogen or synthetic natural gas injection can be a way to store significant amounts of renewable energy. Alone in the EU, the amount of stored energy in gas grid in the form of methane was around 1,200 TWh in 2017. Also by using existing gas infrastructure expensive electricity grid upgrades can be avoided [21].

4.1.1 Synthetic natural gas via CO_2 methanation

Methane is globally an important energy carrier to the energy, industry and transportation sectors. Nowadays major share from the natural gas are from fossil sources. A way to decarbonize the gas grid is to synthesize H_2 produced via electrolysis with CO_2 to produce synthetic natural gas. Either chemical or biological catalytic reaction can be used [61]. A simplified process pathway from renewable power to synthetic natural gas (SNG) process is shown in Figure 24.

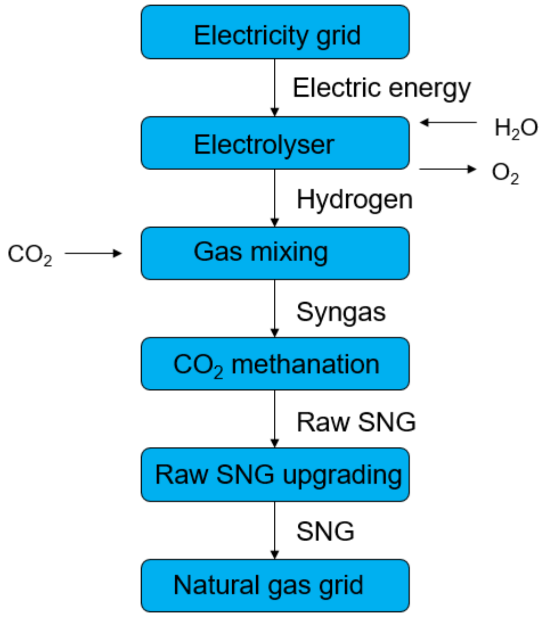

Figure 24. *A simplified process pathway from electricity to synthetic natural gas (adapted from [62]).*

Sites emitting considerable amounts of useable CO_2 for capture, available excess renewable energy sources and possibility for natural gas feeding points into the grid are suitable for this pathway.

Project involving synthetic natural gas manufacturing and injection into gas grid was demonstrated in Allendorf, Germany 2013-2016. In project BioPower2Gas surplus renewable electricity was used to produce hydrogen via PEM electrolyser (400 kW_{el} and 60 Nm^3/h_{H2}) which was methanized using CO_2 from biogas plant by microbiological means. The maximum synthetic methane production rate was 15 Nm^3/h and the gas was injected into national gas grid to serve natural gas customers and fuel markets [63]. Other project in smaller scale was established in Rozenburg in the Netherlands. In Rozenburg 7 kW PEM electrolyser was used and produced synthetic natural gas was injected in Dutch gas grid (35 % efficiency) [64].

Upcoming industrial-scale PtG demonstration Jupiter 1000 in France started its construction phase in December 2017. Commissioning for the project is planned to take place in 2019. Wind farms in the surrounding region generate renewable electricity for green hydrogen and synthetic natural gas production. 1 MW_{el} electrolysis plant will have two electrolysers (alkaline 0.5 MW and PEM 0.5 MW) with 200 m^3/h hydrogen output and a methanation reactor which will combine the hydrogen and CO_2 with 25 m^3/h SNG production. CO_2 is captured from nearby industrial plants. SNG and also part of the hydrogen will be injected into the gas grid [65].

4.1.2 Direct use of hydrogen and injection into the gas grid

According to DVGW research report (2013) [66], existing studies imply that the current natural gas infrastructure is largely suitable for approximately 10 vol-% H_2 in natural gas. Theoretically, this would correspond to roughly 30 TWh of hydrogen in EU 1,200 TWh natural gas grid. However, there are several limitations for the blending and need for research with the regard to some important elements, for example the natural gas storage, particularly pore storage, gas turbines and natural gas tanks. There is also significant variety in maximum hydrogen blend levels, even within European countries, due to natural gas network system and local natural gas composition [58]. The maximum share of hydrogen in natural gas is set to 1 % in Finland [67].

Figure 25 shows the essential elements of the natural gas infrastructure and their researched limits of H_2 compatibility. They constitute first reference values, but in particular research and investigations are still to be carried out to assess the influence of fluctuating concentrations. The evaluation of existing data covers a concentration range from 0 to 70 % by volume of H_2. The suitability of individual elements of the supply chain for higher H_2 concentration is also possible, but must be examined on a case-by-case basis. Despite the optimistic underlying trend, further research can show that dissimilar components and infrastructure parts are not tolerated to the expected extent of H_2, and their adaptation can lead to economic challenges [66].

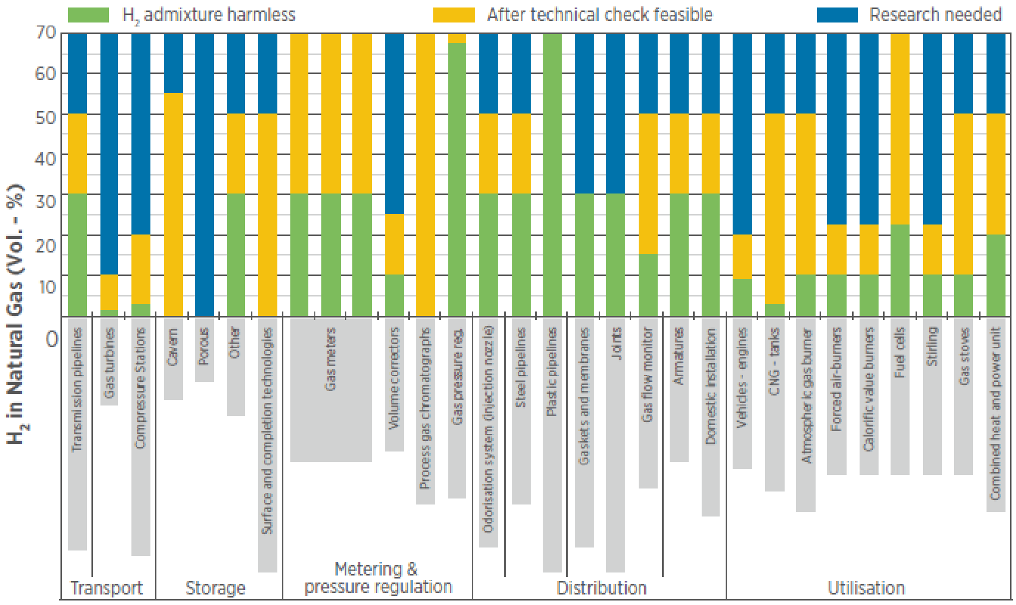

Figure 25. Compatibility of hydrogen with selected components in natural gas grid [21].

Hydrogen's lower energy density compared to natural gas reduces the grid capacity. The reduction in transport capacity, by the addition of 10 % by volume of H_2, is about 5- 6 %. This is initially a purely mathematical reduction attributable to the altered material properties of the mixture, particularly the lower density. If the aim is to maintain the original natural gas transport capacity with admixture of H_2, which therefore requires a higher volume rate because of a decrease in the density, this can only be realized via a stronger pressure on the line segment [66]. At 20 % blend flow rate needs a 10 % increase to offer same energy for end-user [9].

UK has been undertaking the Iron Mains Replacement Programme. The aim of the programme is to upgrade the majority of gas distribution pipes to polyethylene, which is considered to be suitable for 100 % hydrogen. At the moment, this programme is over 50 % complete. Project relating to hydrogen injection into gas grid, the H21 Leeds City Gate project, is a study with a purpose to determine the feasibility of converting the existing natural gas grid in Leeds to 100 % hydrogen. H21 project has shown that the conversion of the UK gas network to hydrogen would reduce UK emissions with 73 % from the heating, transport and power generation [68]. Project HyDeploy hosted on the Keele University campus in Staffordshire will explore the potential of hydrogen blending up to 20 % into the natural gas grid. Trial of hydrogen blending into gas grid begins in summer 2019 and will run a 12 month period. Currently project is at building phase where hydrogen production and supporting equipment are constructed in the site [69].

Germany is also very active on the Power-to-X sector because of increasing amount of renewable energy production and several notable projects have been established during last years. Energiepark Mainz, a large Power-to-Gas plant with 6 MW PEM electrolysis system, was commissioned in 2015 to explore the possibilities of PEM electrolysis technology. According to Kopp M. et al. Energiepark Mainz: Technical and economic analysis of the world wide largest Power-to-Gas plant with PEM electrolysis [8], plant is connected to medium-voltage grid and nearby 8 MW wind farm network. At peak power 6 MW, the hydrogen production efficiency is 59 % but at rated 4 MW power input, the efficiency increases to 64 %. Produced hydrogen can be injected into local natural gas grid (H_2 fraction of 0-15 % [70]) or filled into trailers and delivered to the chemical industry or fueling stations. The research facility is also examining the provision of power grid services and storing renewable energy.

In Ibbenbüren an innovative PEM technology is demonstrated to feed hydrogen into the local gas grid. The 150 kW_{el} electrolyser with hydrogen production rate of 30 Nm^3/h at 14 bar supplies hydrogen into the gas pipes without additional compression. Hydrogen concentration in the gas grid is kept approximately at 1 vol. -%. Electrolyser system with high 71 % efficiency is as well equipped with waste heat recovery (15 %) which increases the overall efficiency to 86 % [32]. The waste heat was supplied to the neighboring grid operator to be used in gas pressure regulation and measuring system for preheating purposes, which reduces the gas consumption of the grid operator. The revenue opportunities from the heat sales are at the same level as the grid operator’s natural gas self-supply costs [71].

Project WindGas Hamburg operation started 2015. The electrolysis system consisted of compact 1.5 MW_{el} PEM electrolyser. Produced hydrogen gas was fed into the natural gas grid. Technical experiences from the project were successful upscaling of the PEM technology to MW range in compact and efficient way. High outlet pressure of the electrolyser makes direct injection of the hydrogen to the natural gas network possible without external compression [72].

4.2 Hydrogen decarbonizing transport

The hydrogen from renewable sources serves as a clean energy source in mobility applications decarbonizing the transportation sector. In fuel cell electric vehicles (FCEV), hydrogen is transferred to electricity with fuel cell technology. Various transportation sectors can be implemented in hydrogen use: passenger cars, busses, aviation, maritime applications, trains and material handling vehicles (forklifts).

Fuel cells are used to make electricity by recombining hydrogen and oxygen as an inverse process to electrolysis. Fuel cell electric vehicles (FCEV) have good range of 400 - 560 kilometers per tank and quick refueling (3 - 5 minutes). Notable progress has been made by developing equipment standards (SAE J2061, SAE J2799) for high-pressure 700 bar and 350 bar refueling stations. Not like conventional vehicles, FCEVs are quiet and do not produce emissions except water vapor and heat. This is, if the hydrogen is produced from renewable sources. Otherwise, FCEVs have no emissions at the tailpipe [16].

Marcel Weeda et al. reports in the IEA Task 28 (2015) [28] that FCEVs, particularly passenger cars are ready for commercialization. The main barrier to large-scale introduction is market development. Significant challenges are related to additional hydrogen production and distribution infrastructure. The leading challenge is to create an acceptable business case for hydrogen refueling station (HRS) networks in their initial phase with possibly long period of underutilization. Reported H_2 fuel consumptions from different car manufacturers range between 0.76 - 1 kg/100 km [73]. To achieve similar fuel costs per kilometer for end-users as today with conventional vehicles, acceptable hydrogen fuel hydrogen price ranges 9 - 10 €/kg. This would mean 5 - 7 €/kg acceptable price for hydrogen delivered to station [9, 74].

Hydrogen has already shown promising results in public transport. Buses are well tested for hydrogen use in mobility and have reached a high technical maturity. Hydrogen buses offer almost same flexibility as conventional diesel buses in terms of range (300 - 450 km) even though it is still too short distance for coaches. Also newer hydrogen buses consume 8 to 9 kg H_2 per 100 km, which means 40 % efficiency advantage compared to diesel buses. The disadvantage for hydrogen buses is still higher production costs which are expected to decrease as production volumes rise [12]. Lorries and heavy-duty hydrogen trucks are not as mature technology as passenger cars and buses, but interest to bring zero-pollutant and low noise vehicles to the roads is growing [12].

For example, in Switzerland, 60 - 80 MW hydrogen supply framework for 1,000 heavy-duty hydrogen trucks and other applications is being built to operate during late 2019 [75].

Rail vehicles are currently typically either driven electrically or by diesel. Hydrogen-based trains, also called as hydrails, can be reasonable option to areas where electric infrastructure cannot be built due to technical, economical or other reasons [12]. First passenger train to utilize hydrogen FC as a power source started operating in Lower Saxony, Germany in September 2018. The Coradia iLint train has maximum speed of 140 km/h and range up to 1,000 km [76]. Hydrogen train is expected to replace diesel trains on the un-electrified rail sections in Germany [16].

FCEVs are already a viable solution in the forklift market. Today, forklifts are available with both internal combustion and electric motors. Fuel cell forklifts have shown advantages over electrical forklifts due to fast refueling and lack of need to change batteries. They offer uninterrupted use and are cheaper to maintain and repair [12].

4.3 Hydrogen decarbonizing industry

Today, most of the hydrogen is consumed in industrial applications. Large industries such as chemical plants, refineries and steel manufactures consume large, over 10,000 Nm^3/h quantities of hydrogen. Currently, this hydrogen is typically produced on-site by SMR. One-third is produced from by-product sources and less than 10 % is merchant hydrogen. Delivery is typically done via pipeline [9, 74]. Replacing SMR with water electrolysis technology can reduce significant amounts of CO_2 emissions in large scale industrial applications.

A steadily growing demand for hydrogen is estimated in refining industry. Hydrogen is used for hydro-treating, hydro-cracking and desulphurization in the refining process [9]. Currently, the largest planned PEM electrolyser plant with a 20 MW capacity will be built in Canada [57]. The facility using hydropower will produce almost 3,000 tons of H_2 annually to major industrial markets. Another recent project Refhyne was launched on January 2018 and scheduled to be in operation in 2020. The 10 MW PEM electrolysis plant will be built at the Shell Rheinland refinery in Germany. The PEM electrolyser will be able to produce about 1,300 tons of hydrogen annually. The hydrogen will be fed into refinery's hydrogen pipeline and utilized in the refinery processes like desulfurization of conventional fuels. Currently hydrogen is produced via two steam methane reformers. The PEM electrolyser will also be selling reserve services to the German Transmission System Operators. 10 MW Refhyne is designed as the building block for future up to 100 MW systems, which makes this project a great introduction to large scale electrolyser integrations into refineries [77].

Steel manufacturing industry is a very carbon intensive area due to use of coal and natural gas as energy sources. Blast furnace is the most common way to produce steel in Europe but integrating hydrogen injection into process is very challenging. However, other way called direct reduction iron (DRI) process has notably less technical issues for hydrogen injection. Two projects in Europe are experimenting the hydrogen implementing in DRI process. H2Future in Austria uses a 6 MW PEM water electrolysis demonstration plant to produce hydrogen for current use in steel manufacturing and provide grid balancing services for additional revenue [74]. HYBRIT in Sweden is in building phase. Aim of the project is to replace today's blast furnace process with DRI and to have completely fossil-free process for steel manufacturing by 2035. Consequently, coke is replaced with hydrogen gas from water electrolysis and water becomes the by-product instead of CO_2. H_2 consumption is around 650 Nm^3/t DRI for 100% H_2 operation [74]. Technology shift can cut Sweden's total CO_2 emissions by 10 % and Finland's by 7 % [78].

Chemical sector has largest percentage (65 %) of the hydrogen demand in industry. Key applications are ammonia, polymers and resins. Like in refineries, hydrogen is produced typically via SMR in chemical industry [21]. Ammonia synthesis, known as Haber-Bosch process, uses high pressure hydrogen and nitrogen in high temperature to produce ammonia. Ammonia production is a major consumer of energy and creator of GHG emissions largely due to SMR. Power-to-Hydrogen is a potential opportunity to reduce the GHG emissions [74].

4.4 Hydrogen for re-electrification

Re-electrification is a pathway, where renewable electricity is stored in hydrogen via water electrolysis during low electricity consumption. Chemical energy is transformed back to electricity via fuel cells or gas turbine during higher electricity demand. This promising technology has potential to store renewable energy in larger capacities (0.01-1000 MW) and for longer time periods (minutes-months) than for example in Li-Ion batteries [61]. Additionally, hydrogen storages do not self-discharge and higher temperature tolerance, which make them practical in extreme conditions. Potential environments for re-electrification applications are in isolated power systems and power backup systems. Stationary fuel cells also offer reduced noise and improved air quality by replacing diesel generators, which also require more maintenance. However, low roundtrip efficiency (around 29 % [9]) and high capital costs make this business case challenging [21].

Hydrogen re-electrification is demonstrated in project Haeolus which integrates 2.5 MW PEM electrolyser system with 100 kW fuel cell in a state-of-the-art wind farm in remote area in Norway. Object of this project is to demonstrate electricity storage, mini-grid and fuel production modes of operation. Production starts late summer 2019 [79].

5. PEM ELECTROLYSER CHARACTERIZATION

The main purpose of this chapter is to study system behavior and process limitations by doing experimental operations for small scale PEM electrolyser with a 30 kW power input and 4 Nm^3/h nominal H_2 output PEM water electrolysis system. Economic calculations are done on the basis of literature and experimental results to evaluate feasible economic environments for PEM electrolyser in Finland.

A Power-to-X demonstration for liquid and solid hydrocarbon production was done in Jokioinen at the St1 biorefinery plant site [80]. The key objective of the experiment was to utilize industrial CO_2 from bioethanol production and H_2 from PEM water electrolysis as a feedstock for hydrocarbon synthesis. CO_2 and H_2 was fed into a mobile synthesis unit and converted into liquid hydrocarbons and waxes. Two main reaction steps took place in the process: 1) reverse water-gas-shift (RWGS) and partial oxidation to transform CO_2 into carbon monoxide (CO) and to reform circulated light hydrocarbons, 2) Fischer-Tropsch synthesis to produce C1 - C40 n-paraffins from CO and H_2. The main task of the PEM electrolyser operation was to study H_2 production and consumption, system stability and behavior.

Following chapters first describe the working principle of the PEM electrolyser used in operations. Then the tests and operation activities to study system behavior and dynamics are presented. Finally, the chosen variables and boundaries for economic calculations are presented.

5.1 EL4N PEM electrolysis system

The PEM electrolyser's (EL4N) components can be divided into two groups. The first group is the stack and the second group includes other components integrated in the system, also called as balance of plant (BoP). Whole system is housed in a 10ft high cube container which is separated in two compartments. One side includes the hydrogen production system and the other includes the compression and storage equipment.

The main characteristics of the electrolyser are presented in Appendix 2. Figure 26 shows a simple block flow diagram of the electrolyser system which was used to produce hydrogen at the test site.

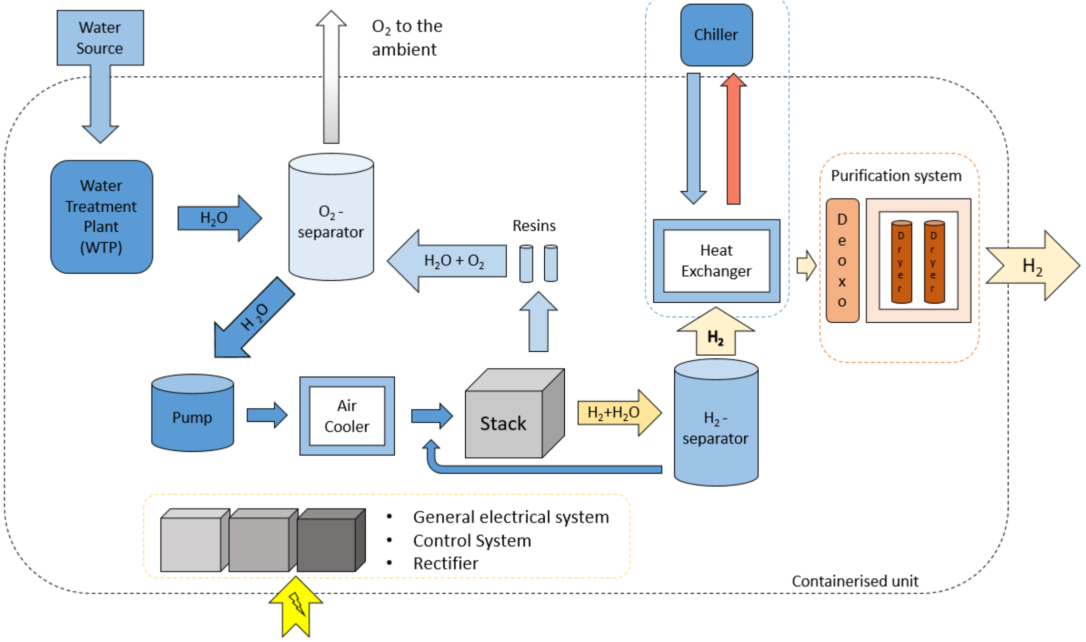

Figure 26. *Block flow diagram of hydrogen production system (adapted from manufacturer H2B2 manual.)*

First, water flows into the water treatment plant (WTP) which adapts the water quality to the stack requirements (< 0.1 µS/cm and Total Organic Carbon (TOC) < 30 ppb). The treatment consists of pre-treatment, reverse osmosis and electro deionization. Tank for the deionized water in the WTP has volume of 30 liters. The purpose of this tank is to store pure water. From the tank, water is pumped with distribution pump to the O_2-separator. In the O_2-separator the oxygen-water mixture dissociates and the oxygen is vented to the outdoors. Water flows to recirculation pump, which supplies water back to the stack through an air-cooler at the rate of around 1,300 l/h (= 0.36 l/s). Air cooler dissipates the heat produced in electrolysis process.

PEM stack uses electricity to split cooled (around 63 °C) liquid water into hydrogen and oxygen. At 100 % load actual power input for whole system ranged between 27 - 29 kW of which the stack consumed 22 - 23 kW. Stand-by power consumption was 1.5 kW. Electrolyser’s general electrical system include wiring, control system (PLC) and rectifier. In this setup, it is necessary to have a rectifier to adapt electrical current to the stack requirements (direct current, DC) as electricity from the grid (alternating current, AC) is used. System has also uninterruptible power supply (UPS) to ensure that electrolyser is able to switch to a safety position in case of power failure.

From the stack, the oxygen-water mixture continues to two ionic exchange resins which enhance the quality of water in the circulation. Then the mixture comes back to O_2-separator which also serves as a water reservoir for the electrolyser.

Hydrogen-water mixture continues to the H_2-separator. From there, hydrogen passes to the heat exchanger for cooling and separated water is recirculated back to the stack inlet. Hydrogen is cooled from 65 - 70 °C to 4 - 5 °C in the heat exchanger. The coolant fluid is water-glycol mixture which is cooled with chiller. In the purification stage hydrogen is purified to reach < 5 ppm water and < 5 ppm oxygen content. Equipment used to achieve this are: catalytic recombiner (DEOXO), which reduces the oxygen content, and two dryers which reduce the humidity content. Only one dryer is used at a time while the other is being regenerated. Dryer operation temperature is around 340 °C. Around 0.41 Nm^3/h of produced hydrogen is used at dryer regeneration. This means that at 100% load, the hydrogen production rate is 4.41 Nm^3/h of which 4 Nm^3/h is available for actual use.

5.1.1 Compression and storage equipment

Figure 27 illustrates a block flow diagram of the compression and storage equipment. First, produced and purified hydrogen enters at 19 - 20 bar a low pressure buffer. Buffer is required to stabilize the compressor operation. Pneumatic driven compressor pressurizes the hydrogen from 20 bar to 55 bar. Air demand for the compressor is about 49 Nm^3/h at 6 bar pressure. Heat exchanger cools the hydrogen after compression. Water-glycol mixture is used as a coolant and it is cooled with chiller. Finally hydrogen can be stored in high pressure storage with volume of 612 l.

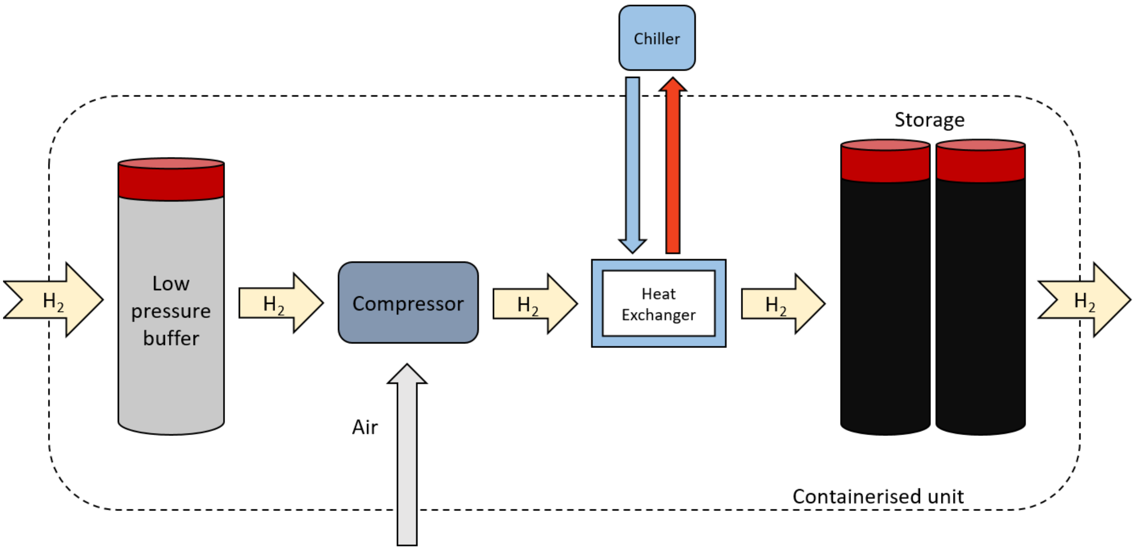

Figure 27. *Block flow diagram of hydrogen storage section (adapted from manufacturer H2B2 manual).*

If hydrogen production is shut down for a time and hydrogen storage is drained, decreasing the pressure in vessels, some arrangements are required when the system is restarted again. Due to low pressure in storage vessels, the pneumatic compressors cycle rate increases over optimal 54 bpm. Too high cycle rate could damage the compressor. Compressor’s control valve must be adjusted manually to keep cycle rate in 50 - 60 bpm range while storage vessels are filling back to around 50 bar pressure. Slightly high cycle rate is allowed during the filling, because as the storage pressure increases, the cycle rate will decrease.

5.1.2 Control and monitoring system

Control system enables operating and monitoring of the electrolyser. EL4N has fully automated and unattended Programmable Logic Controller (PLC) with 7” color touch screen. Electrolyser communication system uses Modbus TCP/IP protocol or Profinet (RJ45 port). Operation can be done remotely or locally. Remote control was enabled via computer or phone application.

In addition, a simple program using Modbus protocol for reading and saving the data from the electrolyser was created with LabVIEW engineering software. Reason for this additional program was to have more options and possibilities to gather the data. For example more variables were able to be monitored with wide adjustable data gathering frequency. Electrolyser’s own data saving system had five minutes time step, which was not practical tool for dynamic operation measurements. However, additional program needed development during the experiments and it was not fully functional during the first weeks.

5.1.3 Electrolyser operation modes

Two operational modes were available to choose from: manual set-point mode and automatic set-point mode. Manual set-point mode lets the user to set the hydrogen production load level between 10 - 100 % (90 - 900 A) of the nominal rate. When the maximum product pressure is achieved, the electrolyser will stop the production. During the hydrogen consumption, when the pressure decreases, the system will continue the operation at the set rate. Automatic set-point mode will run the system at 100 % rate. Hydrogen production is stopped when the buffer reaches 20 bar and as the pressure decreases to 15 bar, hydrogen production will start again.

System operation follows pattern presented in Figure 28. When electrolyser has been shut down, it must go through a cold start-up. The control system automatically does initial check to ensure that there are no alarms or failures active. Then system checks for control loops and levels and starts-up the water recirculation, cooling and ventilation system. Finally, control system starts-up the rectifier and production of hydrogen.

H_2 production stage operates from 0 to 20 bar. At 20 bar buffer pressure, the system switches to stand-by mode. When the buffer pressure reaches 15 bar, the electrolyser starts to produce hydrogen again.

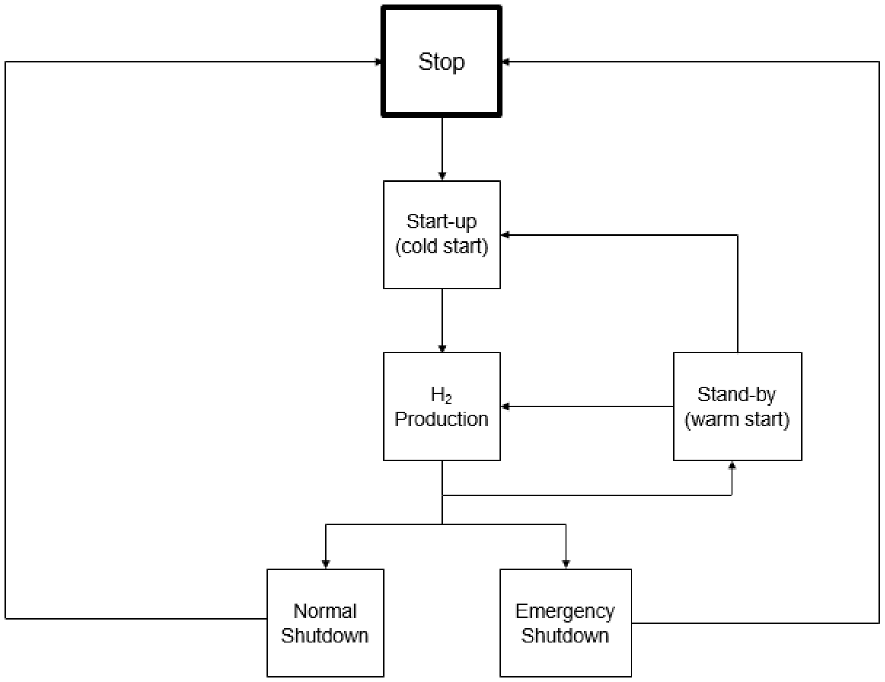

Figure 28. *Electrolyser's operation modes (adapted from manufacturer H2B2 manual).*

During the H_2 production and stand-by modes, the compressor pressurizes hydrogen from 12 bar to 55 bar. Two pilot switches control the pneumatic compressor. One them is installed in upstream and the other in the downstream. The low-pressure pilot switch cuts the air to the compressor when the hydrogen product pressure in buffer drops below 12 bar. This indicates that the electrolyser has been shut down. The high-pressure pilot switch cuts the air supply for compressor when hydrogen pressure reaches over 55 bar in the hydrogen storage. This indicates that the hydrogen consumption has stopped.

The normal shutdown is carried out automatically by the control system. System shuts down the power supply, the rectifier, the cooling and ventilation systems. Electrolyser is depressurized and recirculation pump is shut down. The pump will circulate water for minimum of two minutes before shutdown. The actual time depends on the stack voltage. Compressor is shut down independently through low and high-pressure switches.

The emergency shutdown (ESD) is caused by pressing the emergency button or by alarms show in Table 5. ESD carries out the following sequence: Power supply shuts down, system depressurizes and ventilation is kept running for two minutes.

Table 5. *Emergency shutdown alarms.*

Limit	Description
20 % LEL	Very high hydrogen concentration in the O_2 separator
20 % LEL	Very high hydrogen concentration in the process cabinet
True	Cabinet door opened during operation
50 % LEL	Very high hydrogen concentration in the compressor room
True	Compressor room’s door opened during operation

Electrolyser is recommended to use in 0 °C to 30 °C temperature range. Below zero temperatures may cause freezing in electrolyser’s parts containing water. High temperatures in turn may cause cooling problems. Electrolyser should also be operated in a ventilated area due to oxygen from anode increases the risk of explosion.

5.2 PEM electrolyser experiments

Pilot size PEM electrolyser experiments were done at the St1 biorefinery plant site in Jokioinen. First, electrolyser commissioning and training took place in mid-September 2018. Demonstration of the technologies for public was organized few weeks later in early October. After the demonstration event, experiments continued for about three weeks.

September − October testing period offered varying ambient conditions with -3 − 20 °C temperature range, rain and wind. Especially the low temperatures proved to limit the process. Practical aspects of the operation, such as remote control and system stability, were tested and electrolyser properties were measured during the experiments. Measurement data was gathered and saved by electrolyser’s own system. Later dedicated laptop, running the own data gathering program, allowing smaller time step for the measurements, was also connected to the system.

Electrolyser had 74 available variables in the Modbus table, which could be monitored by own data gathering program. Additionally, electrolyser's control interface, local and remote, presented data of electrolyser's efficiency, produced hydrogen and consumed energy. However, these variables were not saved for future analyzes. Major part of the available variables are illustrated in visualized PI-diagram of the electrolyser system (Appendix 3). In addition to variables in PI-diagram, the system power and individual cell voltages, were interesting variables to analyze. Some of the noteworthy variables did not have option to be measured (O_2-production rate, water inflow) and these values were calculated from the chemical equations or retrieved from the electrolyser's manual.

The electrolyser was used to feed hydrogen to the synthesis unit and operation was mainly done in automatic set-point mode. Electrolyser's ability to operate independently was tested by leaving hydrogen production on without on-site operators. However, process was monitored remotely via mobile application. Part of the electrolyser reliability for hydrogen feed is related to storage and compression stage. In case of technical error in hydrogen production, sufficient enough storage gives time for the repair work. Along with the storage adequacy, the system specific behavior was examined.

Interesting properties of the PEM electrolyser are particularly its ability to quickly ramp-up/down during the operation. The ramping times of the electrolyser stack and the system were measured in respect of input power. Durations of the cold start-ups were tested during the experiments several times and in different ambient temperatures. Cold-start tests required also using of the manual set-point mode. This data was important to know if PEM electrolyser could be used to provide grid services for additional revenue. Also high dynamic performance would be required to operate directly with renewable energy sources.

5.3 Economic calculations

Purpose of the economic calculation was to analyze which operation framework is suitable for PEM electrolyser in Finland. Hydrogen production cost is a useful key performance indicator (KPI) because it allows an economical comparison between other hydrogen production technologies. As the hydrogen cost depends on many factors, different scenarios were chosen to the calculations. The results first show the impact of the system efficiency and CAPEX on production cost value. Additionally, the impact of the utilization rate is presented using several different scenarios. Finally, yearly profits of the PEM electrolyser operation with different operation strategies and modified product selling prices are presented. Values for different scenarios are presented in Table 6.

Table 6. *Values for different scenarios in economical calculations.*

Scenario	Finland 2018	Denmark 2018	Speculative 2030
Electricity price [€/MWh]	FI 2018	DK1 2018	Modified FI 2018 150 % volatility 80 % price
Power input [MW]	10	10	10
Efficiency [%]	60 [30]	60 [30]	75 [36]
Specific investment [€/kW]	1,000 [30]	1,000 [30]	500 [30]
Fixed O&M costs [%/CAPEX]	3 [30]	3 [30]	3 [30]
WACC [%]	5	5	5
FCR-N income, hourly market [€/MW]	2018 hourly market [82]	2018 hourly market [82]	2018 hourly market [82]
Hydrogen selling price [€/kg]	2 - 3.4 [26, 55]	2 - 3.4 [26, 56]	2 - 3.4 [26, 55]
Oxygen selling price [€/t]	24.5 - 80 [39, 53]	24.5 - 80 [39, 53]	24.5 - 80 [39, 53]
Heat selling price [€/MWh]	25.3 [83]	25.3 [83]	25.3 [83]

Electricity price assumption has a significant role in the calculation as it takes the major part of the expenses. Because forecasting the future electricity price is practically impossible, three different price scenarios were used to calculate the hydrogen production cost: Finland 2018, Denmark 2018 (DK1) and speculative future electricity price scenario for 2030. A future scenario was created based on the factors known to have positive effects on economic viability of the PEM electrolyser operation. Speculative 2030 future scenario was created on the basis of Finland 2018 prices with lowered base price, added volatility (the variation in hourly prices), lower specific investment and higher system efficiency. Improved variables chosen to the future scenario are optimistic but still realistic to develop. Realized prices for FI 2018 and DK 2018 were gathered from Nordpool hourly history market data [81]. Transmission fees and taxes were assumed to be 15 €/MWh, as mentioned in theory part, and added to the total electricity price. Electricity price duration curves are illustrated in Figure 29.

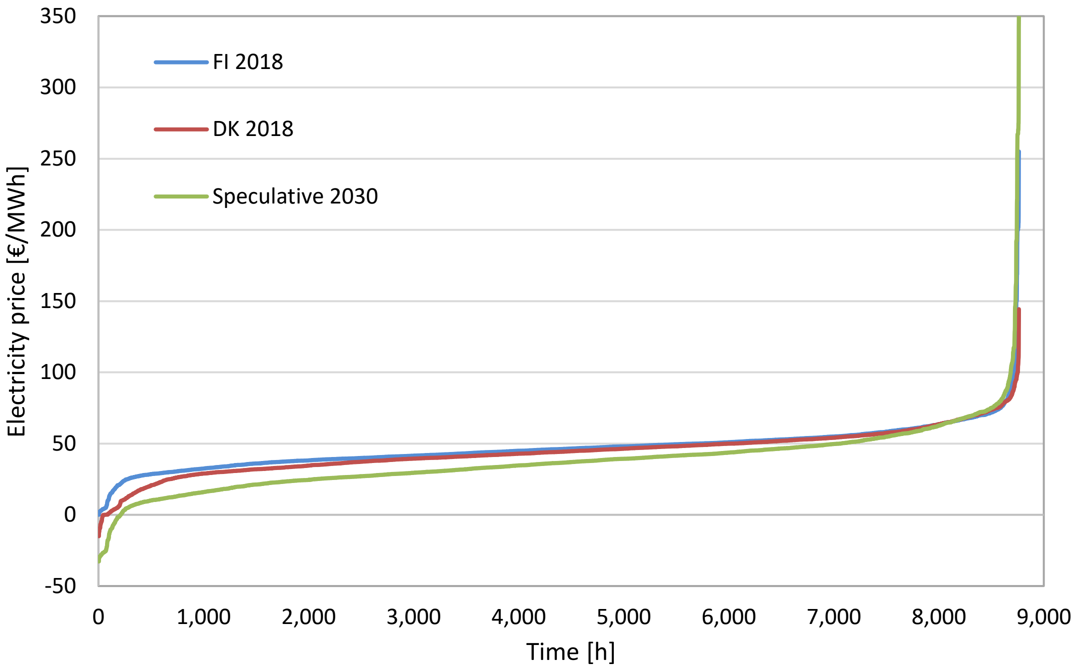

Figure 29. *FI 2018, DK 2018 and speculative 2030 electricity prices (without transmission fees and taxes).*

The prices follow similar pattern, but as expected, the speculative price scenario has highest volatility and cheapest average price. Some key numbers from these trends are also presented in Table 7. A noteworthy thing is that DK 2018 and Speculative 2030 have hours with negative electricity prices.

Table 7. *Key numbers from electricity price trends.*

Electricity price scenario	**FI 2018**	**DK 2018**	**Speculative 2030**
Average price 8760 h [€/MWh]	46.8	44.1	37.4
Average price 7800 h [€/MWh]	35.1	40.7	32.5
Average price 4400 h [€/MWh]	33.9	33.0	22.6
Min/Max price [€/MWh]	0/255.0	-15.0/144.3	-32.8/349.8
Negative electricity price hours [h]	0	50	207

Average prices for different annual operation hours are calculated from the cheapest hours of each scenario. These numbers provide direction how much annual electrolyser utilization rate effects the electricity costs. Minimum/Maximum price and negative electricity price hours support the need for grid balancing in DK 2018 and in Speculative 2030 scenarios.

Current CAPEX and OPEX values for 10 MW, presented in chapter 3.5.1, were used in the calculation. MS Excel's PMT function was used to calculate CAPEX payment. 5 % weighted average cost of capital (WACC) and 20 years economic lifetime were used as arguments in the PMT function. The present value for the investment was assumed to be CAPEX with added 15 % installation cost and 30 % investment subsidy. Also hydrogen storage cost estimate of 2 M€ was included. Fixed O&M costs are assumed to include all labor, maintenance and water costs.

As presented in chapter 3.5.4, additional revenue for PEM electrolyser operation is possible from grid balancing services. In Finland FCR-N market has most potential for extra income. Offered capacity needs to be symmetrical and 55 % set-point was chosen for calculation, as it offers 45 % up and down regulation with assumed 10 - 100 % electrolyser load range. Also constant 55 % load was used in FCR-N operation calculation, because historical data shows that up and down regulation is needed almost equally. For FCR-N hourly market, the scenarios with optimized operation hours are calculated using realized FCR-N hourly prices from 2018 [82]. The hourly market was used due to higher average revenue (22.8 €/MW) than yearly market (13.5 €/MW) in 2018. As FCR-N revenue is auction based with limited market size and the incomes may change yearly, the scenarios without FCR-N are also calculated.

Side products oxygen and heat are also possible revenue sources. Oxygen is assumed to be utilized in all calculations with 24.5 €/t price. Higher value for oxygen (80 €/t) is used in a calculation, which also assumes higher value for hydrogen. If heat can be utilized from the electrolysis operation, a higher system efficiency and additional incomes can be achieved. The revenue opportunities from the heat sales are assumed to be at the same level with the natural gas cost as was done in project Ibbenbüren (chapter 4.1.2). The used value for natural gas 25.3 €/MWh (LHV) is the average cost from 2018 in Finland and does not include taxes [83]. The amount of recoverable waste heat was estimated to be 50 % from the electric energy which was not converted into hydrogen.

6. RESULTS AND DISCUSSION

This chapter presents the results and conclusions from the test operations in Jokioinen, and economical calculations. First, measured and studied properties of the pilot scale PEM electrolyser are presented and the second research question “What are the process limitations for 30 kW small scale PEM electrolyser system operation in Finland?”, is answered. Next, results from the calculations for economic operation framework of PEM water electrolysis system in Finland is analyzed. This subchapter also gives answer to third research question “Which factors have the most significant impact on economic feasibility of PEM water electrolysis in Finnish conditions?“. Finally, last subchapter includes other noteworthy aspects of the PEM water electrolysis operation and general discussion about the thesis results.

6.1 Measured properties of the pilot scale PEM electrolysis

Altogether the electrolyser was running 212 hours including commissioning and repair work. After commissioning, the electrolyser did not necessarily require the attendance of operator on-site when operating in automatic mode. This allowed operating the electrolyser in more flexible way and an option for continuous 24 hours a day operation. However, different special occasions during the experiments, such as compressor cycle adjustment, required manual on-site work. Otherwise only monitoring for possible technical errors which could lead to automatic shut-down and consequently disturb the synthesis unit’s operation was needed.

Operating the electrolyser was environmentally friendly. Products released into ambient were oxygen, water and occasionally hydrogen. PEM electrolyser operation was silent and only significant noise came from the pneumatic compressor during the hydrogen compression into high pressure storage. PEM electrolyser is also very compact and has a little footprint, as the whole system was fit into a 10ft high cube container.

Pilot size PEM electrolyser’s mass balance is illustrated in Figure 30. Simplified model for mass balance includes water (untreated and treated), hydrogen and oxygen. Tap water with approximately 8.0 kg/h input rate, was purified in the water treatment plant. Required amount of purified water for the stack was around 4.0 kg/Nm^3_{H2} at 100 % load. Untreated water was purged out. Produced hydrogen rate was 0.4 kg/h of which 0.036 kg/h was used to regenerate dryers. Oxygen production rate of 3.1 kg/h was calculated from chemical reaction because no measurement data was available from oxygen output.

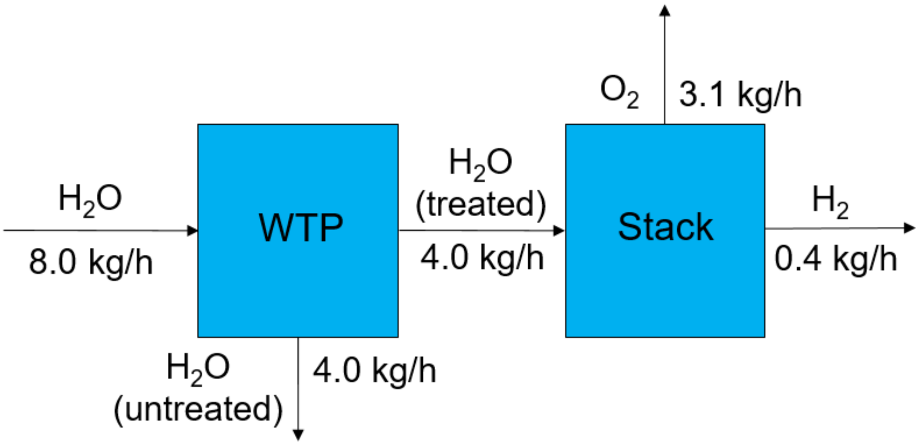

Figure 30. *Mass balance of the pilot PEM electrolysis at 100 % load.*

The energy balance for the pilot scale PEM electrolysis system is presented in Figure 31. The total input electricity is divided into BoP and stack blocks. 5.0 kWh of total 28 kWh input electricity is consumed by the auxiliary equipment and the rest 23.0 kWh is used by the stack. The system efficiency was around 47 % at 100 % load level.

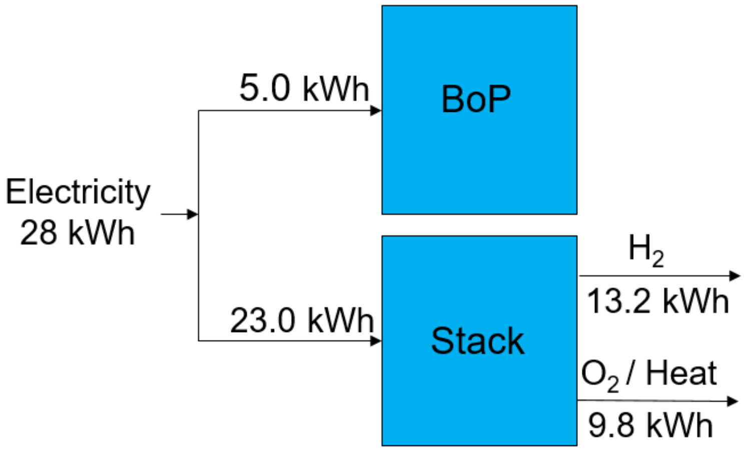

Figure 31. *Energy balance (LHV) at 100 % load for pilot PEM electrolysis system.*

With approximately 57.4 % stack efficiency 13.2 kWh of hydrogen (LHV) and 9.8 kWh of O_2/heat was produced from 23.0 kWh electricity input at 100 % load. Stack has a high 3 A/cm^2 current density, that increases the electricity consumption and consequently decreases the efficiency, but lowers capital investment and allows more compact system size. Heat utilization could increase the system efficiency but it would require specific system design and end-use application for produced heat at temperature around 66 - 68 °C.

Table 8 shows the estimated hydrogen production cost and produced amounts of H_2 and O_2 for the pilot scale PEM electrolyser operation for one year operation based on the values from test operations. The electricity costs used were realized and modified FI 2018 electricity prices. Modified electricity prices are same as used later in the economic calculations for Speculative 2030 scenario. Electricity prices also include network charges and taxes. Other estimated operational costs were assumed to be water and maintenance costs but as mentioned earlier, the electrolyser was able to operate independently and no labor costs are assumed in O&M. The value for maintenance work was based on the study Roadmap for Flanders [39]. Water costs are included into fixed O&M and not calculated separately proportional to operation hours due to small significance (1 - 2 % of electricity costs). Also, calculation does not include CAPEX in the H_2 production cost, which makes it significantly lower.

Table 8. *Estimated hydrogen production cost for pilot scale PEM electrolyser.*

Electricity price [€/MWh]	**Utilization rate [%]**	**O&M (excluding electricity) [€/kW/a]**	**H_2 production cost [€/kg]**	**Produced H_2 [kg/a]**	**Produced O_2 [kg/a]**
58.9 (FI 2018)	90	80	4.9	3,105	24,595
57.6 (FI 2018)	50	80	5.3	1,752	13,874
56.2 (Modified FI 2018)	90	80	4.7	3,105	24,595
46.35 (Modified FI 2018)	50	80	4.5	1,752	13,874

The H_2 production cost for pilot scale PEM electrolyser according to the calculation is 4.5 - 5.3 €/kg. The utilization rates of 90 % and 50 % convert to annual operation hours of 7,800 and 4,400 respectively. The electricity cost is lower with smaller utilization rate because the average electricity price is calculated from cheapest hours. However, due to fixed O&M costs the H_2 production cost is lower with higher utilization rate. The produced amounts of H_2 and O_2 show that even the hydrogen is the main product, oxygen is produced significantly more in terms of mass. It is also worth to note that this calculation did not take in account any revenues from the oxygen.

6.1.1 Buffer and high pressure storage behavior

Pilot electrolyser system was designed to be able to operate smoothly with the synthesis unit. Storage was required to ensure continues synthesis unit operation in case of technical problems in electrolyser system. Also, hydrogen buffer in this specific system was needed to stabilize compression stage, which resulted in dynamic power behavior.

Figure 32 shows the system and stack power behavior with varying product output flows. In addition, effect of the closed buffer tank on the electrolyser dynamics is shown in the beginning of the timeline.

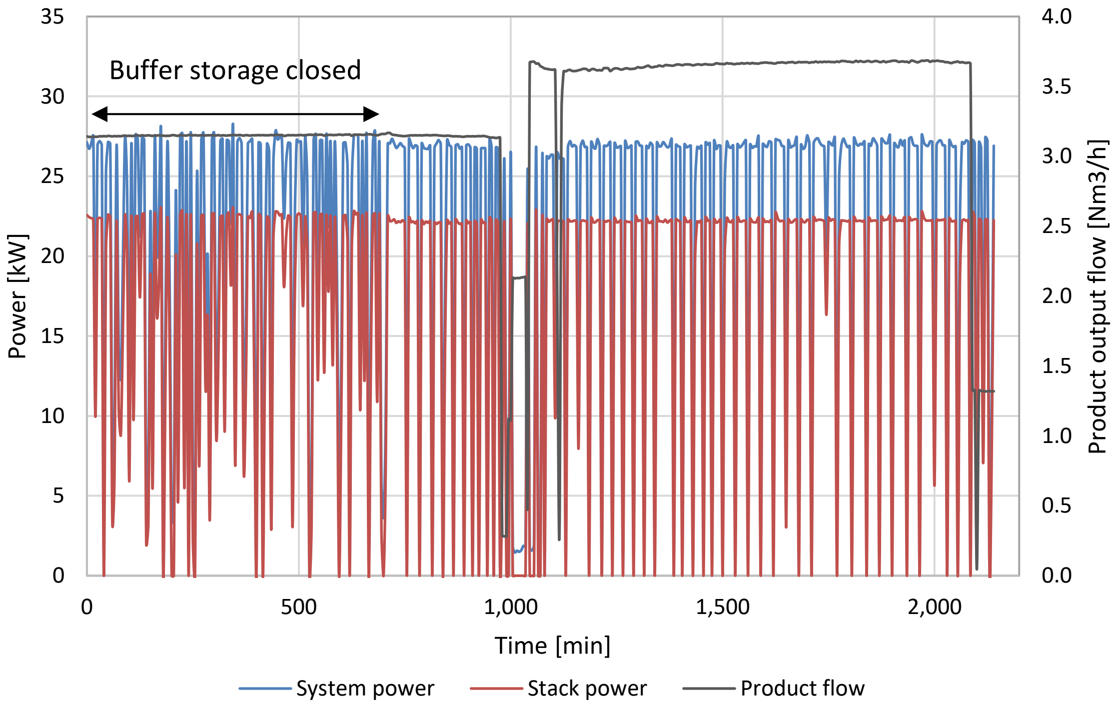

Figure 32. *System and stack power (kW) behavior with varying product flow output (Nm^3/h) into synthesis unit.*

Operation with closed buffer tank led to irregular system behavior. When the buffer tank was opened, power curves started to ramp-up and down more consistently. As can be seen in the Figure 33, the ramping cycle rate depends on product output flow. Ramping is more frequent with lower product output flows and with increasing output flow also stack can produce hydrogen longer without ramping down.

Altogether, this kind of dynamic power ramping during hydrogen production might be problematic for optimal, electricity price driven, operation and particularly providing grid balancing services. Without buffer mechanism stabilizing compression stage more constant power consumption could be achieved. However, during the storage filling, illustrated in Figure 33, the system and stack powers stayed almost constant.

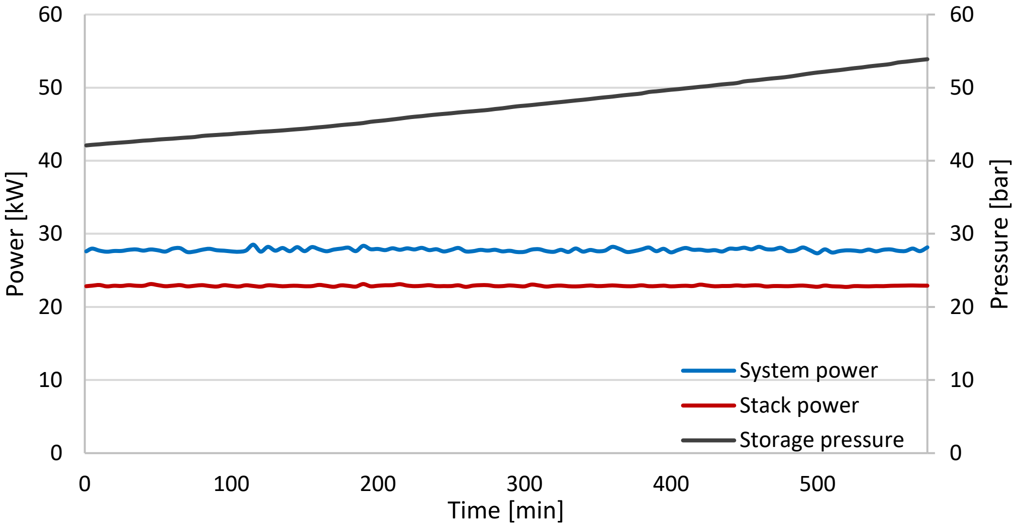

Figure 33. *System and stack powers during the high pressure storage filling.*

High pressure storage with volume of 0.612 m^3 could store approximately 2.6 kg of hydrogen at maximum pressure of 55 bar. Theoretically, at 100 % load with hydrogen production rate of 4 Nm^3/h, filling the storage takes around 7h 15 min. However, experimental data showed 7 bar/h filling rate at 100 % load, which converts to around 8 hours filling time. Storage was drained during the operation with synthesis unit at around 0.8 - 4.0 Nm^3/h hydrogen output rate. Figure 34 illustrates the storage pressure decrease rate at 0.8 Nm^3/h and 4.0 Nm^3/h output flows.

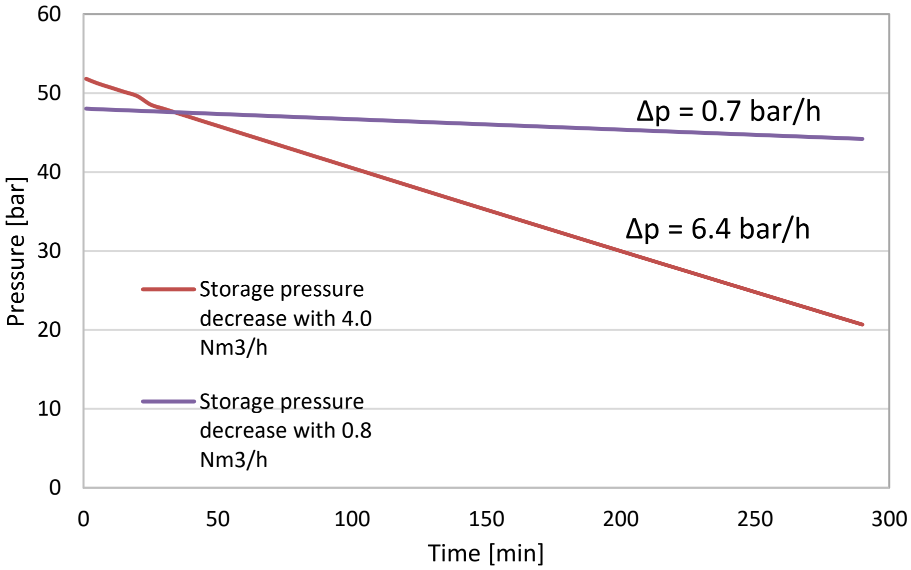

Figure 34. *Storage pressure decrease rate with 0.8* Nm^3/h *and 4.0* Nm^3/h *output flows.*

Pressure decrease rate was 6.4 bar/h and 0.7 bar/h for 4.0 and 0.8 Nm^3/h output flows respectively. It can be assumed, that storage pressure decrease with output flows between 0.8 - 4.0 Nm^3/h follow similar linear behavior between these lines. The time period that operator can feed hydrogen without running the electrolyser depends on pressure decrease rate. With longer time period the operator has better change to utilize cheap electricity price hours and avoid high cost hours. Storage adequacy depends, in addition to output flow, on hydrogen pressure requirement of the end-use application. With illustrated decrease rates, the storage is sufficient to keep hydrogen flow to end-use application for 5.5 - 50 hours with assumed minimum pressure requirement of 20 bar (55 - 20 bar). With pressure range of 55 - 0 bar, storage would be enough for around 8.5 - 78.5 hours. However, this is impractical range because typically end-use applications will require pressurized hydrogen.

6.1.2 Ramp-up/down, warm start-up and cold start-up times

Results from warm start-up and ramp-ups/downs within 10 % - 100% load range are presented in Table 9. Warm start-up and ramp-ups within operation load range times were tested for stack power and system power.

Table 9. *PEM electrolyser properties on ramp-up and ramp-down.*

Variable	Ramp-up	Time
Stack power	Stand-by → 100 %	81 s
System power	Stand-by → 100 %	90 s
Stack power	Within load range	< 1 s
System power	Within load range	< 4 s
Variable	**Ramp-down**	**Time**
Stack power	100 % → Stand-by	< 1 s
System power	100 % → Stand-by	< 2 - 3 s (2.5 - 3.5 kW) 8 - 14 min (1.5 kW)
Stack power	Within load range	< 1 s
System power	Within load range	< 3 s

Results from the test operations are in line with the ramp-up and ramp-down rates for PEM electrolysis presented in chapter 3.5.6. Compared to the alkaline electrolysis ramping rates, the test results show much better performance both in ramp-up and ramp-down situations.

Figure 35 illustrates the ramp-up from stand-by to 100 % load. Warm start-up, meaning a ramp-up from stand-by mode to 100 % load, took 81 seconds for stack and around 90 seconds for the system power. After the initial quick increase, the power growth showed linear behavior.

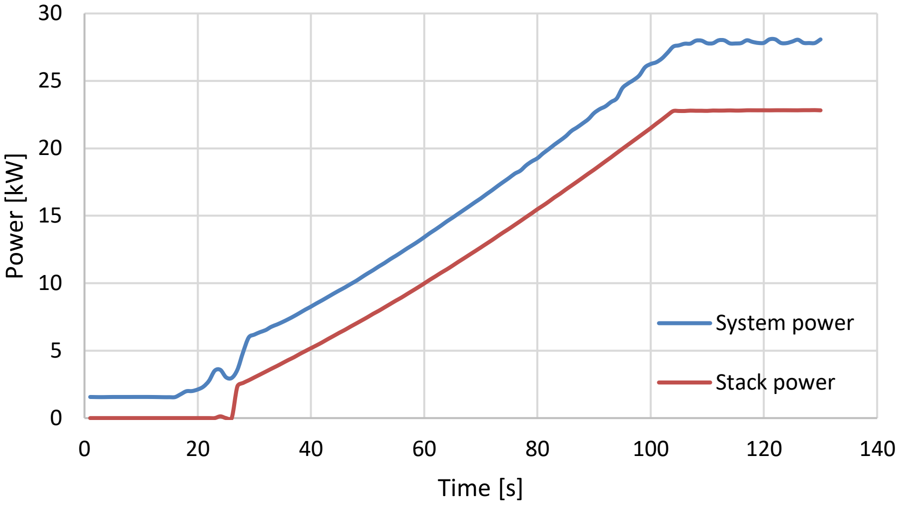

Figure 35. System and stack power as a function of time in warm start-up.

According to the data from the ramp-downs, the stack power dropped from around 22.8 kW to 0 kW in less than one second. However, the system power showed less immediate behavior. System power dropped quickly, in 2-3 seconds, to 2.5 kW - 3.5 kW power range and then slowly to the 1.5 kW as is illustrated in Figure 36. This slow drop is caused by auxiliary equipment, which kept running after the stack shut down. For example water flow to stack inlet is kept running at production level after ramp-down for five minutes. Also cooling system is kept on after shut-down for sufficient time to keep temperatures on acceptable levels.

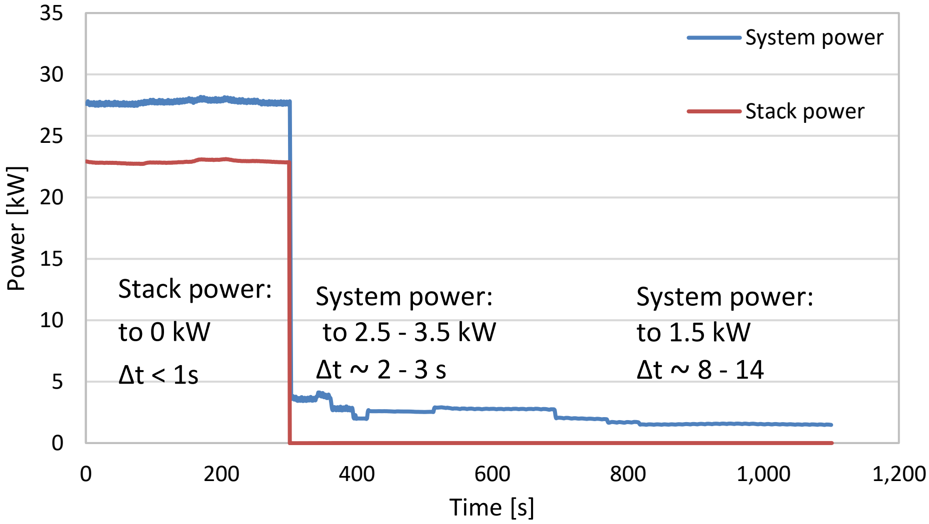

Figure 36. Typical system and stack power dynamics on electrolyser ramp-down from 100 % load to stand-by.

Figure 37 illustrates a ramp-down from approximately 85 % load to 10 % minimum load. The ramp-down speed was less than one second for the stack power and under 3 seconds for the system power. However, as can be expected, there was no notable slow decrease in power input after the initial drop. Then, power is ramped up to 40 % load and after a while, to 60 % load. Stack power ramped-up in under one second and system power in under four seconds.

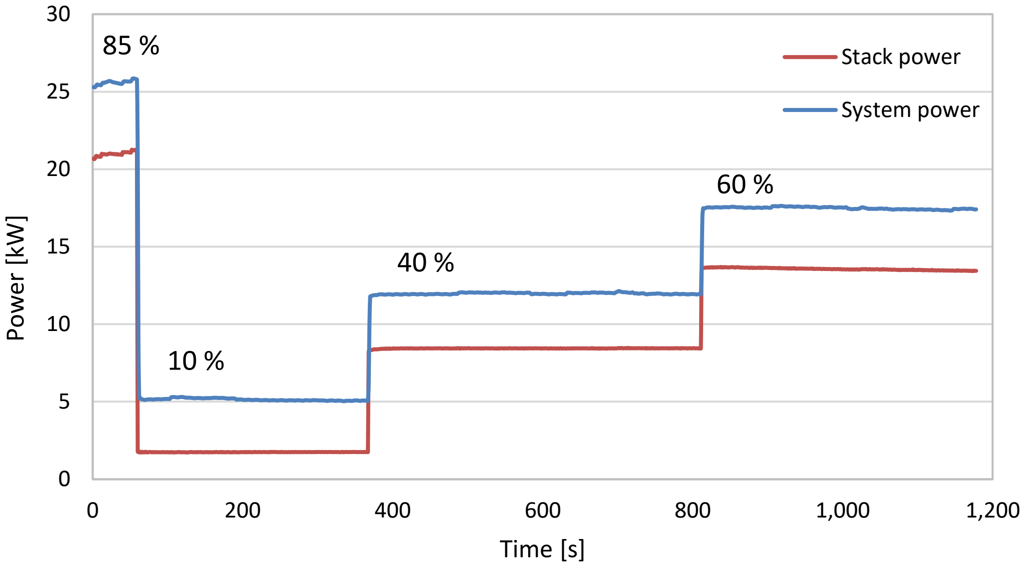

Figure 37. *Ramp-down and ramping up dynamics within load range for system and stack power.*

PEM electrolyser stack was very fast to ramp down and up in certain cases. To get more precise results, a shorter time interval for measurement would have been needed. However, the knowledge of ramping speed less than one second is enough for practical operation.

Cell voltages reacted slowly to the ramp-down. Voltages stayed approximately constant after ramp-down for 174 seconds and then it took around 94 seconds for all cell voltages to drop near zero as the power supply controlling stack voltage set voltage to 0 V. As Figure 38 illustrates, cell voltages increased individually after the initial drop. This is due to recombination of H_2 and O_2 remained in the stack acting as a fuel cell. System has a drain resistor which acts when system reaches the state of a fuel cell. It took approximately 67 min for H_2 and O_2 to be consumed and all cell voltages to level to zero.

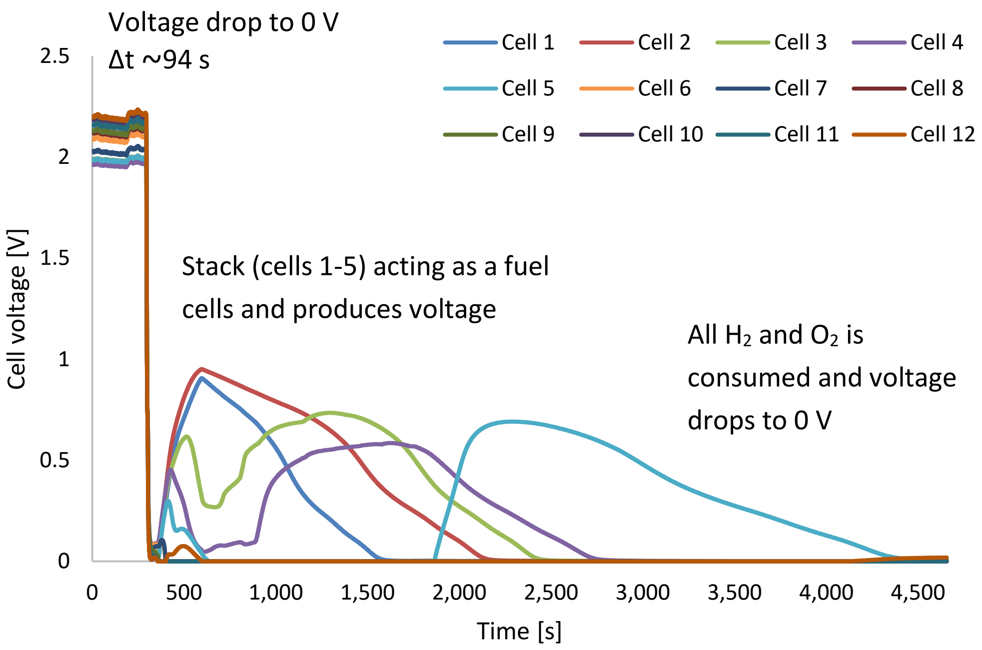

Figure 38. *Cell voltages behavior on ramp-down from 100 % load to stand-by mode.*

However, cell voltage behavior changed when ramp-down was executed in operation range 100 % - 10 % (Figure 39). The delay between power input change and cell voltage change stayed same 173 seconds, but unlike in ramp-down to stand-by mode, the ramp-down and up times were < 1 s. Voltages also stayed constant at new load level as the stack will not act as a fuel cell when power is applied.

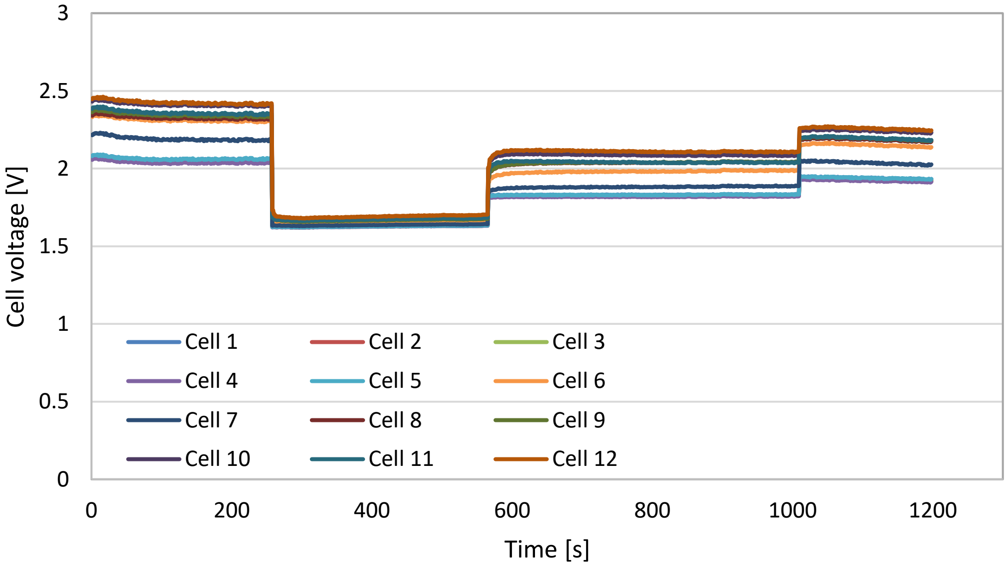

Figure 39. *Cell voltages dynamics on ramp-down and ramp-up.*

A notable voltage difference between cells occurred at different operation loads. The difference seemed to be larger at higher operation load. This voltage difference was due to higher resistance in upper cells caused by heat. Ohmic losses increase with increased power.

A notable property of the electrolysis system was cold start-up time which varied in range of 15 - 30 min depending on the ambient temperatures and duration of the shut-down. The start-up times presented in chapter 3.5.6 for alkaline and PEM electrolysis also have quite significant variance. Start-up times for pilot size PEM electrolyser in test operations were moderately longer than the literature estimates, which can result from low ambient temperatures in the test site. However, alkaline electrolysis start-up times are reported to be up to several hours, which are significantly longer than test operation results for pilot PEM system. Near zero temperatures affected the cold start-up time by decreasing the circulating water temperature. This increased the stack's resistance which in turn led to increase of too high (> 2.5 V) cell voltages (Figure 40).

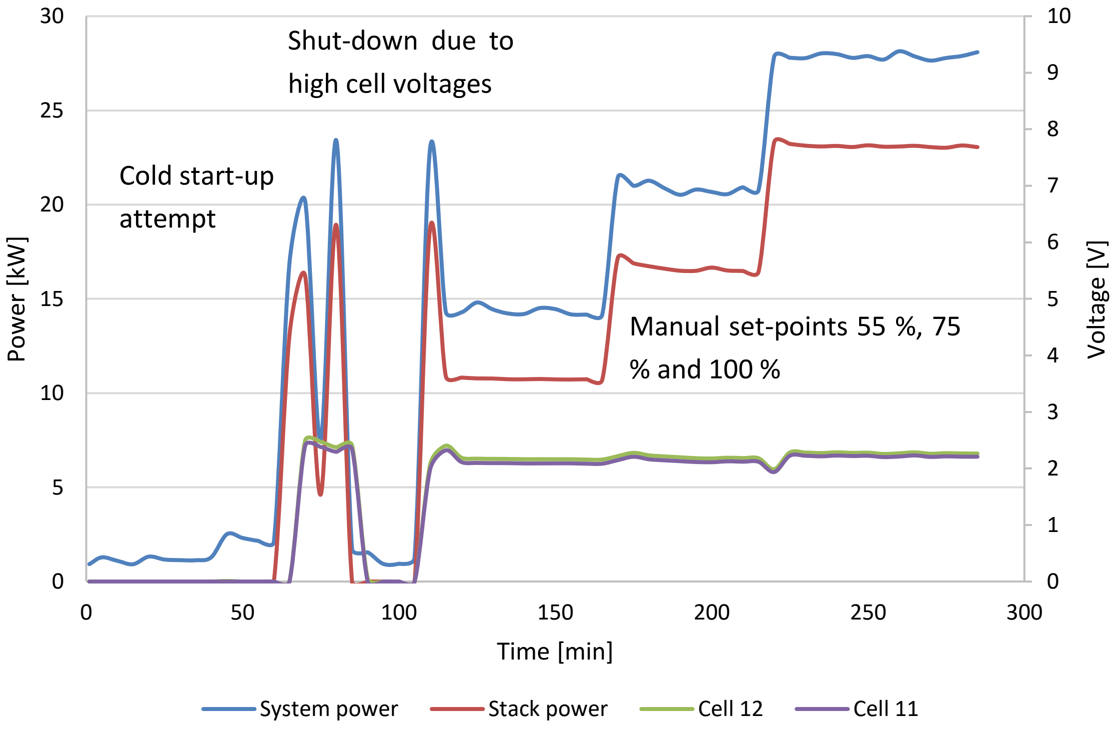

Figure 40. *System shut-down after cold start-up in cold ambient conditions following with manual start-up.*

As a result, the electrolyser's safety mechanism turned off the system while it was trying to increase the stack power. To avoid the shut-down, manual set-point was used to start the system. First, load set-point was set to 55 %, then 75% and finally to automatic mode, which increased the load to 100 %.

6.2 Economic calculations

Economic calculations aim to find profitable PEM electrolyser operation scenarios using current and speculative but realistic to develop future variables. Electrolyser system efficiency has a very direct effect on hydrogen production cost. Figure 41 shows hydrogen production cost as a function of LHV efficiency and specific investment with high 7,800 h (90 %) annual utilization rate. Using average FI 2018 electricity price, production cost varied between 6.7 - 2.1 €/kg with selected 0.3 - 0.95 efficiency range. Specific investment in range of 1,000 - 250 €/kW decreased the hydrogen production cost on from 3.6 to 3.3 €/kg.

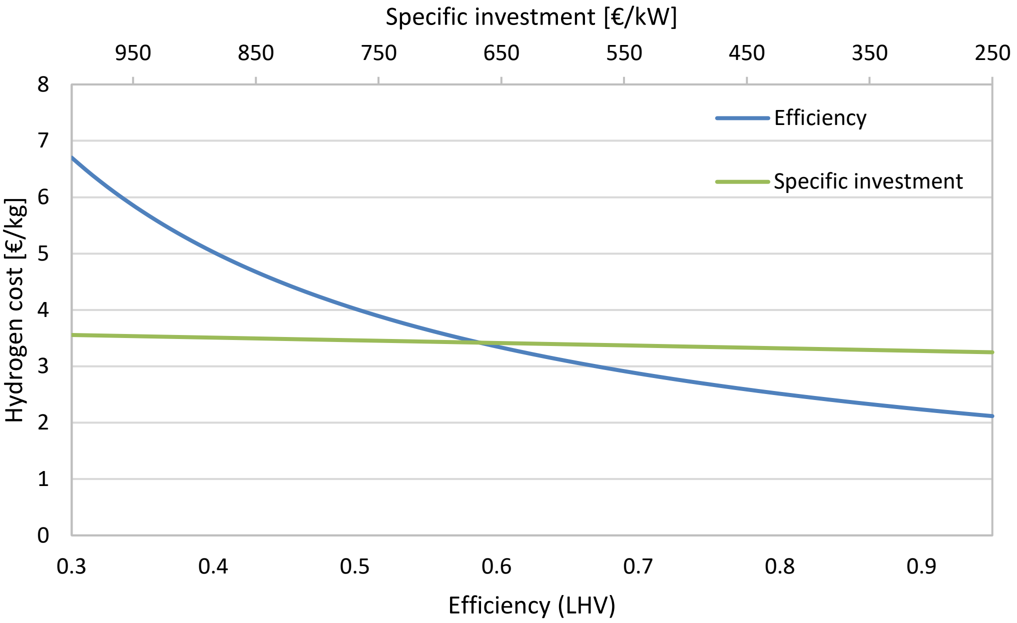

Figure 41. *Hydrogen cost as a function of efficiency and specific investment with 7800 annual operation hours using average FI 2018 electricity price.*

Very high system efficiencies can be achieved by utilizing the waste heat, which cannot be calculated with same value as hydrogen. Therefore the effect of increased efficiency on hydrogen cost is less significant after around 0.75 efficiency (LHV), which was estimated to be highest system efficiency for 2030 in terms of kWh/kg_{H2} (chapter 3.4). Reduction in CAPEX shows little effect in hydrogen production cost with these high operation hours. On the other hand, with low operation hours, the effect would be significant as mentioned in chapter 3.5.2.

Figure 42 illustrates the hydrogen cost as a function of annual operation hours for selected economic scenarios. Cost curves for different scenarios are calculated using average price from realized electricity prices and with added yearly CAPEX, OPEX and oxygen revenue (24.5 €/t). The current target cost range for hydrogen (2 €/kg) is based on current SMR production cost. However, this cost may increase in the future due to increase of CO_2 emission allowance costs and natural gas cost. As mentioned in chapter

3.5.6, these increases can result to SMR hydrogen production cost of 3.4 €/kg, which is presented as a “Future cost target”.

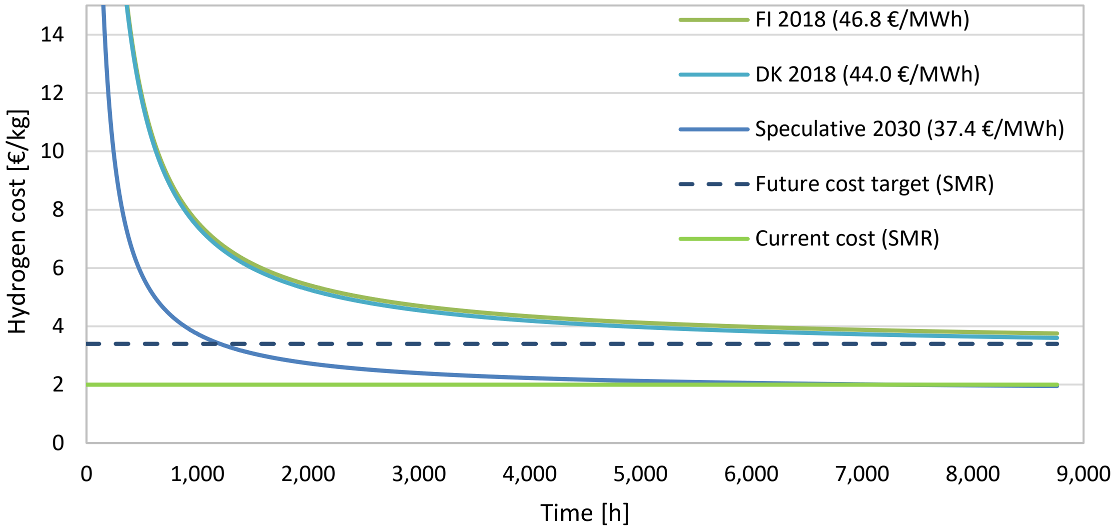

Figure 42. *Hydrogen cost for different scenarios as a function of annual operation hours.*

With average FI 2018 (46.8 €/MWh) and DK 2018 (44.0 €/MWh) electricity prices hydrogen production cost will not reach current or future cost target without additional revenues. Speculative 2030 (37.4 €/MWh) scenario reaches the target cost at around 2,000 h and settles at around 2.0 €/kg production cost, which would make it barely profitable scenario.

Naturally, it is reasonable to operate the electrolyser only during hours when the revenues are higher than costs. Figure 43 presents the distribution of yearly costs and incomes for different scenarios with optimized operation hours. Optimizing makes operating the electrolyser profitable in terms of operational costs (mainly electricity). However, annual CAPEX and fixed O&M costs must be covered and this kind of optimization does not necessarily lead to positive profit if the electrolyser utilization rate is low. Additionally, profitability with and without CAPEX is illustrated in the calculations. Even without CAPEX, operation may be unfeasible as revenues will not cover fixed O&M costs, including for example labor, and electricity.

Scenarios with FCR-N operation were calculated at full load, 55 % load (FRC-N) or not at all, depending on which setup is the most profitable. Result data including profit (k€/a), levelised cost of hydrogen (LCOH, €/kg), utilization rate (%) and the amount of produced hydrogen (kt/a) is presented in Table 10.

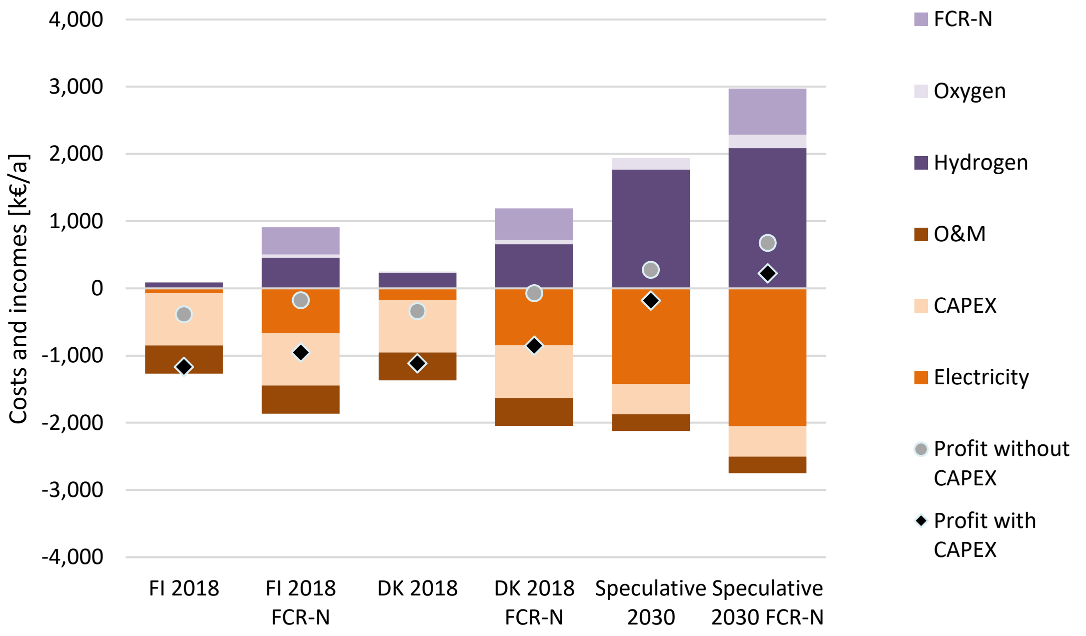

Figure 43. *Yearly costs and revenues of 10 MW PEM electrolyser operation with optimized operation hours.*

With optimized operation hours only Speculative 2030 FCR-N scenario becomes economically viable with CAPEX making 222 k€/a profit. This can be explained by the very low utilization rates, especially for FI/DK 2018 and FI/DK 2018 FCR-N price scenarios. In Speculative 2030 and Speculative 2030 FCR-N scenarios more practical utilization rates of 45 % and 53 % respectively are achieved. Impact of the low annual operation hours can been seen in cost side, where CAPEX and O&M are relatively high compared to the electricity. Also very little hydrogen is produced and large share of revenues come from FCR-N services. Such situation is not ideal because of uncertain nature and limited size of the FCR-N auction market.

Table 10. *Results for yearly 10 MW PEM electrolyser operation for different scenarios with optimized operation hours.*

	FI 2018	**FI 2018 FCR-N**	**DK 2018**	**DK 2018 FCR-N**	**Speculative 2030**	**Speculative 2030 FCR-N**
Profit without CAPEX [k€/a]	-389	-176	-338	-75	273	678
Profit with CAPEX [k€/a]	-1,168	-955	-1,117	-854	-182	222
LCOH [€/kg]	27.7	6.2	11.7	4.6	2.2	1.8
Utilization rate [%]	3	15	7	21	45	53
H_2 produced [kt/a]	0.1	0.2	0.1	0.3	0.9	1.0

Operation with heat utilization is not currently very common for PEM electrolysers but has been demonstrated and some future plant designs include heat recovering in the system. Next calculation (Figure 44) shows the impact of the heat utilization in the yearly incomes.

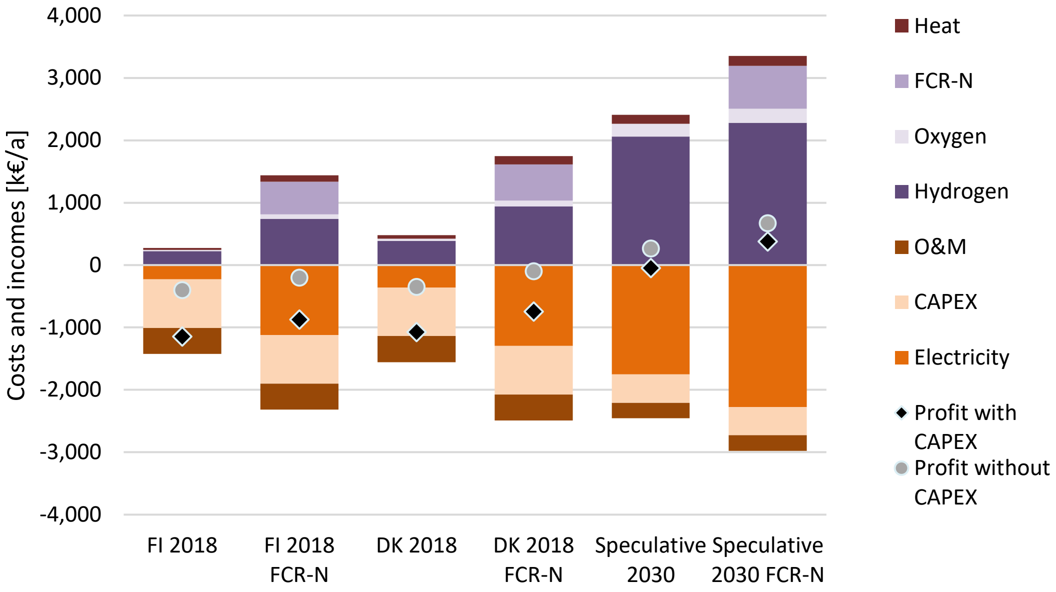

Figure 44. *Yearly costs and revenues of 10 MW PEM electrolyser operation with optimized operation hours and waste heat utilization.*

As expected, the heat utilization improves the profitability of the all scenarios. However, the heat utilization is assumed to only bring revenues and the possibly higher investment costs have not been taken into account. Still only the Speculative 2030 FCR-N scenario makes positive profit with CAPEX. With current FI 2018 and DK1 2018 prices the operation is rarely feasible and utilization rates stay low between 7 - 8 % without FCR-N incomes. Heat utilization is a good way to improve electrolyser efficiency but represents small share of the revenues. In FI 2018 and DK 2018 scenarios the heat revenues are around 14 % of the hydrogen revenues. In the Speculative scenarios with higher system efficiency the heat utilization revenues have smaller 7 % part of hydrogen incomes. The result data is presented in Table 11.

Table 11. *Results for yearly 10 MW PEM electrolyser operation with optimized operation hours and waste heat utilization.*

	FI 2018	FI 2018 FCR-N	DK 2018	DK 2018 FCR-N	Speculative 2030	Speculative 2030 FCR-N
Profit without CAPEX [k€/a]	-401	-202	-351	-100	263	671
Profit with CAPEX [k€/a]	-1,148	-856	-1,075	-745	-48	376
LCOH [€/kg]	12.3	4.4	7.5	3.6	2.0	1.7
Utilization rate [%]	7	24	8	30	52	58
H_2 produced [kt/a]	0.1	0.4	0.2	0.5	1.0	1.1

Speculative future scenarios have shown best potential for viable business cases. Clearly, lower electricity prices and higher volatility result effectively to better profitability. However, electricity prices are difficult to predict and might act in unfavorable way. Other method to improve the electrolysis profitability is to increase the product values. It is realistic to assume that values could increase due to increasing demand of hydrogen and oxygen. In addition higher costs for dominant hydrogen production via SMR, could increase the selling price of hydrogen. Oxygen selling does not impact the viability as much as hydrogen, but more optimistic oxygen value can also be implemented due to wide range in estimated selling prices. Figure 45 shows the calculations for scenarios with increased hydrogen price (3.4 €/kg), which is the possible future hydrogen production cost for SMR and oxygen price (80 €/t), which is higher value for oxygen estimated in literature (chapter 3.5.4).

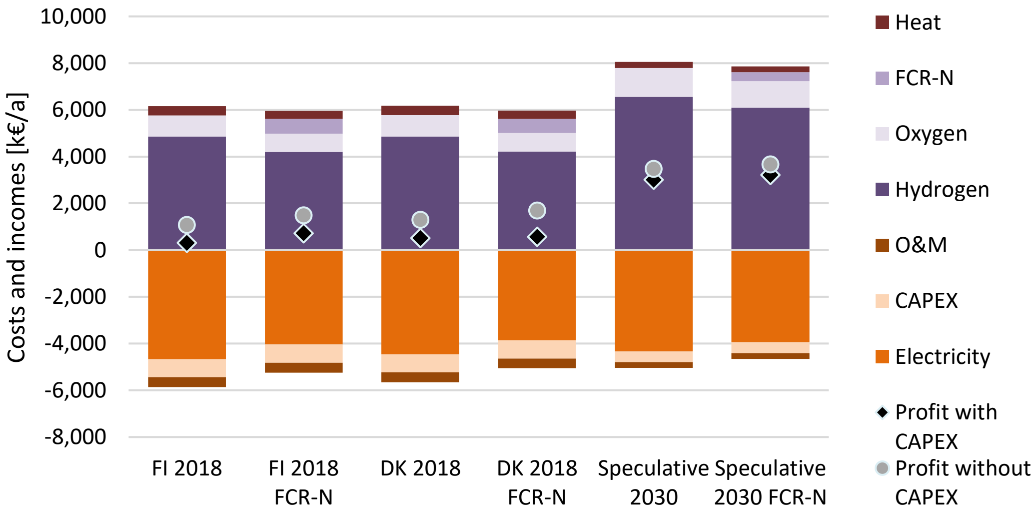

Figure 45. *Yearly costs and incomes of 10 MW PEM electrolyser with optimized operation hours, heat utilization and higher product values.*

All operation scenarios made positive profit and even covered CAPEX. In all scenarios main revenue came from hydrogen sells, which is more stable revenue source than FCR-N services. Oxygen revenues were approximately 20 % of hydrogen revenues with chosen values. Due to higher hydrogen value, the share of heat utilization has decreased to 8 % and 4 % of hydrogen revenues for FI/DK 2018 and Speculative 2030 scenarios, respectively. The electricity purchase price covers major share of the costs. CAPEX has the second largest share. However, PEM electrolysis investment costs are relatively high and factors, such as timeframe, in calculations determine how heavy impact they have on annual profits. The result data is presented in Table 12.

Table 12. *Results for yearly 10 MW PEM electrolyser operation with optimized operation hours, heat utilization and higher product (H_2 and O_2) values.*

	FI 2018	**FI 2018 FCR-N**	**DK 2018**	**DK 2018 FCR-N**	**Speculative 2030**	**Speculative 2030 FCR-N**
Profit without CAPEX [k€/a]	1,081	1,491	1,296	1,688	3,469	3,670
Profit with CAPEX [k€/a]	302	712	517	562	3,013	3,214
LCOH [€/kg]	3.2	2.8	3.0	2.6	1.8	1.6
Utilization rate [%]	91	78	91	79	98	91
H_2 produced [kt/a]	1.4	1.2	1.4	1.2	1.9	1.8

The high utilization rates of 78 - 98 % in the scenarios convert to around 6,800 - 8,600 annual operation hours. The highest utilization rate of 8,600 annual operation hours may be even practically difficult to achieve due to maintenance work.

According to economical calculations, the PEM electrolyser operation was not feasible at current FI/DK 2018 electricity prices, H_2 cost target and chosen variables for other factors. Speculative 2030 scenarios showed best feasibility with chosen optimistic values for CAPEX, electricity price and efficiency. Major share of the revenues come from the hydrogen and consequently with increased H_2 cost target (3.4 €/kg) all scenarios became feasible. Additional revenues were small compared to the hydrogen sells but together added up to be very relevant income.

6.3 Overall discussion

PEM electrolyser showed good dynamic properties with very quick ramping times and a shorter time interval for measurement would have been needed to get more accurate results. However, ramping speed less than one second is enough for practical operation. BoP was slightly slower to react for ramp-downs but as the major part of the electricity consumption consists of the stack, slower decrease time affects only small part of the consumption. Providing grid balancing services requires under three minutes full activation time which is achieved in the warm start-up but not from the cold start-up. Therefore, PEM electrolyser should be always operating at least at low load level due to long and uncertain cold start up times if operation takes place in cold ambient conditions. Issues regarding the ambient temperatures appeared at cold start which required, a manual, slow ramping to heat the system before full load operation. Also, operation at subzero temperatures would require special design of the system to avoid water freezing.

Hydrogen production cost is sensitive to many factors. The most significant parameter impacting the hydrogen cost in PEM electrolysis is electricity costs. Other significant costs come from CAPEX and O&M costs, which have smaller impact as utilization rate increases. Chosen timeframe for the investment was 20 years, which is equivalent to technical lifetime of the plants. With shorter timeframe CAPEX would be larger, resulting to higher hydrogen production cost. The chosen scale of the electrolyser plant for this economic calculations (10 MW) was relatively large, but corresponding to state-of-the-art PEM electrolysis plants. Multi MW-size plants are typically required in the industrial applications and as discussed later, the possible near term hydrogen use in Finland will be mostly in industrial sector. Also, according to the estimates of capital cost development in chapter 3.5.1, the economy-of-scale effect becomes less significant after reaching 10 MW size.

It is worth to note, that the calculation model considers only if particular hour is profitable or not and no limits are set to how long the electrolyser can be without producing hydrogen. Practically this kind of optimizing is possible only if the hydrogen storage is adequate to feed the end-use applications when electrolyser is not running. Optimization of the electrolyser and storage buffer sizes are crucial for uninterrupted and profitable operation.

Also, as mentioned in chapter 3.4, the best system efficiency is not typically achieved at 100 % load but at around 40 % load level, which could play a role in electrolyser plant procurement decisions. Calculation also assumes no electricity consumption at hours, when the operation is not profitable, because no hydrogen is produced. However, the auxiliary equipment have small power consumption at stand-by mode (5 % from nominal power input in the pilot scale electrolyser). Also experimental operation pointed out, that it is practical to run the electrolyser at least at minimum load due to uncertain cold start-up times. This is because of retaining the fast ramping abilities, which is particularly important when providing FCR-N services. Against this background, it is not viable to operate at all if the utilization rate would be very low.

Revenues from the electrolyser operation comes mainly from the hydrogen sells. Possible additional incomes come from FCR-N services, oxygen sells and possible heat utilization. FCR-N operation made all scenarios more feasible with 2018 hourly prices. It is noteworthy the mention, that the model assumed FCR-N operation to take place only on hourly market. Practically operator can provide capacity for both yearly and hourly markets as described in chapter 3.5.4. This could be even more profitable method, but has not been considered in this calculation. However, frequency control reserve service revenues are auction based and future auctions are not granted to offer same revenue for balancing services. Because of this, planned electrolyser operation should be feasible with hydrogen and oxygen selling revenues instead of relying on additional grid services.

Different end-use applications define the side revenue possibilities and requirements for H_2 and O_2 production rates. They determine the optimal size for the electrolyser system. Optimizing the hydrogen generation and storage capacity for flexible and optimal operation is crucial. Larger systems require higher investments costs but because of larger production capacity, hydrogen production cost is lower. Higher storage capacity also offers better flexibility and stability. End-use application also determines the required hydrogen pressure, which could in some cases allow leaving the compression stage out as PEM electrolyser can operate at high pressures (chapter 3.5.3 and project WindGas Hamburg).

For all scenarios, high utilization rates were achieved in the calculation with higher product prices. However, despite of favorable impact of high utilization rate on profitability, best scenarios with 8,600 annual operation hours may be even unpractical due to needed maintenance and repair work. On the other hand, multi MW-scale PEM electrolysers are typically composed of individual MW-size stacks. This could enable maintenance for the part of the stack while others are still running and only some of the capacity is lost during the repair work. This would be essential for industrial applications where constant hydrogen production is typically required. Low annual operation hours proved to be unfeasible due to relatively high CAPEX and fixed O&M costs which need to be covered.

Hydrogen end-use applications in Finland are currently in industrial applications, which would require multi MW-scale PEM electrolysis plants. Major share of hydrogen is consumed by oil refining sector. Smaller shares go to chemical industry, mining and heat & electricity. In addition to industrial applications, options for hydrogen use mentioned in chapter 4 were gas grid injection, methanation and use in transportation sector as a fuel. Injecting hydrogen into natural gas grid in Finland is limited to 1 % but hydrogen methanation could raise injection limit significantly. Synthetic methane could also be used as a fuel supply for gas cars, which would create more self-sufficient fuel supply.

Hydrogen could be directly used as a fuel for FC vehicles but this would require major improvements on HRS infrastructure and significant increase in FC vehicles in Finland. Even though there are plans for around 20 HRS installations by 2030, the hydrogen use in transportation is in very early stage in Finland.

7. CONCLUSIONS

PEM water electrolysis and Power-to-X systems are seen as potential technologies to achieve GHG emission reductions and limit the global warming to 1.5 °C above pre-industrial levels. Moreover, Power-to-X systems are also seen as a solution to balance electricity grid in the case of higher share renewable energy production which would increase the intermittent energy production. One technology cannot solve all problems but hydrogen has already versatile end-use applications and potential to make significant reductions in CO_2-emissions in energy sector, limiting the global temperature rise.

PEM water electrolysis technology for hydrogen production is commercially available but still needs developing to reach its full potential. At the moment, most of hydrogen is produced from natural gas via SMR which emits CO_2 emissions. However, SMR is very mature technology and currently the most feasible method to achieve lowest hydrogen production costs. PEM electrolysis is still operating at small markets which puts the large expected cost reductions into context. Consequently, PEM electrolysis could achieve SMR feasibility through technical development. In addition to hydrogen sales, revenues for PEM electrolyser operator can be expected from the side products, oxygen and waste heat. Electrolyser can also offer capacity to FCR-N market to balance electricity grid and gain extra incomes.

Projects with PEM electrolysis in Power-to-X concepts have been demonstrated recently globally and especially in Europe. Many sectors, such as transportation, industry, and heating can benefit from hydrogen utilization. Projects aim to scale up the plant sizes and improve the technology to be more affordable and available. Demonstrations have been done for example to supply industrial applications, blend hydrogen to natural gas grid, install hydrogen refueling stations and combine hydrogen with CO_2 to create synthetic methane. Ambitious projects to replace fossil fuels with hydrogen are planned and partially started. Building a suitable infrastructure for large scale hydrogen transportation and utilization by replacing the natural gas grid to be fit for 100 % hydrogen. Also a noteworthy attempt to replace today’s blast furnace process with DRI and to have completely fossil-free process for steel manufacturing by 2035.

Practical part of the work included test operations with 30 kW and 4 Nm^3/h nominal H_2 output pilot size PEM electrolysis system. The main task of the PEM electrolyser operation was to study H_2 production and consumption, system stability and behavior. Experimental operation of the pilot scale PEM electrolyser showed excellent dynamic properties and stable hydrogen production. However, with system efficiency of 47 % (LHV) hydrogen production would not be feasible. Hydrogen storage was used as a buffer for possible technical errors. According to calculations, storage could feed the synthesis unit from around 5 hours upwards from full fill, depending on the output rate and required end-use pressure. The cold ambient temperatures caused issues with the cold start. Consequently, a manual ramping to heat the system before full load operation was required. Also, operation at subzero temperatures would require special design of the system to avoid water freezing.

Based on the economic calculations, only speculative 2030 with FCR-N scenario with optimistic assumptions was feasible without raised hydrogen and oxygen prices. The scenarios with FI 2018 and DK 2018 electricity prices could not operate profitably and utilization rates remained low with assumed 2 €/kg hydrogen price. However, with higher future 3.4 €/kg hydrogen and 80 €/t oxygen prices all scenarios made profit. The results are also sensitive for other factors, such as electricity purchase price, system efficiency, utilization rate, and CAPEX. The distribution of yearly costs and incomes for different scenarios showed that main incomes are received from hydrogen sells. Additional revenues from oxygen and heat have significantly smaller but relevant share. FCR-N operation proved to be beneficial in all scenarios but relative significance decreased as utilization rate increased. Furthermore, FCR-N market revenues are limited and auction based. Therefore future auctions are not guaranteed to offer same revenue for balancing services. Feasible electrolysis operation required high utilization rate, in which case costs were dominated be electricity costs other costs played smaller role.

Feasible operational framework for PEM electrolyser can be challenging to achieve in Finland with current system electricity prices and hydrogen target cost. However, if the factors, such as higher hydrogen price, oxygen and heat utilization, and FCR-N revenues can be fully utilized, feasible operation is possible. The development of the electricity price has a major role in the viability of the PEM water electrolysis. Hydrogen in Finland is mainly utilized in industrial sector which allows larger scale operation and consequently lower relative hydrogen production costs. Other possible end-use applications are hydrogen blending into natural gas grid (1% limit), methanation and direct use in the transportation. However, hydrogen infrastructure in these options are at very early stage.

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APPENDIX 1: POWER-TO-X APPLICATIONS AND PROJECTS

Country	Project	Product	Plant size	Year	Reference
Austria	H2Future	H_2	6 MW	2021	[74]
Canada	Air Liquide	H_2	20 MW	2020	[57]
Canada	Enbridge P2G	H_2	2.5 MW	2017	[84]
Denmark	HyBalance	H_2	1.2 MW	2017	[85]
France	Jupiter 1000	H_2 / CH_4	1 MW	2014-2020	[65]
France	GRHYD	H_2	100 kW	2013	[86]
Germany	Refhyne	H_2	10 MW	2018	[87]
Germany	Hydrogen fueling station for FC busses	H_2	1 MW (400 kg/day)	2018	[88]
Germany	MefCO2	CH_4	1 MW	2018	[89]
Germany	WindGas Brunsbuttel	H_2	2.4 MW	2017	[90]
Germany	WindGas Hamburg	H_2	1.5 MW	2015	[72]
Germany	Ibbenbüren	H_2 / Heat	150 kW	2015	[32]
Germany	BioPower2Gas	CH_4	400 kW	2013-2016	[91]
Germany	Energiepark Mainz	H_2	3,9 MW	2015	[8]
Germany	Hassfurt	H_2	2,1 MW	2016	[92]
Norway	Haeolus	H_2 / Electricity	2 MW + 100 kW FC	2018	[79]
Sweden/ Finland	HYBRIT	H_2	multi-MW scale	2016-2035	[78]
Switzerland	Hydrospider AG	H_2	2 MW	2019	[75]
Thailand	EGAT	H_2 / Electricity	1 MW + 300 kW FC	2017	[93]
The Netherlands	Rozenburg	CH_4	7 kW	2013-2018	[64]
Runcorn, Cheshire, UK	Project Centurion feasibility study	H_2 / CH_4	100 MW	2018	[94]
UK	HyDeploy	H_2	500 kW	2017-2020	[69]
UK, Cobham	HYFIVE 1 HRS	H_2	207 kW, 20bar, 80kg/day	2017	[95]

APPENDIX 2: THE MAIN CHARACTERISTICS OF THE ELECTROLYSER (ADAPTED FROM MANUFACTURER H2B2 MANUAL)

Hydrogen Gas Production	
Max. Nominal Hydrogen Flow	4 Nm^3/h
Hydrogen production range	10-100 %
Operation pressure	Up to 20 bar
Hydrogen purity	99,999 %; < 5 ppm O_2; < 5ppm H_2O
Electrical requirements	
Voltage	400 V ± 10 % (3Ph+N+PE)
Frequency	50 Hz ± 5 %
Power (BoP + Stack)	30 kW
Tap feed water	
Consumption	Approx. 8 l/hr
Conductivity	< 2,000 µS/cm (T 25 °C)
Pressure	2-6 bar
Temperature	+5 °C to +30 °C
Water after WTP	
Consumption	< 1 l/Nm^3 H_2
Quality	< 0,1 µS/cm, TOC < 30 ppb

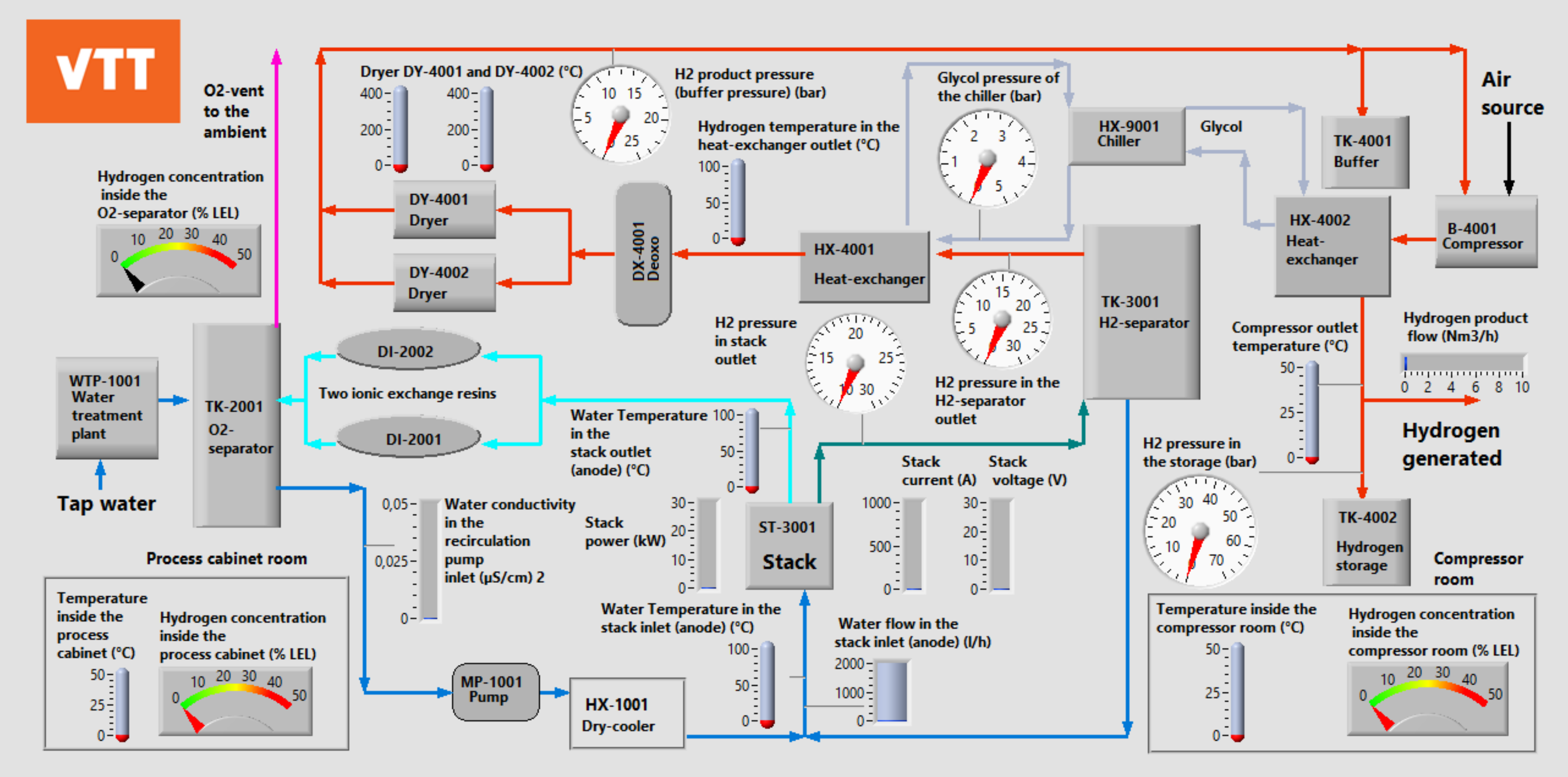
Control system	
PLC	Fully automated and unattended with 7” color touch screen
Communication	Modbus TCP/IP or Profinet (RI45 port)
Environmental conditions	
Temperature range	+5 °C to +30 °C
Humidity	0 to + 95 % (non-condensing)
Air ventilation	Available from a non-hazardous area
Installation area	Indoor/outdoor
Approx. weight	1,000 kg
Standards and regulations	CE

Low pressure buffer	
Storage type	Type I
Pressure	Up to 200 bar
Volume	50 l
Compressor	
Compressor type	Pneumatic
Pressure	Up to 55 bar
Air consumption	49 Nm^3/h (6 bar)
Air quality	Class 4

High pressure storage	
Storage type	Type II
Pressure	Up to 250 bar
Volume	612 l

Container dimensions and weight	
Dimensions (L*W*H)	Container: 10 ft. HC (2,99 x 2,44 x 2,89) m Cabinet: 1,8 m x 0,8 m x 2,1 m
Approximate weight	5,000 kg

APPENDIX 3: VISUALIZED PI-DIAGRAM OF THE ELECTROLYSER SYSTEM



APPENDIX 4: DATA FOR COSTS AND REVENUES OF ECONOMIC CALCULATIONS

10 MW/ Optimized hours/a	FI 2018	FI 2018 FCR-N	DK 2018	DK 2018 FCR-N	Speculative 2030	Speculative 2030 FCR-N
Electricity [k€]	-73.0	-668.3	-175.2	-850.6	-1,420.9	-2,048.7
CAPEX [k€]	-778.4	-778.4	-778.4	-778.4	-455.4	-455.4
O&M [k€]	-415.9	-416.6	-416.2	-416.9	-246.4	-247.0
Hydrogen [k€]	90.7	457.6	230.4	656.7	1,768.5	2,085.6
Oxygen [k€]	8.8	44.5	22.4	63.9	172.0	202.8
FCR-N [k€]	0.0	406.5	0.0	471.9	0.0	684.7
Profit [k€]	**-1,167.7**	**-954.7**	**-1116.8**	**-853.5**	**-182.2**	**222.1**
LCOH [€/kg]	27.7	6.2	11.7	4.6	2.2	1.8
Utilization rate [%]	3	15	7	21	45	53
Produced H_2 [kt/a]	0.1	0.2	0.1	0.3	0.9	1.0

10 MW/ Optimized hours/a/ Heat utilization	FI 2018	FI 2018 FCR-N	DK 2018	DK 2018 FCR-N	Speculative 2030	Speculative 2030 FCR-N
Electricity [k€/a]	-229.0	-1,119.9	-359.9	-1,297.1	-1,752.5	-2,273.2
CAPEX [k€/a]	-778.4	-778.4	-778.4	-778.4	-455.4	-455.4
O&M [k€/a]	-416.1	-417.1	-416.4	-417.4	-246.9	-247.3
Hydrogen [k€/a]	222.8	741.3	387.4	943.4	2,061.9	2,282.4
Oxygen [k€/a]	21.7	72.1	37.7	91.8	200.5	222.0
FCR-N [k€/a]	0.0	522.0	0.0	579.8	0.0	686.9
Heat [k€/a]	31.3	104.2	54.4	132.6	144.9	160.4
Profit [k€]	**-1,148**	**-875.8**	**-1.075.3**	**-745.3**	**-47.6**	**375.7**
LCOH [€/kg]	12.3	4.4	7.5	3.6	2.0	1.7
Utilization rate [%]	7	24	8	4	52	58
Produced H_2 [kt/a]	0.1	0.4	0.2	0.5	1.0	1.1

10 MW/ Optimized hours/a	**FI 2018**	**FI 2018 FCR-N**	**DK 2018**	**DK 2018 FCR-N**	**Speculative 2030**	**Speculative 2030 FCR-N**
Electricity [k€/a]	-4,663.5	-4,045.3	-4,457.9	-3,857.2	-4,339.4	-3,946.5
CAPEX [k€/a]	-778.4	-778.4	-778.4	-778.4	-455.4	-455.4
O&M [k€/a]	-420.8	-420.1	-420.8	-420.2	-250.1	-249.6
Hydrogen [k€/a]	4,857.3	4,196.2	4,864.7	4,215.9	6,562.5	6,091.5
Oxygen [k€/a]	906.5	783.1	907.8	786.8	1,224.7	1,136.8
FCR-N [k€/a]	0.0	630.2	0.0	614.8	0.0	385.8
Heat [k€/a]	401.2	346.6	401.8	348.2	271.0	251.6
Profit [k€/a]	**302.2**	**712.3**	**517.2**	**561.7**	**3,013.3**	**3,214.1**
LCOH [€/kg]	3.2	2.8	3.0	2.7	1.8	1.6
Utilization rate [%]	91	78	91	79	98	91
Produced H_2 [kt/a]	1.4	1.2	1.4	1.2	1.9	1.8