LAPPEENRANTA UNIVERSITY OF TECHNOLOGY
Faculty of Technology
Degree Programme in Energy Technology

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POTENTIAL FOR SYNTHETIC GAS IN FINNISH INDUSTRY

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ABSTRACT

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Potential for synthetic gas in Finnish industry

Master's thesis

2015

126 pages, 33 figures, 10 tables, 3 appendices

Examiners: Professor Esa Vakkilainen, MSc in Technology Katja Kuparinen

Keywords:
natural gas, synthetic natural gas, hydrogen economy, industrial process fuels

Finland, other Nordic countries and European Union aim to decarbonize their energy production by 2050. Decarbonization requires large scale implementation of non-emission energy sources, i.e. renewable energy and nuclear power. Stochastic renewable energy sources present a challenge to balance the supply and demand for energy. Energy storages, non-emissions fuels in mobility and industrial processes are required whenever electrification is not possible. Neo-Carbon project studies the decarbonizing the energy production and the role of synthetic gas in it.

This thesis studies the industrial processes in steel production, oil refining, cement manufacturing and glass manufacturing, where natural gas is already used or fuel switch to SNG is possible. The technical potential for fuel switching is assessed, and economic potential is necessary after this. All studied processes have potential for fuel switching, but total decarbonization of steel production, oil refining requires implementation of other zero-emission technologies.

TIIVISTELMÄ

Lappeenrannan Teknillinen Yliopisto
Teknillinen tiedekunta
Energiatekniikan koulutusohjelma

Heikki Lindfors

Synteettisen maakaasun potentiaali Suomen teollisuudessa

Diplomityö

2015

126 sivua, 33 kuvaajaa, 10 taulukkoa, 3 liitettä

Tarkastajat: Professori Esa Vakkilainen, Diplomi-insinööri Katja Kuparinen

Avainsanat:
maakaasu, synteettinen maakaasu, vetytalous, prosessiteollisuuden polttoaineet

Suomi, muut Pohjoismaat ja Euroopan Unionin maat ovat sitoutuneet dekarbonoimaan energiantuotantonsa vuoteen 2050 mennessä. Tämä tapahtuu muuttamalla energiantuotantoa päästöttömiin energianlähteisiin, eli uusiutuvaan energiaan ja ydinenergiaan. Haasteena on katkoittaisen uusiutuvan energiantuotannon tasaaminen. Siksi on välttämätöntä varastoida energiaa, dekarbonoida liikennepolttoaineet sekä vaihtaa teollisuuden polttoaineet päästöttömiin polttoaineisiin jos sähköistäminen ei ole mahdollista. Neo-Carbon hanke tutkii synteettisen kaasun potentiaalia energiantuotannon dekarbonoinnissa.

Tässä työssä tarkastellaan tarkemmin terästuotannon, öljynjalostuksen, sementintuotannon ja lasintuotannon prosesseja, jossa käytetään maakaasua tällä hetkellä tai polttoaineen vaihto maakaasuun on mahdollista. Työ keskittyy teknillisen potentiaalin arviointiin, ja taloudellinen lisäarvionti kyseisten prosessien kohdalla on tarpeen. Kaikissa tarkastelluissa prosesseissa löydettiin potentiaalia, mutta täydellinen dekarbonointi vaatii myös muiden vähäpäästöisten teknologioiden käyttöönottoa terästuotannossa, öljynjalostuksessa ja sementin valmistuksessa.

FOREWORD

This thesis was made for Neo-Carbon research project, implemented by Lappeenranta University of Technology, VTT and Finland Futures Research Centre. I felt delighted and motivated to work in a professional team on such a progressive topic in field of energy technology.

Professionally I would like to thank Esa Vakkilainen, who was an inspirational advisor for this thesis work, as well as a great professor through my studies.

Personally I would like to thank Jussi and Victoria, who made working in the energy department of LUT more fun and enjoyable. Last, I would like to thank Hanna for reminding me about the life after the master's thesis.

This thesis is dedicated to all the engineers who work hard to make the world more sustainable.

In Lappeenranta 27.3.2015

Heikki Lindfors

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ABBREVIATIONS

BF	Blast furnace
BF-BOF	Blast-furnace/basic oxygen furnace
BFG	Blast furnace gas
CO_2	Carbon dioxide
CO_2-eq	Carbon dioxide equivalent
COG	Coke oven gas
DRI	Direct reduced iron
EAF	Electric arc furnace
EU-ETS	European Union emission trading scheme
FCC	Fluid catalytic cracking
GHG	Greenhouse gas
HM	Hot metal
HVFO	High viscosity fuel oil
IEA	International Energy Agency
LDG	Converter off gas
LNG	Liquefied natural gas
LPG	Liquefied petroleum gas
LULUCF	Land use, land use change and forestry
MET	Metallic charge
MTBE	Methyl-tert-buthylether unit
NG	Natural gas
NGI	Natural gas injection
NO_x	Nitrous oxides
PEM	Polymer electrolyte membrane electrolyzer
SEC	Specific energy consumption
SNG	Synthetic natural gas
SOE	Solid oxide electrolyzer
STD	Standard system
TAME	tert-amyl-methylether unit
TCC	Thermofor catalytic cracking
TGR	Top gas recycling
TPES	Total primary energy supply
VOD	Vacuum oxygen decarburization

1 INTRODUCTION

This thesis studies the potential for synthetic natural gas and hydrogen gas in Finnish process industry. The thesis topic is related to Neo-Carbon project by Lappeenranta University of Technology, VTT and Finland Futures Research Centre which studies synthetic natural gas and hydrogen made from excess electricity from renewable energy sources such as wind and solar power via hydrolysis and methanation. Synthetic natural gas can act as an energy storage for the electricity generated by stochastic renewable energy sources, as a fuel for mobility, or as a fuel for industrial processes. Renewable fuels such as synthetic natural gas are needed to reduce emissions from energy production due climate reasons, while providing reliable and affordable energy for the industry and the society in general.

Synthetic natural gas (SNG) is man-made methane gas. The qualities of the synthetic gas are similar to those of natural gas, and therefore it can substitute natural gas in combustion processes. Furthermore, it can replace other gaseous fuels, such as liquefied petroleum gas, and possible other fuels, such as oil or coal, providing the process is re-optimized for gaseous fuels.

The focus of this thesis is natural gas and hydrogen consumption in Finnish industry. The goal is to identify processes in Finnish industry using natural gas as fuel. This thesis also studies use of hydrogen in industrial processes. Hydrogen is formed by water electrolysis, or methane steam reforming process. Direct fuel use of both synthetic natural gas (SNG) and hydrogen are more efficient than transforming energy from power to gas and back to power, as energy losses in fuel combustion are avoided. Therefore, synthetic natural gas from renewable energy sources to fuel industrial processes will be economically viable sooner than using synthetic natural gas as energy storage medium.

1.1 Aim and definition of the thesis

The current industrial consumption of natural gas and hydrogen are studied in this thesis to identify the potential for synthetic methane and hydrogen gases from power-to-gas technology. Aim is to study the industrial processes in metal, chemical, glass and cement manufacturing industries which use natural gas and could be switched to use hydrogen and methane gases generated in power-to-gas –process, and the processes using another (fossil) fuel as energy source which could be switched to use renewable electrolysis gas as a fuel. This thesis also identifies the processes that use hydrogen as raw material, namely in chemical industry. This thesis is conducted as a literature review.

The metal, chemical, cement and glass industry sectors are globally major natural gas consumers. These sectors are studied due their high energy consumption, high carbon emissions, and/or high current consumption of natural gas. Example manufacturing processes and their fuel use are described in detail, in chapter 6. Pulp and paper industry, energy industry, energy storage systems and traffic fuel use of methane and hydrogen are not the focus of this thesis.

This thesis is conducted as a literature review. Main sources are existing studies on the natural gas consumption. Current natural gas usage in industry is inquired directly from some companies, whose processes are studied in this thesis. Comprehensive study of natural gas consumption in Finland has been done by Stefan Malin and Prosessi-Insinöörit in 1988, ordered by Neste Oil, which was the distributor of natural gas in Finland at the time. The study identified the industrial processes that use natural gas as fuel in late 1980's, excluding the petrochemical industry (Neste Oil's own process). However, as the study is made 30 years ago, the data it represents is out of date and might not be relevant to situation in 2015. Therefore, more recent data is used whenever possible.

1.2 Research questions

This thesis aims to find answers to following questions:

- Which are the largest industrial processes currently that apply natural gas as fuel in Finland?
- Which industrial processes apply hydrogen as fuel or raw material in Finland?
- Is there potential for electrification of the processes that currently apply natural gas as fuel?
- Is there potential for other processes to use gaseous fuels that apply other fuels at the moment?
- How much the fuel substitution to SNG would reduce emissions of the studied processes?

1.3 Structure of the thesis

This thesis represents industry cases in metal, chemical, cement and glass industries that apply natural gas as fuel or hydrogen as a raw material. These cases are described in the chapter 6. Chapter 2 different energy storage systems for renewable energy sources. Chapter 3 describes the chemical and physical qualities of natural gas and hydrogen and their use as a fuel, as well as the technical requirements for burners for the combustion of gaseous fuels. Chapter 4 introduces the power-to-gas process, including the hydrolysis and methanation processes. Chapter 5 provides an overview of the energy sector in Finland and the Nordic countries, including the electricity and heat production, industrial energy use, natural gas use and logistics and the carbon dioxide emissions from energy sector. Chapter 6 introduces the industrial processes with potential for fuel substitution with SNG.

Figure 1.1 depicts the vision of Finnish Gas Association of the role of gas in future energy system. In the vision the gas is used as regulating power, energy storage, as fuel in heat pumps and fuel cells and as traffic fuel. The energy is provided with renewable energy sources (wind, solar and hydro), nuclear power, and, if required, regulating power sources (gas, peat, oil, coal). In the vision of Finnish Gas Association the gas originates from traditional sources as natural gas and LNG, as well as SNG via power-to-gas technology (water electrolysis and methanation). The Neo-Carbon project focuses on power-to-gas technology, which is described in chapter 4 of this thesis. This thesis introduces current and potential applications for gas, and as natural gas and SNG are chemically equivalent, the origin off the gas is less relevant to the application where the gas is consumed.

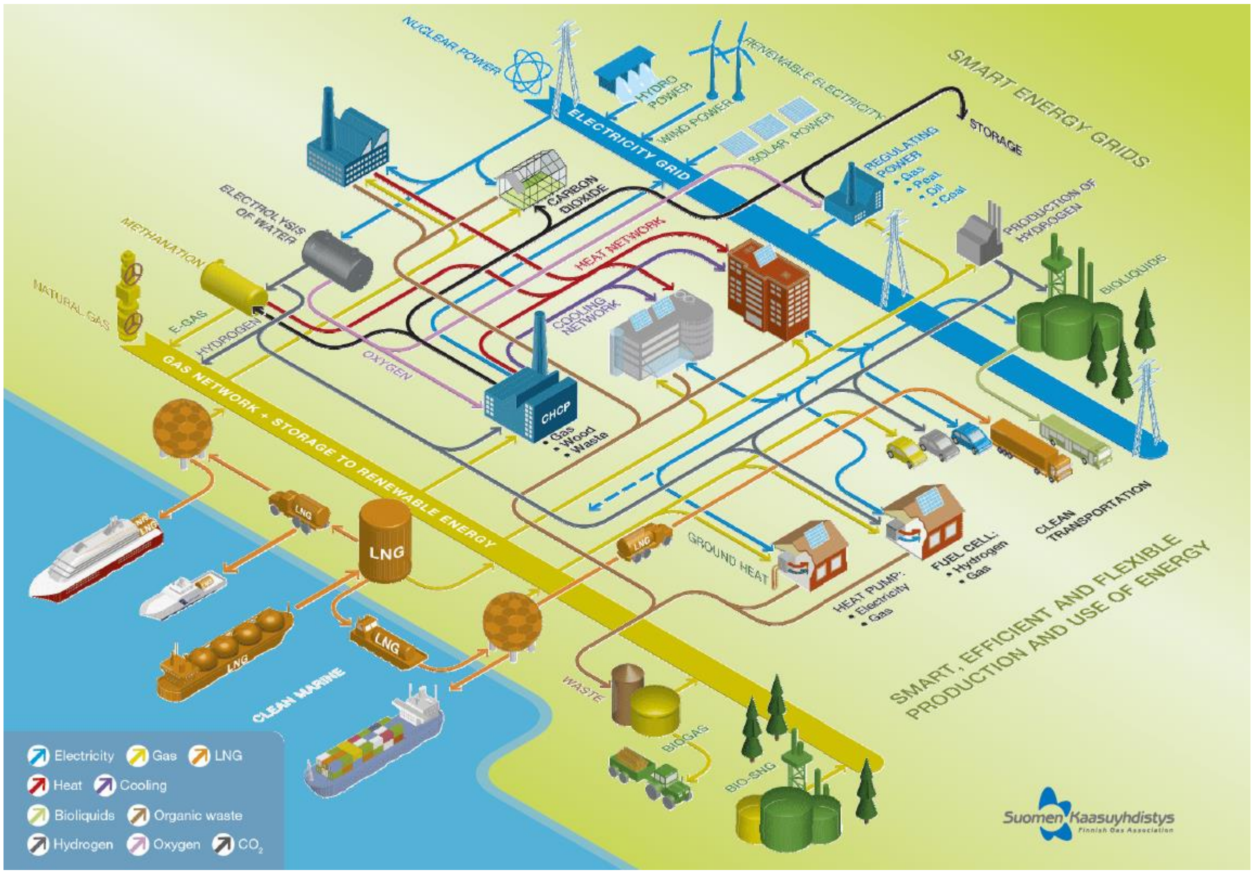

Figure 1.1: Role of gas in smart energy system (Finnish Gas Association)

Figure 1.2 depicts global greenhouse gas (GHG) emissions in year 2000. Iron and steel, chemical, and cement sectors are major carbon dioxide (CO_2) emitters, and therefore examined more closely in this thesis. Additionally, glass industry is also examined, as natural gas has high share of total energy in glass manufacturing. One or more example processes in each of these industries are described, and potential for power-to-gas technology is assessed for the process in question. The industrial processes are described in chapter 6 of this thesis.

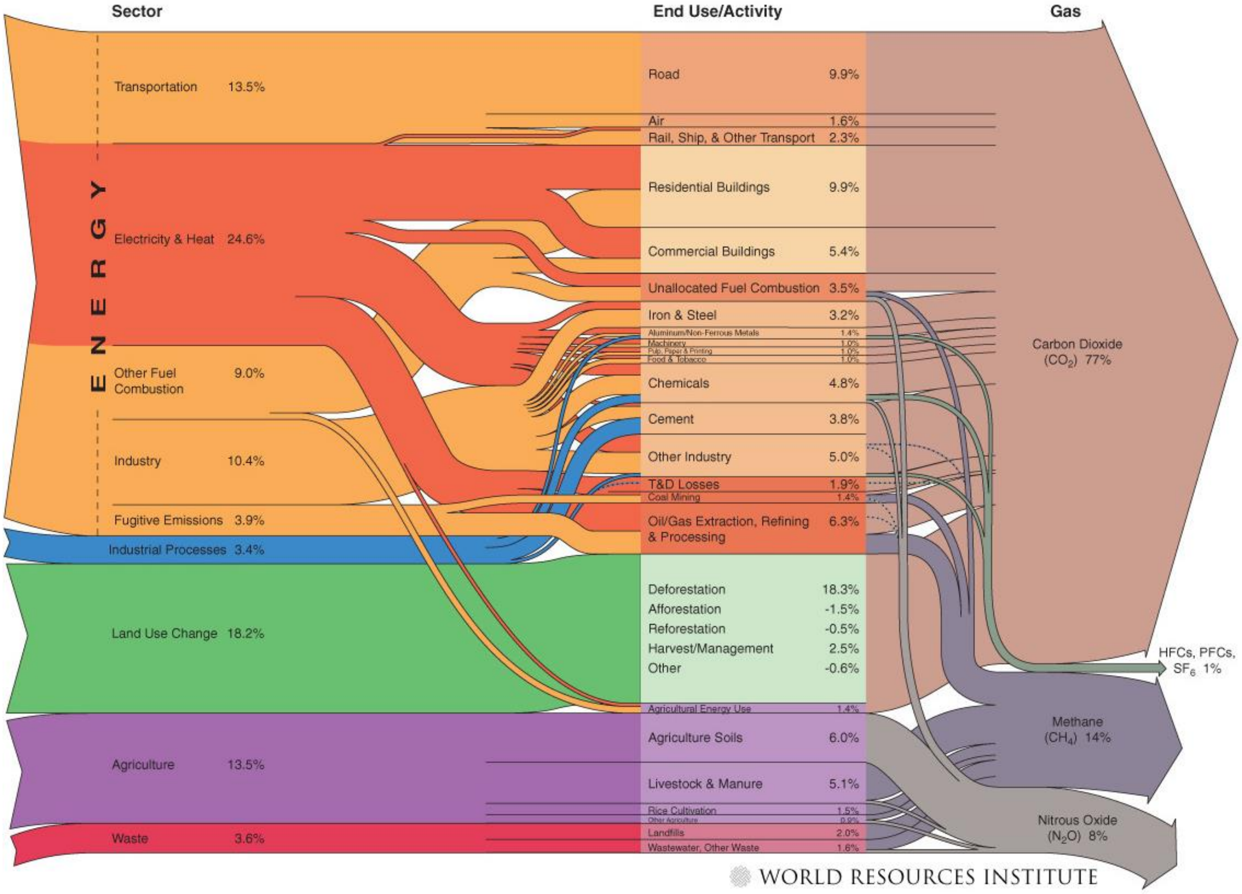

Figure 1.2: World greenhouse gas emissions flow chart (World Resources Institute)

The industrial processes have different characteristics of energy use. The processes may purchase all its energy in form of electricity or heat. The process might acquire some of its energy from raw material processing, such as coke oven gas utilization in ironmaking, and purchase the rest. Or the process might be self-sufficient in energy, or even produce excess energy, such as pulp and paper manufacturing. Processes in the same industry manufacturing same end-product have distinct sub-processes that create a unique energy use profile. Production volume, heat and electricity requirements, sub-processes, support processes, energy efficiency and auxiliary conditions determine the total energy use of the industry. The processes studied in this thesis all have thermodynamical minimum requirement for the process energy use; melting of raw materials in ironmaking and glass manufacturing, crude oil distillation, temperature and pressure requirements of oil refining and cement clinker chemical reaction require certain amount of energy input to reach the required temperatures and other boundary conditions for desired reaction to occur.

The industrial processes manufacturing the same end-product have different energy-use profile. Therefore, it is difficult to determine the typical energy consumption of specific industrial process, even though hundreds of cases of specific process exist worldwide. Therefore, an example case process in Finnish process industry is picked in each studied industry sector and its energy use profile is studied to estimate the potential for power-to-gas generated SNG in the industry sector as a whole.

2 STORAGE OPTIONS FOR RENEWABLE ENERGY SYSTEM

In this chapter, energy storages are discussed generally. Energy storages are required at the times when the energy production is higher than the consumption. This occurs especially in the markets with high share of stochastic renewable electricity production at the times of high wind or high solar production. Power-to-gas technology can be used as an energy storage to produce synthetic natural gas (SNG) from excess electricity. The characteristics and efficiency of power-to-gas technology is compared to other storage methods in this chapter.

In past years, the European Union has made conscious plan to switch the energy production more and more to renewable energy sources. Recently, the US and China, amongst other regions, have started to implement similar policies as well. Main goal of such policies is to mitigate climate change, but renewable energy sources will also reduce energy imports, improving the current account. Some countries, such as the Nordic countries, already had high share of renewables due availability of natural resources, usually hydropower and biomass. Some countries where share of fossil fuels have traditionally been high, such as Denmark and Germany, have achieved to reach high shares of renewables in recent years mostly based on increase of wind, solar and bioenergy generation.

The renewable energy is implemented in all sectors of energy production; hydro, wind and solar in power production, biomass in heat (both building heating and industrial process heating), combined heat and power (CHP) production, and liquid biofuels, gas and electricity in mobility sector. Renewable energy sources have their advantages, disadvantages and problems. Hydropower and biomass have stable production and can be dispatched on demand, but are dependent on availability of local resources. Wind and solar power are available all around the world, but they have fluctuating production curve. The production fluctuates due changing climate conditions, which affect any area with high share of wind or solar power, such as Germany, Denmark, Spain, UK and Italy. Therefore, production have to be stabilized in these areas. The shares of renewable energies are not yet high enough to make this issue critical, however this issue must be addressed in the future.

Different options to manage with fluctuating production include expansion of electricity grid, load management and energy storages. Grid expansion and load management, in both consumption and alternative energy sources should be developed, as they offer solution for the balancing at reasonable cost, with technologies that are already available. However, the potential for both load management and grid expansion is limited, and therefore also the energy storages are needed when the share of renewable energy becomes high.

There are number of different storage technologies, ranging from options that have been available for several decades, to technologies current being developed. The storage technologies can be roughly divided into three categories: mechanical storages (kinetic and potential) such as pumped hydro storages and flywheels, chemical storages such as power-to-gas, and electrical storages such as batteries. Optimal storage for specific energy system depend on several factors. Lehner et al. (2014) list the following variables, which should be considered when assessing the energy storages:

- Storage capacity
- Maximum charging/discharging power
- Possible storage duration
- Efficiency/utilization
- System benefits
- Storage losses
- Total storage potential of all plants
- Temporary availability, guaranteed capacity (time of the day, seasonal dependability)
- Investment costs
- Operational costs (resources, emissions)
- Economic impact (value added effects)
- Site conditions, need for topographic intervention
- Existing infrastructure on site, i.e. power grid
- Conversion possibility, requirement for reconversion
- Public acceptance for new infrastructure projects, environmental impacts

Addressing all these variables is complex. Regarding long-term storages (days-to-months) the following parameters are primary: high storage capacities, high volumetric storage density, system benefits, flexible site-specific modifiability, decentralized application possibility and the possible storage duration (Lehner, et al., 2014). Chemical storages, such as power-to-gas, meet these parameters well.

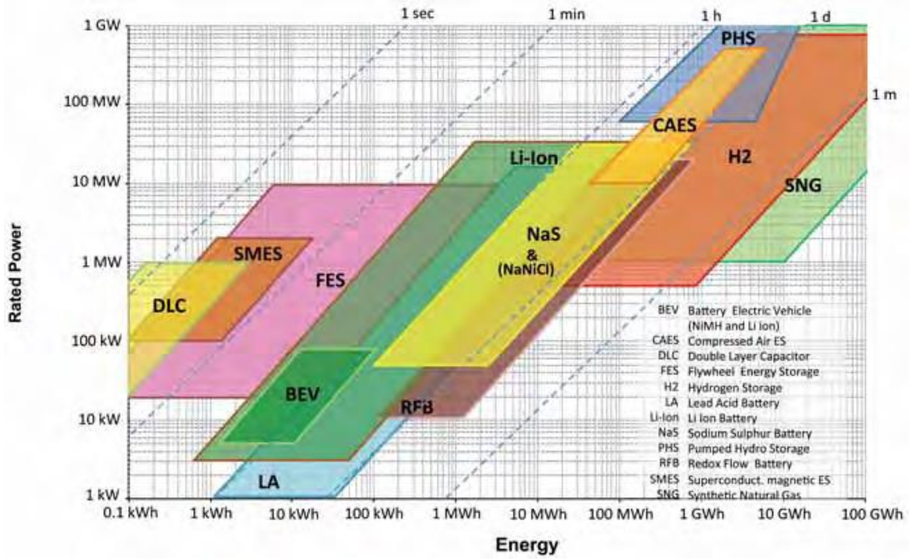

Figure 2.1: Different energy storage comparison by power, timescale and energy (Franhofer ISE)

Figure 2.1 illustrates the suitable ranges of power, time and energy for different energy storage systems. The double layer capacitator, flywheel and superconductor storages are most suitable for low-power, seconds-to-minutes scale energy storage, battery storages are most suitable for low-power, minutes-to-hours timescale storage, and compressed air, pumped hydro storages and power-to-gas applications are most suitable for high energy, days-to-months timescale energy storage applications. In the Nordic countries the months-timescale energy storage applications have critical role in renewable energy-based energy system, as solar energy can be collected in high amounts in long summer days, and the consumption is the highest in dark and cold winter months.

The efficiency, capacity rating and time scale of different storage technologies are compared in the table 2.1.

Table 2.1: Efficiency, capacity, and time scale comparison of different storage technologies (Lehner, et al., 2014)

Technology	**Efficiency**	**Capacity rating [MW]**	**Time scale**
Pumped hydro storage	70-85%	1-5000	Hours-months
Li-Ion battery pack	80-90%	0,1-50	Minutes-days
Lead acid battery	70-80%	0,05-40	Minutes-days
Vanadium redox battery	65-85%	0,2-10	Hours-months
Sodium sulfur (NaS) battery	75-85%	0,05-34	Seconds-hours
Nickel cadmium (NiCd) battery	65-75%	45	Minutes-days
Flywheel	85-95%	0,1-20	Seconds-minutes
Compressed air	70-75%	50-300	Hours-months
Power-to-gas	30-75%	0,01-1000	Minutes-months

In order to provide electricity with renewable energy sources, different energy storage mediums as well as demand, extended grid capacities are needed to provide energy without intermissions. Pumped storage is well-established energy storage method. Water is pumped into a reservoir in high altitude, and released through turbines when the electricity is needed. The efficiency of the system in 70-85%, which is comparatively high. However, the availability of potential sites for pumped hydro varies between regions; mountainous areas have more available sites. There are not enough available sites for pumped hydro for large-scale integration of renewable energies (Bajohr, et al., 2011) (Klaus). Furthermore, pumped hydro reservoirs dramatically alter the landscape, and therefore the public acceptance of new projects is generally low. Compressed air storage pressurizes air to store energy, which is reconverted to electricity by turbines. The drawbacks include very large space requirement and high cost. Additionally, part of the energy is converted to heat, which should be utilized to increase efficiency. Flywheel is a short-term mechanical storage medium, which can capture of release large amounts of energy within seconds. However, it is not suitable for long term energy storage. Rechargeable batteries are widely used in small scale consumer applications (electronics, cars). However, in large scale their high specific cost limit their use as energy storage.

Power-to-gas process can provide a fuel for traffic, energy storage from electricity which is needed when the share of stochastic electricity production (wind, solar) becomes high, or to fuel industrial processes which are fueled by fossil fuels now and are challenging to electrify. The industrial processes with potential to switch to power-to-gas generated methane in metal, petrochemical, fertilizer, glass and cement manufacturing industries are discussed in this thesis. Beside the flexibility to use methane in various forms, power-to-gas offers high volumetric energy density. However, the power-to-gas suffers from efficiency losses in all energy conversion steps. Power-to-gas technology is discussed in more detail in chapter 4.

3 QUALITIES OF NATURAL GAS AND HYDROGEN

The methane and hydrogen gases have distinct characteristics compared to other fuels, which affect the application where the fuel is used. Chemical and fuel characteristics are described. Furthermore, suitable burner applications are described for the gaseous fuel use. The characteristics of the gaseous fuels are different from solid fuels, which affect the combustion process, and fuel switching might have an impact other process machinery.

3.1 Qualities of natural gas

The quality of natural gas is dependent on the source; different gas fields have different gas composition. Natural gas consists mostly of methane (CH_4). Additionally, other hydrocarbons are present, such as ethane (C_2H_6) and propane (C_3H_8), as well as inert gases such as nitrogen (N_2). (Ala-Outinen, 1991)

Natural gas used in Finland originates from Siberia, and is considered to be purest in the world, consisting 99% of methane. Impurities, such as sulfur compounds, are very low (less than 0,001g/m^3n), and share of inert nitrogen is less than 0,9%. The Siberian gas has no mechanical impurities, such as resins or condensates. (Ala-Outinen, 1991)

Typical chemical composition of Siberian natural gas (Blomster, 1988):

Methane (CH_4)	98,9 %
Ethane (C_2H_6)	0,16 %
Propane (C_3H_8)	0,02 %
Nitrogen (N_2)	0,87 %
Carbon dioxide (CO_2)	0,02 %
Oxygen (O_2)	0,002 %

Natural gas is lighter than air. In normal conditions (0 °C, 1 bar), its density is 0,723 kg/m^3, thus its relative density compared to air is 0,56. There is significant difference to liquefied gases, such as propane, which has a density of 2,0 kg/m^3 and relative density of 1,56. The energy density of natural gas is lower than liquefied gases due lower molecular weight of methane (16 g/mol), than for example propane (44 g/mol). Therefore, 2,6 times greater volumetric flow is required to get same amount of energy compared to propane. (Ala-Outinen, 1991) (Blomster, 1988)

Natural gas is colorless, odorless and non-toxic gas. It combusts in air in 5-15 volume-% range, and has a flash point of 650 °C. To make natural gas observable, tetrahydrotiofene (C4H8S) is mixed in it. This odorant is also used in liquefied gases, and it burns perfectly and is therefore not present in flue gases. Boiling point of methane is -162 °C (Ala-Outinen, 1991). The lower heating value of Siberian natural gas used in Finland is 9,89 kWh/m^{3}n (or 35,60 MJ/m^{3}n), which practically varies very little (Ala-Outinen, 1991) (Higher heating value is 11,0 kWh/m^{3}n, however this is rarely used.) (Blomster, 1988).

3.2 Qualities of hydrogen

Hydrogen is the lightest and simplest chemical in existence, consisting of one proton in the nucleus, and one electron. It is 15 times lighter than the air in gaseous form, with density of 0,0899 kg/Nm^3. It has the highest energy content per mass, with higher heating value (HHV) of 3,54 kWh/Nm^3 (39,42 kWh/kg), which is 2,5 higher than methane and 3 times higher than gasoline, and lower heating value of 33,33 kWh/kg. (Ursúa, et al., 2012) Hydrogen gas consists of two hydrogen atoms (H_2), however hydrogen rarely occurs in nature in this gaseous form, but rather in molecular form with other compounds, for example water (H_2O). It is colorless, odorless and non-toxic gas. It combusts in air in 4-75 volume-% range, and has a flash point of 574 °C. The combustion forms water (H_2O), which is the only flue gas from hydrogen combustion. Boiling point of hydrogen is -253 °C (Pirilä, 1981).

Hydrogen gas can be manufactured from natural gas by steam reforming, or from water by electrolysis. Both these processes result in pure hydrogen gas (H_2). (Pirilä, 1981)

3.3 Suitable burners for natural gas

Methane gas is applied in industry as either fuel, or raw material for chemical industry. In this chapter, the suitable burners for fuel use is discussed. First, different burning techniques are discussed in general level.

There are several burner types for different applications. Some are suitable for only natural gas, some for other fuel types as well. (Huldén, 1972, pp. 7/1-2) et al. divide burners into three main groups:

a. *Diffusion burners*. In diffusion burners, the mixing of fuel and oxygen occurs after the fuel exits the burner by diffusion. Therefore, the flame speed is limited by the rate of diffusion.

b. *Burners with premixing*:

    b.1 Burners with partially complete premixing. In these burners, only part of oxygen is premixed with fuel in the burner.

    b.2 Burners with imperfect premixing. In these burners part or all oxygen is premixed with fuel in the burner. Premixing is imperfect, so the fuel-oxygen mix is heterogeneous.

c. *Kinetic burners*, a.k.a. premixing burners. In kinetic burners the fuel and oxygen are perfectly mixed in the burner. In these burners, the flame speed is only dependent on the time of mixing the fuel and oxygen molecules.

Different processes have different requirements for the flame, and therefore the choice of the burner is dependent on the industrial process. The requirement for the flame might be, according to (Huldén, 1972, pp. 7/3-4) et al.:

The flame must be long and illuminative. This requirement is common with rotating kilns which might be used for drying or inducing chemical processes. Example processes are cement production and lime kilns, as well as some steel industry kilns (i.e. open hearth furnaces). Diffusion burners are optimal for these kind of processes. The fuel and oxygen are injected into the kiln in separate parallel streams. The method can be applied for natural gas, as well as coal or oil. However, when using natural gas the flame is less illuminative which results in reduced heat radiation, and natural gas requires less injected oxygen (at some cases, none).

The flame must fit in limited space. Limited combustion space requires this, which might occur in steam boilers which are optimized for other fuel types. Burners with imperfect premixing are often used in steam boilers. By injecting oxygen tangentially, the length and angle could be adjusted.

The flame must be short and nonluminous. Burners used for drying and heat treatment processes in glass- and ceramics industries and metallurgy require this type of flame. Kinetic burners using natural gas are optimal to meet this requirement. Coal and oil combustion might not be suitable to meet this requirement.

The heat transfer must occur mainly by radiation. This is required in those drying and heat treatment processes, where product does not tolerate high temperatures, and/or contact of flue gas to product would impair the quality of the product. Suitable solution in this case is to use type of kinetic burner which is optimized for produce radiant heat. These burners premix fuel and oxygen, and the mixture is injected through small pipes, porous material, or metal net. The combustion occurs after this, because the speed of the mixture through the net or porous material is greater than the flame speed. When the mixture enters combustion space, its velocity reduces and combustion occurs fast, practically on the surface of the burner, and starts to radiate. Coal and oil cannot be used in this type of burner, only natural gas.

Table 3.1 summarizes the requirements for combustion process, represents industrial applications and suitable burners to meet these requirements.

Table 3.1: Burner flames for different applications (Huldén, 1972, p. 7/5)

Requirements for combustion	Example applications	Common burner types	The burner is suitable to use:		
			coal	oil	gas
1. Long illuminative flame	Rotating kilns: cement, limestone, mesa, drying Melting pots: glass, metal	Diffusion burner	yes	yes	yes
		Combined diffusion- and imperfect premix burner	yes	yes	yes
2. Limited space for flame; illuminative flame	Steam boilers Hot water boilers Hot gas generators Waste combustion Heat treatment processes	burners with imperfect premixing	yes	yes	yes
3. Short nonluminous flame	Drying ovens Heat treatment ovens Crucibles Spot heating	kinetic burners	no	no	yes
		burners with imperfect premixing	no	no (yes)	yes
4. Maximal heat transfer by radiation	Drying Heat treatment	kinetic burners	no	no	yes

4 POWER TO GAS TECHNOLOGY

The power-to-gas technology is described in this chapter. The overall process is described first. Plant scales are estimated, synergy potentials are described as they are relevant to industrial power-to-gas applications, logistics are described and power-to-gas efficiency is estimated from the recent studies. Furthermore, current technologies for electrolysis and methanation processes are described in the chapters 4.2 and 4.3 respectively.

Power-to-gas is emerging energy storage technology that might have a key role in future energy system. The product is renewable hydrogen or methane, which can be used in electricity production, mobility, or process fuel, using current energy infrastructure. Even though the power-to-gas technology is in demonstration phase, the applications for it are mature. Figure 4.1 depicts a gas fueled vehicle by Audi, and how power-to-gas can create a new value chain for a traditional mobility application.

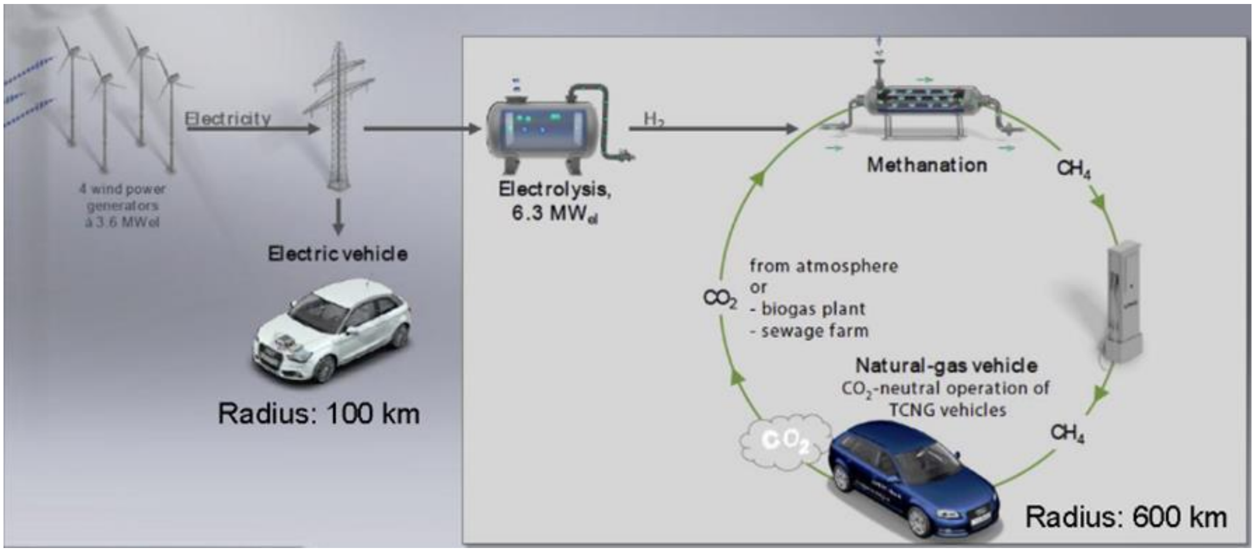

Figure 4.1: Natural gas vehicle in power-to-gas value chain (Audi)

As the figure 4.1 depicts, gas powered vehicles are superior to current electric vehicles, another emerging technology in the mobility sector. The origin for the energy is the same (electricity), but the power source of the end use application differs. Even though the energy provider needs to invest in electrolysis and methanation technologies, the end user prefers the better performance of the car. Similar cases can be found in industry, where the gaseous fuels are better than electrification of the processes, which is the topic of this thesis. To understand the value chain upstream from the end-use application, power-to-gas process is described in this chapter.

4.1 Power-to-gas process

Renewable energy is transferred to the power grid. In power grid, there has to be constant balance between supply and demand. The power grid has to be at constant 50 Hz frequency, and more than 0,1 Hz variation from the frequency increases the likelihood of blackouts. As of 2014, the grid stability can be easily maintained in Nordpool grid, as there is large amount of flexible hydropower potential available in Nordpool, which can be ramped up or down very fast depending on the demand. However, as the share of stochastic renewable energy sources increase, there will be demand for other means to balance demand and supply, such as demand side control or energy storages.

Power-to-gas concept is an energy storage, where excess electricity is transformed into hydrogen (H_2) and oxygen (O_2) by water electrolysis. Oxygen can be released to the air, or preferably applied in industrial processes, such as chemical or metal industry, or in oxyfuel combustion to increase the efficiency and/or temperature of the combustion process. However, the demand for oxygen depends on local conditions, particularly the distance to the potential consumers, consumer demand and storage capacity. The actual product is hydrogen which can be transported by either own hydrogen transportation grid, as admixture in the natural gas grid, or by road or rail transport. Hydrogen can be stored in dedicated facilities, or in natural gas storage infrastructure.

Hydrogen can then be transferred back to electric power at demanded times, or as feedstock to industry. Particularly, chemical, petrochemical and metal industries consume hydrogen in their processes, which currently is supplied from natural gas by steam reforming. However, the use of hydrogen is limited by lack of hydrogen infrastructure, the capacity of industrial applications, or the maximum allowable content in natural gas grid. Therefore, hydrogen can further transformed into methane (CH_4) by methanation process. Hydrogen and carbon dioxide (CO_2) can be transformed into methane by chemically or biologically catalyzed reaction. This methane is called synthetic natural gas or substitute natural gas (SNG). The by-product from this process is steam (H_2O). The carbon dioxide can be supplied from industrial processes, fossil fuel plants, biogas plants or from atmosphere or sea water. The concentration of carbon dioxide is much lower in atmosphere and sea water, making them energy-intensive sources. Pure carbon dioxide sources are rare, making carbon capture an important sub-process in power-to gas concept.

SNG is chemically and physically similar to natural gas and no changes to natural gas infrastructure or processes are necessary. Therefore, SNG can be applied and stored in the natural gas infrastructure without limitations. Transport and storage options for natural gas already exist; there are 134 subsurface storage facilities in Europe, with aggregate capacity of 94 billion m^3 of natural gas, equivalent of up to 1000 TWh. Likewise, applications for methane exist; gas can be transformed back to power with high efficiency in combined cycle plants, utilized as fuel in transportation or used to power or as feedstock to industrial processes. This makes power-to-gas concept not only economically interesting, but also avoids conflicts with permits from authorities and with public acceptance. (Lehner, et al., 2014)

4.1.1 Plant scale

The scale of power-to-gas plants may vary from a few hundred kW up to several hundred MW. The scale of the plant affects the process, and therefore the efficiency; small power-to-gas plant (few 100 kW) may use carbon dioxide from biogas plants, and may use biological methanation.

MW scale power-to-gas plants require industrial carbon dioxide sources, and preferably chemical methanation. (Lehner, et al., 2014)

4.1.2 Synergy potentials

The power-to-gas process utilizes excess energies from renewable energy sources, stabilizes the power grid or substitutes transport capacities by natural gas grid, converts renewable energy for long-term storage. The main purpose of the power-to-gas plant can be any of these, or combination of them. Due these requirements, the plant has to be flexible, easily up-scalable and modular. The affordability is dependent on, for example, annual operating hours, the electricity cost, the cost of methane or hydrogen and oxygen, cost of carbon capture and utilization of excess heat from the process. The methanation is an exothermic process, and released heat can be utilized in carbon capture for methanation. The excess oxygen from the methanation process depends on the industrial user; for example oxyfuel combustion, where the temperature and efficiency of the combustion process is boosted by injection of pure oxygen to the flame, requires large amounts of oxygen. (Lehner, et al., 2014)

4.1.3 SNG and hydrogen logistics

The optimal logistics for power-to-gas product gases is to apply them locally in industrial processes or power production, or to use current gas infrastructure and logistics chain to transport them to utilization sites. Unlimited injection of SNG into the gas grid is possible, as the natural gas consist mainly of methane. However, the methanation reaction produces steam as a side product, and not all hydrogen and carbon dioxide is converted to methane. Therefore, upgrade of SNG is required prior to gas network injection.

Injection of hydrogen into natural gas grid is possible in some extent, however several issues have to be considered. Admixture with hydrogen lowers the Wobbe-index and heating value of natural gas. Natural gas quality requirements must be complied, usually 5 to 15 % admixture of hydrogen is possible. (Myller-Syring & Henel, 2014) The pipelines can tolerate admixture of hydrogen up to 30 %, but the leakage rates might increase. (Florisson, 2014; Myller-Syring & Henel, 2014) Hydrogen has lower volumetric heating value than natural gas; an admixture of 10 % hydrogen lowers the transport capacity by 5-6 %. However, the full transport capacities of natural gas grid are used only few days a year. (Myller-Syring & Henel, 2014) Domestic appliances can tolerate up to 20 % hydrogen, but gas turbine manufacturers limit the hydrogen content to 1 to 2 %. However, laboratory tests of gas turbines have shown the possibility of up to 14 % hydrogen content. (Myller-Syring & Henel, 2014) Gas motors show similar considerations; hydrogen admixture lowers the methane number in automotives, which might result in exceedance of knocking limit. (Myller-Syring & Henel, 2014) Myller-Syring et al. conclude that concentration of hydrogen should be limited to 2 % in case filling stations are connected to gas grid, and 10 % if no filling stations, gas turbines or gas motors are connected. Current natural gas logistics in Finland are introduced in chapter 5.5.

4.1.4 Efficiency of power-to-gas process

Energy losses occur in all energy conversion processes. Therefore, it is economic to use electric power directly whenever possible. The potential to use electric power can be increased by increasing grid transfer capacity, or by increasing demand, for example by electrification of industrial processes. However, there is limited potential for both grid expansion and demand increase, and therefore energy storage options are necessary when the share of stochastic renewable energy generation increases in the future.

The first product of power-to-gas process chain is hydrogen, which can be utilized in the chemical, petrochemical and metal industries. The industrial utilization of hydrogen requires the location of industrial consumer and electrolysis plant to be in vicinity of each other, or hydrogen transport infrastructure, which is poor as of 2015. Storage of hydrogen would enable buffering and decoupling at the demand side. Natural gas grid is potential storage for hydrogen, but the share of hydrogen cannot exceed the requirements of the grid and end-use application.

The second product of power-to-gas process chain is synthetic methane (SNG). Methane can be utilized unrestricted in any existing process that apply natural gas as fuel, as well as in natural gas infrastructure. The conversion from hydrogen to methane can be done either chemically, with reported efficiency of 70-85%, or biologically, with more than 95% efficiency. (Grond, et al., 2013)

Methane can be used to convert back to electricity in combined cycle power plants. Advantage of this is the potential to locate the power-to-gas plant anywhere in vicinity of natural gas grid, regardless of the location of the renewable energy source. Disadvantage of this is the lowest efficiency of all possibilities. Slightly better conversion rate can be achieved by producing electricity from hydrogen. Gas turbines, fuel cells and reverse fuel cells can be utilized for this purpose. Hydrogen can also be utilized in mobility, however the fuel cell powered cars and hydrogen infrastructure are not present in large scale as of 2015.

Table 4.1: Efficiencies for different power-to-gas process chains (Sterner, 2009)

Path	Efficiency (%)	Boundary conditions
Electricity to gas		
Electricity → Hydrogen	54-72	Including compression to 200 bar (underground storage working pressure)
Electricity → Methane (SNG)	49-64	
Electricity → Hydrogen	57-73	Including compression to 80 bar (underground storage working pressure)
Electricity → Methane (SNG)	50-64	
Electricity → Hydrogen	64-77	Without compression
Electricity → Methane (SNG)	51-65	
Electricity to gas to electricity		
Electricity → Hydrogen → Electricity	34-44	Conversion to electricity: 60%, compression to 80 bar
Electricity → Methane → Electricity	30-38	
Electricity to gas to CHP		
Electricity → Hydrogen → CHP	48-62	40 % electricity and 45 % heat, compression to 80 bar
Electricity → Methane → CHP	43-54	

Both power-to-gas and gas-to-power conversion processes produce excess heat, and the utilization of this process heat in industrial processes or district heating would increase the overall energy efficiency of the process. Table 4.1 summarizes the efficiencies of different power-to-gas process chains. However, no straightforward conclusions should be made from efficiencies alone; systemic, economic and macroeconomic aspects should also be considered. (Lehner, et al., 2014) Also, these values represent the current technologies, and the efficiencies might improve in the future.

4.2 Water electrolysis

Although hydrogen is the most abundant element in the universe, it cannot be found in pure state in nature and therefore has to be manufactured. At the moment (2014), 96 % of the hydrogen is produced from fossil fuels and 4 % from water via electrolysis. Hydrogen can be produced from hydrocarbons by steam reforming and partial oxidation from methane, or by coal or biomass gasification. Biomass can also be processed biologically to form hydrogen, e.g. by fermentation. Hydrogen can be produced from water via electrolysis, and possible in the future by thermal and photocatalytic decompositions. When moving towards renewable energy system, hydrogen production from water by electrolysis, powered by renewable electricity sources, is most interesting option. After drying and purifying the hydrogen from oxygen, the purity can reach 99,999 %, which is great advantage against hydrogen production from fossil fuels and biomass.

Electrolysis is based on applying direct electric current to water to dissociate its molecules into hydrogen (H_2) and oxygen (O_2). The current flows between two electrodes immersed in an electrolyte to raise the ionic conductivity. The general electrolysis reaction is:

$$H_2O \rightarrow H_2(g) + \tfrac{1}{2}O_2(g) \qquad (1)$$

Ursúa et al. (2012) describe the electrolysis process as follows:

"In the electrolysis process, the electrons are taken or released by the ions at the electrodes surface, generating a multiphasic gas-liquid-solid system. The reduction half-reaction takes place at the cathode. The electrons flow to this electrode from the outside circuit and polarize it negatively. The oxidation half-reaction occurs at the anode. The electrons leave the anode to the outside circuit, polarizing it positively. Hydrogen is hence generated at the cathode and oxygen at the anode."

There are three electrolysis methods available; advanced alkaline electrolyzers are matured to the stage of large-scale hydrogen production. Polymer electrolyte membrane electrolyzers are commercially available, but their costs are high and they seem more suitable for low-scale applications. Solid oxide electrolyzers produce hydrogen from steam at high temperature, and are currently at R&D stage and show great promise in terms of efficiency and therefore the operating costs.

4.2.1 Alkaline water electrolyzers

Alkaline water electrolysis is an old method to produce hydrogen and oxygen from water. The method was widely available in the beginning of 20th century; there were more than 400 alkaline water electrolyzers in use in 1902, and is the most common electrolyzer technology nowadays. The technology is reliable and safe, and the lifetime of the machinery can reach up to 15 years.

Figure 4.2 illustrates the operating principle of alkaline water electrolyzer cell. The cell has two electrodes with gas-tight diaphragm in between. The cell is filled with potassium hydroxide (KOH) solution (sodium hydroxide or sodium chloride are sometimes also used), to maximize ionic conductivity. The drawback of this principle is the corrosive character of the solution. The cell operates in temperature range of 65 °C to 100 °C, and usually has 47 % to 82 % efficiency.

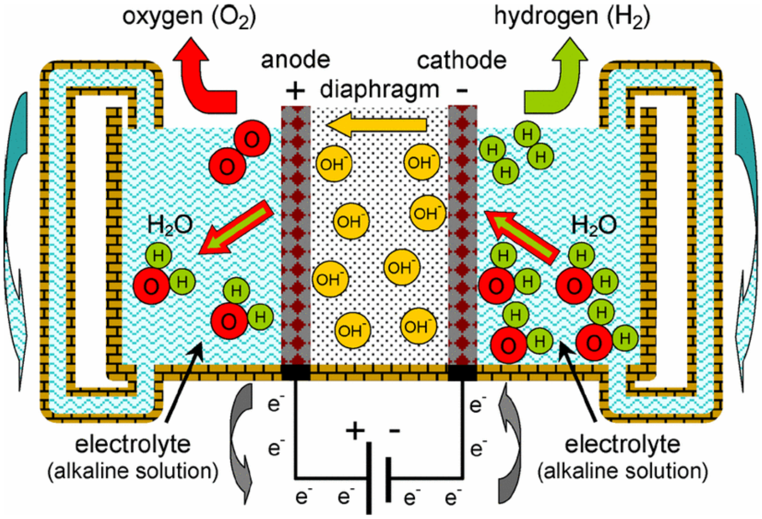

Figure 4.2: Schematics of an alkaline electrolysis cell

Hydrogen is formed at the cathode, where water is reduced:

$$2H_2O(l) + 2e^- \rightarrow H_2(g) + 2OH^-(aq.) \qquad (2)$$

The formed hydroxide anions (OH^-) cross the diaphragm to the anode. The hydroxide anions recombine on the surface of the anode, producing oxygen and closing the electric circuit:

$$2OH^-(aq.) \rightarrow \tfrac{1}{2}O_2(g) + H_2O + 2e^- \qquad (3)$$

Purity of hydrogen and oxygen can reach up to 99,9 % and 99,7 %, respectively, without external purification process. However, the purity of the feed water needs to be significantly high, with electronic conductivity of 5 µS/cm or less. (Ursúa, et al., 2012)

4.2.2 Proton exchange membrane electrolyzers

The ion exchange polymers were developed in the 1950's for the space program, and in 1966 General Electric introduced the first water electrolyzer based on the proton conducting concept using a polymer membrane as the electrolyte. The first commercial application came to market in 1978.

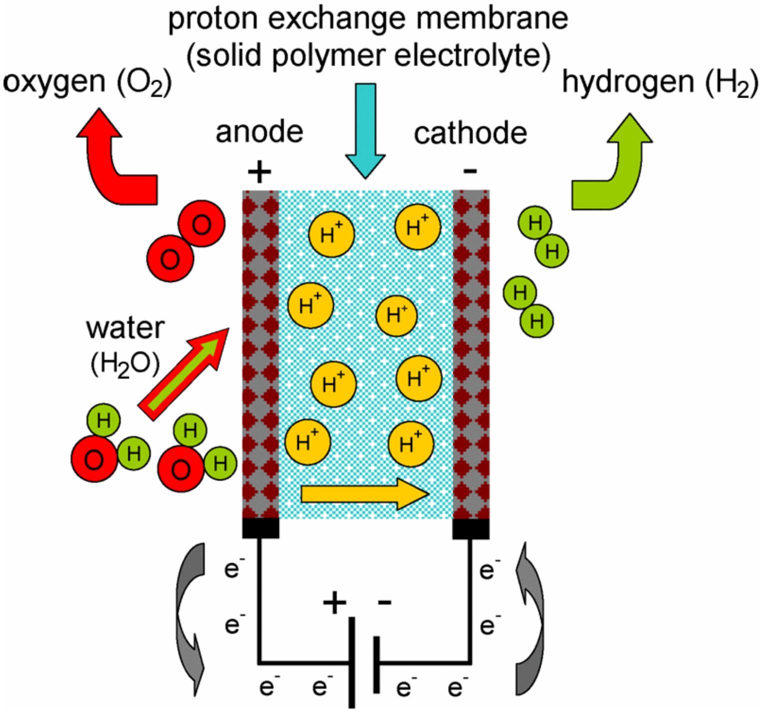

Figure 4.3: Schematics of a proton exchange membrane electrolysis cell

Figure 4.3 illustrates the operating principle of proton exchange membrane (PEM) electrolysis cell. The electrolyte is gas-tight polymeric membrane, with a cross-linked structure and acid character, due to the presence of functional groups of sulfonic acid ($-SO_3H$) type. These groups conduct the proton (H^+) through the ion exchange mechanism. The electrodes consists of noble metals such as platinum or iridium.

The water is oxidized at the anode, producing oxygen, electrons, and protons according to:

$$H_2O(l) \rightarrow \frac{1}{2}O_2(g) + 2H^+(aq.) + 2e^- \qquad (4)$$

The protons (H^+) cross the membrane to the cathode where they are reduced, closing the circuit and producing hydrogen:

$$2H^+(aq.) + 2e^- \rightarrow H_2(g) \qquad (5)$$

Purity of hydrogen can reach up to 99,99 % without external purification process. However, the purity of the feed water has to be even higher than in alkaline water electrolysis, with electronic conductivity of 1 µS/cm or less.

PEM electrolyzers are commercially available. However, they have high investment cost, mostly due cost of membranes and noble metal electrodes. PEM electrolyzers have shorter lifespans than alkaline electrolyzers, and their hydrogen production capacity should be improved for large-scale hydrogen production. (Ursúa, et al., 2012)

4.2.3 Solid oxide electrolyzers

Solid oxide electrolyzers (SOE) are an emerging technology. The concept of solid oxide electrolyzers were developed in the 1960’s. In SOE, the electrolysis of steam occurs in high temperatures (600 ° C – 900 ° C), resulting in high efficiency of the process as significant part of the energy can be provided with heat, instead of more expensive electricity.

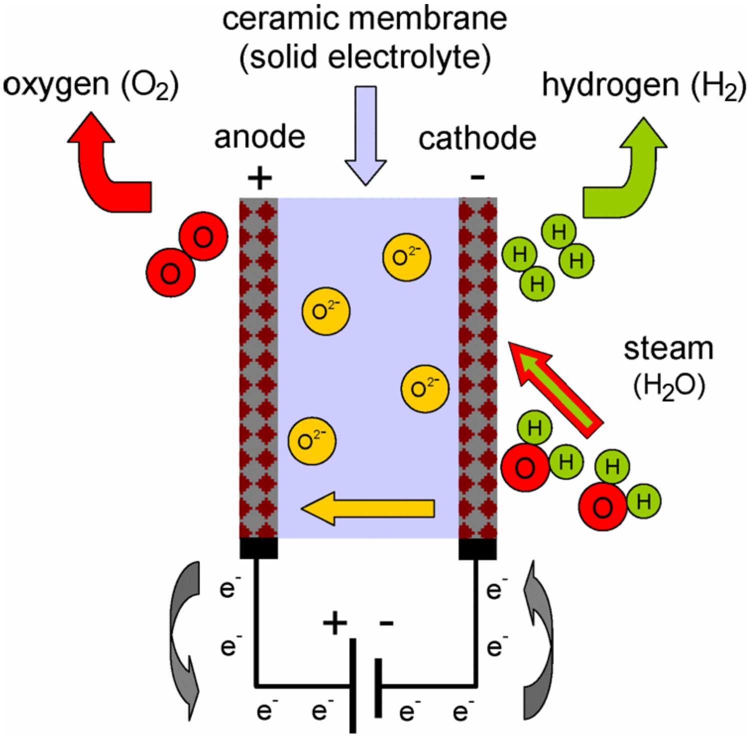

Figure 4.4: Schematics of a solid oxide electrolysis cell

Figure 4.4 illustrates the operating principle of solid oxide electrolysis cell. The electrolyzer usually consists of gas-tight film of yttria (Y_2O_3)-stabilized zirconia (ZrO_2), which provides high ionic conductivity in high temperatures.

Water and recycled hydrogen are fed to the cathode, where water is reduced to hydrogen:

$$H_2O(g) + 2e^- \rightarrow H_2(g) + O^{2-} \qquad (6)$$

The oxide ions (O^{2-}) pass through the solid electrolyte to the anode, where they recombine and close the electrical circuit:

$$O^{2-} \rightarrow \tfrac{1}{2}O_2(g) + 2e^- \qquad (7)$$

The high-temperature lowers the energy intensiveness, due significant part of input energy can be supplied as heat instead of more expensive electricity, and therefore the operating costs of electrolysis process. The technology is attractive at the sites where high-temperature heat source is available, such as in nuclear or geothermal plants. While high-temperature operation of the cell allows lower cell voltages and increase the electrochemical reactions, stability of the materials and sealing issues persist. Moreover, the process requires purification of hydrogen, as the hydrogen and steam are mixed in the cathode, which results in higher capital costs than with liquid water electrolysis. The electrolysis cell degradates faster than in other electrolysis methods, and this stability in the long term is main issue at the moment. Currently, the SOE technology is in R&D stage, and it is estimated that it will take at least a decade before the technology is commercially available. (Ursúa, et al., 2012)

4.2.4 Current status

Alkaline and polymer electrolytic membrane (PEM) electrolyzers are commercially available. However, only alkaline electrolyzers have large enough capacity to meet the large demand for hydrogen or SNG applications. Current high prices of both systems divest investments for electrolyzer applications. Significant reduction of both investment and operating costs should be reached for large scale market penetration. In this regard, advanced solutions such as solid oxide electrolyzers (SOE) can provide more economical solution. However, development of SOE technology is required to improve the performance and lifetime of SOE systems.

4.3 Methanation

Methanation is synthesis of methane gas from hydrogen via biological or chemical path. It is the second step of power-to-gas process. The theory of methanation has been known for more than a century, and the technical application is mature and has been available for several decades. This process has been used to produce substitute natural gas (SNG) from coal or biomass.

Coal-to-gas process was developed in the 1970's, in reaction to oil crisis. The process included the gasification, gas cleaning and conditioning, methanation and gas upgrading sub-processes, before injecting SNG to the natural gas grid. Industrial plants have been operated in US, and there is one coal-to-liquids plant in South Africa. The development continued around the year 2000, focusing on using biomass as feedstock to form gas (biomass-to-gas) or liquid fuels (biomass-to-liquid). New process were required, as biomass feedstock resulted in different synthesis gas compositions, and smaller plant scale compared to coal-to-gas process.

4.3.1 Chemical methanation

The Sabatier reaction, which is the basis of methanation process is described as:

$$CO_2(g) + 3H_2(g) \leftrightarrow CH_4(g) + H_2O(g) \qquad \Delta H^0_R = -206{,}2\ \text{kJ/mol} \tag{8}$$

In combination with the shift conversion:

$$CO_2(g) + H_2(g) \leftrightarrow CO(g) + H_2O(g) \qquad \Delta H^0_R = +41{,}2\ \text{kJ/mol} \tag{9}$$

With combination of (8) and (9), a formation of methane and water from carbon dioxide and hydrogen can be described as:

$$CO_2(g) + 4H_2 \leftrightarrow CH_4(g) + 2H_2O(g) \qquad \Delta H^0_R = -165{,}0\ \text{kJ/mol} \tag{10}$$

As can be seen from (9), the CO_2 in methanation process is first converted to CO. Reactions (8) and (10) and strongly exothermic. The product gas leaving the reactor contains the methane, but also steam, carbon monoxide and unconverted educts.

There are several principles for methanation process (Bajohr, et al., 2011):

2-phase systems (gaseous educts, solid catalyst):

- Fixed bed
- Fluidized bed
- Coated honeycombs

3-phase systems (gaseous educts, liquid heat carrier, solid catalyst):

- Bubble column

Fixed bed methanation

Fixed bed methanation is methanation in static bed with catalyst pellets spread in the bed. The educt gases are injected to the bed in 250 °C -300 °C temperature range, but as the methanation is strongly exothermic reaction, the bed temperature increases significantly. Therefore, thermal controls in the bed are required, to avoid formation of hot spots which would destroy the catalyst. The process is split into columns, with gas cooling, gas recycling and heat recovery in between each column.

Fluidized bed methanation

Fluidized bed methanation occurs in turbulent reactor, which consist solid catalytic particles and educt gases. The turbulence occurs due high pressure gas injection, and therefore the fluidized bed is limited to certain gas flow range and therefore constant operation. The turbulence causes abrasion, to both catalyst and reactor. The main advantages of this reactor type is good heat release and large contact area with the catalyst material in the bed.

Bubble columns

Bubble columns include a 3-phase system: gaseous educts, solid catalyst and liquid heat carrier medium. The liquid phase promotes the heat release from exothermic reactions, resulting in isothermic temperature profile and reduced abrasion.

4.3.2 Biological methanation

Besides forming methane via chemical process route, formation in a biological system is also possible by using bio-catalysts (enzymes). Methanogenic bacteria, from Archaea domain, produce these enzymes. Methanogenesis is known process in biogas processes. Two reactions are known:

Acetoclastic methanogenesis:

$$CH_3COOH(g) \leftrightarrow CH_4(g) + CO_2(g) \qquad \Delta G^0_R = -33{,}0 \text{ kJ/mol} \qquad (11)$$

And the hydrogenotrophic methanogenesis:

$$CO_2(g) + 4H_2 \leftrightarrow CH_4(g) + 2H_2O \qquad \Delta G^0_R = -135{,}0 \text{ kJ/mol} \qquad (12)$$

Equation (12) is equivalent to equation (10). Acetoclastic reaction (11) is dominant path in biogas production, but the hydrogenotrophic reaction (12) occurs in a biogas plant with mixed microbe population. Biological methanation plant can be utilized to use both pathways (integrated methanation). Integrated methanation has been demonstrated in laboratory and pilot plants. Hydrogen is used as a co-input with manure or sewage sludge. The reported hydrogen conversion efficiency has been reported to be up to 80 %, depending on the pressure and mixing intensity. The selective hydrogen utilization (selective methanation) optimizes process conditions in a bioreactor. The selective methanation can be linked to a bioreactor, but independent units are also possible which would then need a carbon source. Hydrogen conversion efficiency of 90 % at 55 °C temperature has been reported in laboratory conditions.

Biological methanation is emerging technology besides the chemical methanation process path. Advantages of biological methanation include moderate temperatures (30 °C to 60 °C), atmospheric pressure, and high tolerance against pollutants in the feed gases. Disadvantages include limitation of mass transfer between gas and liquid phases, the microbes require certain conditions, such as appropriate salinity levels, and the biological methanation is limited to low scale applications. (Lehner, et al., 2014)

4.3.3 Methanation in power-to-gas process

Due to intermittent nature of renewable energy sources, the supply of hydrogen for power-to-gas process is fluctuating. The methanation process cannot tolerate frequent start-ups, shut-downs or load changes, and therefore a hydrogen storage is needed at power-to-gas process plant. The other educt gas, carbon dioxide, needs to be stored in similar fashion. Moreover, the educt gases need to be compressed to the operational pressure of methanation process. The hydrogen from electrolysis process is available in high purity (>99,99 %), and the main impurity is oxygen. However, in regards of the other educt gas, carbon dioxide, the gas composition is a concern, beside the capture cost. The carbon dioxide can be derived be biomass or fossil fuel plants. Another possible source of carbon dioxide is from atmosphere, however the concentration is low and therefore capture cost is high. Other gases should be minimized from methanation process; steam content should be minimized, nitrogen is an inert gas in methanation process, and oxygen might affect the catalyst activity.

The carbon capture is technically possible, but not economically viable at the moment. The capture costs of different CO_2 capture processes vary from 25 to 60 €/t CO_2 (Ursúa, et al., 2012), which is much higher than the current carbon price of EU emission trading system. In case of biological methanation process, the carbon originates from untreated biogas, and therefore the biological path has much lower operational cost structure. However, the biological methanation process is not available in large scale. The heat from methanation process can be integrated in CO_2 capture process, or in other industrial processes. This lowers the costs in both cooling of the methanation process, as well as heating the carbon capture process. The site carbon capture is often in industrial processes or power plants utilizing a fossil fuel source, and therefore no new investments are necessary for steam turbines. (Lehner, et al., 2014)

5 CURRENT ENERGY PRODUCTION AND INDUSTRIAL ENERGY CONSUMPTION IN FINLAND

The energy sector in Finland is described in this chapter, with emphasis on industrial energy consumption. Energy sector in an important boundary condition for process industry, as the energy expenses have major effect on the profitability of the industry, and future energy production affects the potential for power-to-gas technology. As the energy sector changes slowly, the future energy system depends on the energy policies of today. First, the energy sector is described in general. Then the combined heat and power (CHP) production is described as it has special importance in country with cold climate and requirement for process heat in industry. Overview of industrial energy use is provided. Natural gas use and natural gas logistics are described as the markets and availability determine the use of gaseous fuels in industry. Finally, carbon dioxide (CO_2) emissions are described, with emphasis on emissions from energy use.

Finland is an industrialized country in northern Europe, with population of 5,4 million. Finland is energy-intensive country, due cold climate and energy-intensive industries, especially forest industry. Energy production causes majority of greenhouse gas emissions (80 %), including energy industry (37 % of GHG emissions), industrial energy production (14 %), domestic transporation (20 %) and other energy production (building heating, agriculture, forestry and fishery, other transportation and fuel evaporation, total 9 %) (Statistics Finland, 2013). Due cold climate, heating consumes a significant part of total energy consumption. Heating of the buildings used to be higher than industry until mid-1960’s, however since then energy-efficient heating solutions has been implemented (mainly district heating), while energy consumption of the industry has steadily grown until economic downturn in 2008. Nowadays, the consumption of industry is over half in both total final consumption (TFC) and electricity, which is the higher that in any other OECD country. (VTT, 2007)

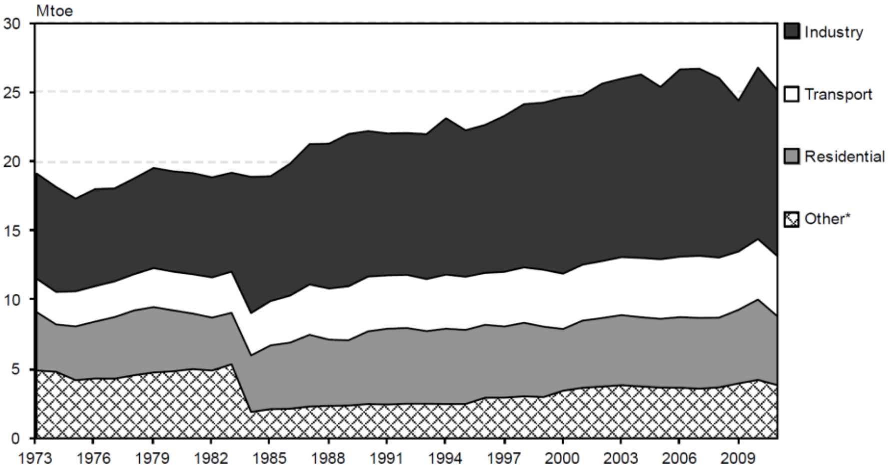

Figure 5.1: Total final consumption by sector, 1973-2011 (IEA, 2013)

Due Finland’s cold climate and energy-intensive industries, Finland has high per-capita energy use. TPES per capita was 6,5 toe in 2011, second highest of European IEA members. Total final consumption (TFC) was 25,2 Mtoe in 2011. Electricity has high share in final consumption. Industry is the largest sector in energy consumption, representing 47,5% of TFC in 2011. In Finland, industry has higher share than any other OECD country (VTT, 2007). The government projects the final consumption of industry to account 50% of TFC in 2020 and 2030. The residential sector accounted 20% of TFC in 2011. The commercial and other services sector accounted 15,3% of TFC in 2011. Transport accounted for 17,2% of TFC in 2011, which is the lowest percentage amongst IEA countries. Furthermore, the government forecasts indicate a reduction in energy use in transport sector, to 12,6% of TFC in 2030. (IEA, 2013)

Nordic countries aim to be carbon neutral by 2050. Sweden and Denmark have included this goal in their legislation. IEA have made different scenarios how the carbon neutral society could be reached. IEA lists energy efficiency, CCS and bioenergy generation as key technologies. Transport sector is especially challenging to decarbonize, from current 80 Mt/a emission level to 10 Mt/a in 2050. All Nordic countries are strong economies, with already high share of renewable energy. However, there is a large amount of energy intensive industry in the Nordics and cold climate with sparse population (IEA, 2013).

5.1 Energy production

Finland has diversified energy mix, with high shares of biofuels and nuclear energy. Domestic primary energy sources include hydropower and different biomasses (forest industry side streams, wood chips, forest residues and peat). Imported primary energy sources in Finland are fossil fuels (oil, coal and natural gas), uranium for nuclear power production and electricity imports. As Finland has no fossil fuel sources, little uranium mining and no uranium enrichment facility the degree of self-sufficiency in energy is low. Approximately 70% of TPES are imported, mostly from Russia. Crude oil constitutes a large share of the energy imports. Russia is the only source of natural gas, which is delivered via one double pipeline (VTT, 2007). Co-production has major share in energy production, and most heat consumed is produced in combined heat and power (CHP) plants. Biomass has large share in energy mix, and its availability is linked with the production of forest industry. (VTT, 1999)

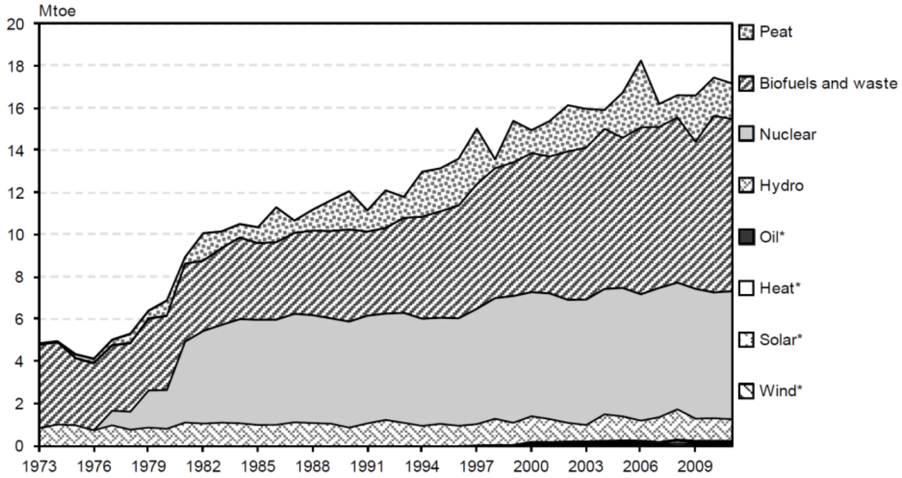

Figure 5.2: Energy production by source, 1973-2011 (IEA, 2013)

Figure 5.2 represents the Finnish Total primary energy supply (TPES) from 1973 to 2011. TPES in Finland is 34,7 Mtoe. TPES has fluctuated in recent years, from 32,2 Mtoe in 2000 to 37,3 in

2006. On average, the TPES is growing 0,7 per annum, and is projected to reach 38,2 Mtoe in 2030. Oil represents 26,4% of TPES, which is relatively low compared to other OECD countries. Biofuels accounted 23,3% of TPES, and is projected to remain relatively constant in the future. Nuclear power represents 17,4% of TPES. However, two new nuclear plants (Olkiluoto 3, Fennovoima Hanhikivi 1) are under construction or planned at the time of the writing (2014), which will increase the share of nuclear to 38,4% of TPES by 2030 (IEA, 2013). Regarding the fossil resources in Nordic countries, oil and gas resources are abundant in the North Sea (in territorial waters of Denmark and Norway). In future scenarios, energy efficiency, increase of biomass and wind power production and deployment of CCS are critical in reaching the emission reduction targets.

5.1.1 Electricity production

Finnish electricity market was deregulated in 1995, and was integrated into Nord Pool electricity exchange. The Nordic countries have different electricity generating profiles, which can balance each other; Norway uses almost entirely hydropower to generate its electricity, Sweden produces over 90% of its electricity with hydropower and nuclear power, while Denmark has traditionally generated most of its electricity with thermal power. Nowadays Denmark and Sweden generate more and more power by wind turbines, and common electricity market (with more than half of all power is generated by hydropower) help to balance the periods of low wind. In Finland, electricity generation is highly diversified, as can be seen from the table 5.1. (VTT, 2007). The total consumption of electricity was 87,7 TWh in 2010. 12% of the consumed electricity was imported from Sweden, Norway and Russia. The total electricity production capacity in Finland is approximately 17,0 GW. (Statistics Finland, 2012)

Table 5.1: Electricity acquisition 2011 (Statistics Finland, 2012)

Production and total consumption of electricity 2011 [GWh]	
Nuclear power	22266
Oil	380
Hard coal	9134
Natural Gas	9202
Other fossil	616
Total fossil	*19333*
Peat	5054
Hydro power	12278
Wind power	481
Black liquor and other concentrated liquors	5067
Other wood fuels	5073
Other renewables	418
Renewable fuels total	*23318*
Other energy sources	421
Total production	*70390*
Net imports	13851
Total acquisition	*84241*

Domestic electricity production in is presented in the table 5.1. Nuclear power was the largest source of electricity in 2011 (33% of domestic production). Other major sources include hydro power (25%), black liquor and other wood fuels (14%), hard coal (10%), natural gas (10%) and peat (5%). Oil is used mainly for peak power production or regulating power (1%). Wind power does not have large share in total production (1%) as of 2011. (Statistics Finland, 2012) Electricity consumption per capita is the second highest in IEA countries after Norway, standing at 26,0 MWh per capita, compared to IEA average of 10,3 MWh per capita. (IEA, 2013)

The electricity generation capacity is changing gradually: hydropower, wood fuels, peat, municipal waste fuels and wind power have increased in past decade. CHP production has increased in 1983 to 2001, and currently approximately one third of electricity is produced in CHP plants. (VTT, 2007)

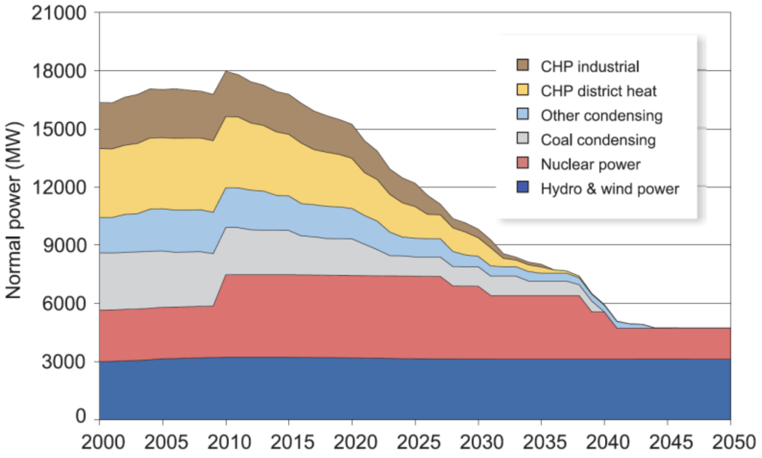

Figure 5.3: Estimated shut-down schedule of the present electricity generating capacity (IEA, 2013)[1]

There is a link between economic growth and energy use. In Finland, the energy use has not grown as quickly as energy use in recent years, but the energy use increased steadily until the economic downturn in 2008. When the economy starts to grow again so will the energy consumption, if no significant measures to energy efficiency will be implemented. No large scale electricity generating plants have been started in Finland since the deregulation of the electricity market, due poor competitiveness of Finnish condensing power plants in years of high precipitation. Furthermore, the operation of Olkiluoto 3 nuclear power plant, that was originally intended to be opened in 2009, has been delayed until at least 2018 (World Nuclear News, 2014). This has resulted in growing electricity imports, particularly from Russia. Condensing power was used as base load power until deregulation, but its decreased competitiveness has led to low utilization rates in recent years. However, if Finnish condensing power capacity were to be shut down, the security of Finnish electricity supply would be threatened during low precipitation years. The shut-down schedule of Finnish power plants is represented in figure 5.3. As can be seen from figure 5.3, besides nuclear and hydro power plants, all Finnish power plants will expire by 2050. Therefore, complete structural change in power generation is required in any case. Due climate

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[1] It should be noted that in this figure the Olkiluoto 3 nuclear power plant is depicted to start production in 2010, which has not realized as of 2015. Also, this figure does not include the planned Fennovoima Hanhikivi 1 power plant.

reasons, future Finnish energy system is likely to be based mostly on non-emission energy sources (renewable and nuclear energy).

Nordic countries have one common electricity market (Nordpool). Nordic countries already have high shares of renewable energy, with large differences in energy sectors of different countries. Sweden relies on biomass, hydro and nuclear power. Finland have biomass, hydro (to lesser extent than Sweden), nuclear and fossil thermal power. Denmark currently uses large amount of coal in thermal power generation, but has high share of wind and biomass as well, and ambitious targets to increase renewable power in the future. Norway produces most of its electricity by hydropower. Iceland generates most of its electricity with hydropower, with some geothermal power production.

5.2 Combined heat and power (CHP) production

The total heat consumption in Finland was 152 TWh in 2011. The consumed heat is produced in industrial plants, to provide heat for the processes, or in CHP plants, for district heating which includes heating buildings, service water, and industrial processes. Figure 5.4 represents the fuel use by production mode in electricity and heat production in 2012. 76% of fuel used in thermal power plants is produced in CHP plants (Statistics Finland, 2012), which is efficient solution for Finnish conditions, as the consumption of both heat and power are higher in the winter months.

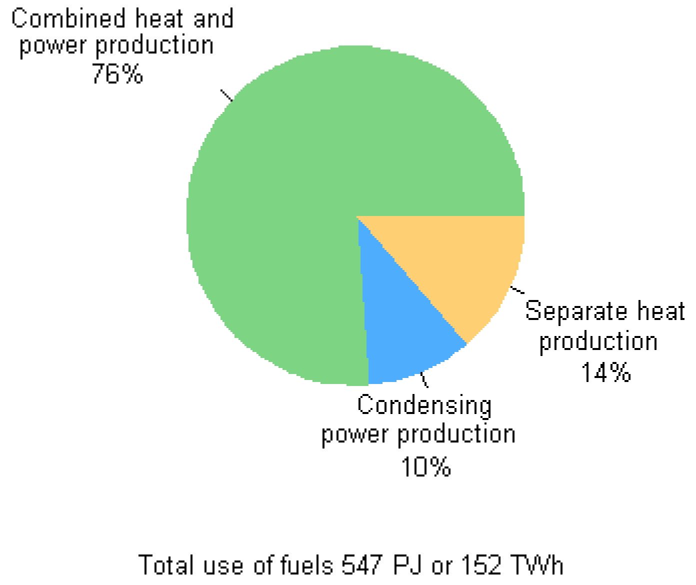


Figure 5.4: Fuel use by production mode in electricity and heat production 2012 (Statistics Finland, 2012)

Due the geographical location of Finland, energy is needed for building heating most of the year. District heating is an efficient solution to provide heat in winter months in densely populated areas. District heating was first started in 1940 in Helsinki Olympic village, and the network started to expand through the capital area in 1950's. The district heating has been applied in smaller towns since then, which has also increased the share of electricity produced by CHP plants. Nowadays 46% of Finland's total heat demand is delivered by district heating (in 2010), with total quatity of 31,2 TWh of heat (2011), there are approximately 8000 km of district heating pipelines, and 2,73 million Finns have their homes heated by district heating. Approximately three fourths of heat used in district heating is produced in CHP plants. Heat produced in the power plant is collected by heating surfaces in the boiler. In CHP power plants, approximately 25-30% is converted into electricity and 55-60% is applied as heat, while in condensing power plants approximately 40% is converted into electricity. (VTT, 1999; Finnish Energy Industries, 2013)

Figure 5.5 shows how the fuel inputs have changed since 1976. In the 1970's, almost all of the heat provided for district heating were supplied by coal and oil. There was a significant decrease in oil consumption for district heating in late 1970's and early 1980's, and oil has only marginal share in district heating production nowadays. The share of peat has increased from late 1970's until 1984, natural gas increased its share in heating market since first half of 1980's and biomass has increased its share since mid-1990. The share of coal has decreased in past 20 years, but its share remains significant nonetheless. (Finnish Energy Industries, 2013)

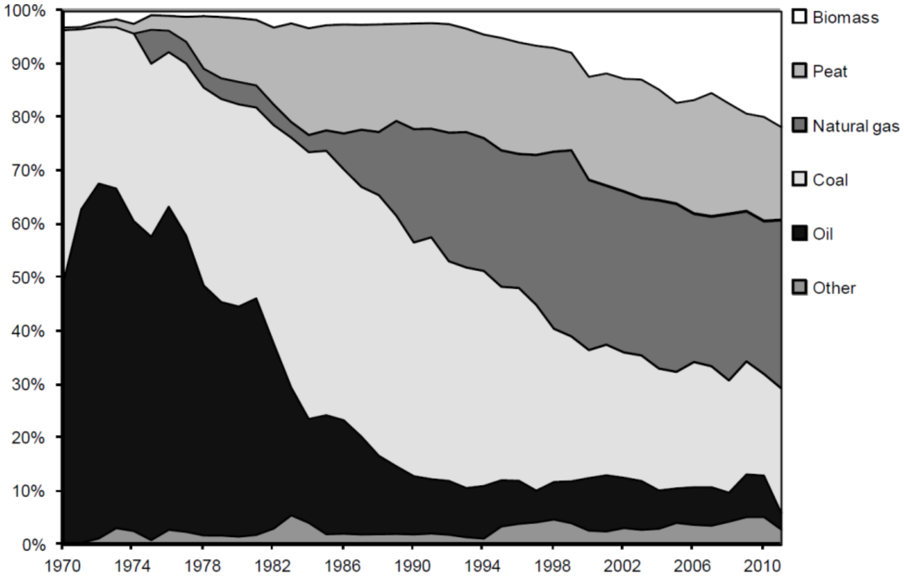


Figure 5.5: Share of energy inputs for district heating and related CHP, 1976-2011 (Finnish Energy Industries, 2013)

5.3 Industrial energy consumption

The industry is the largest energy consumer in Finland. It consumes approximately half of both TFC and electricity consumption. Figure 5.6 represents the consumption of electricity in manufacturing industry. Forest industry is the largest energy consumer, with 55% of electricity consumption. Forest, chemical and metal industries together consume 91% of electricity in industry. (Statistics Finland, 2012)

Current economic downturn and structural change of Finnish industry might have a profound impact to the electricity consumption of the industry. Current major industrial sectors might shrink considerably, and on the other hand new industries might grow significantly.

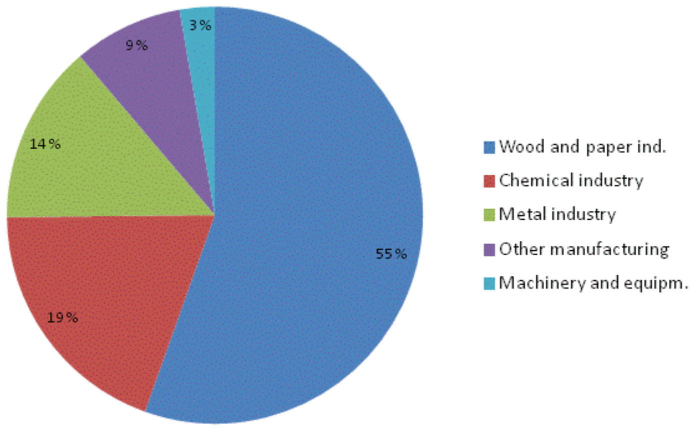


Figure 5.6: Total electricity consumption in manufacturing (Statistics Finland, 2012)

Electricity consumption in industry has grown compared to other forms of energy, as shown in figure 5.7. This trend is expected to continue in the future. At the same time, energy consumption per unit produced has reduced. Automation, switching from direct fuel use to electricity use, recycling and material-efficient processes have increased the consumption of electricity. New machinery consume less power per unit produced than the old, replaced machines. Honkapuro et al. (Honkapuro, 2009) have estimated that there is potential for 21% electricity efficiency improvements by 2050 in Finnish industry. This potential includes optimization of processes, especially adopting new process technology, and improving energy efficiency of pumps, blowers and pneumatic appliances. (Finnish Energy Industries, 2013, pp. 45-46)

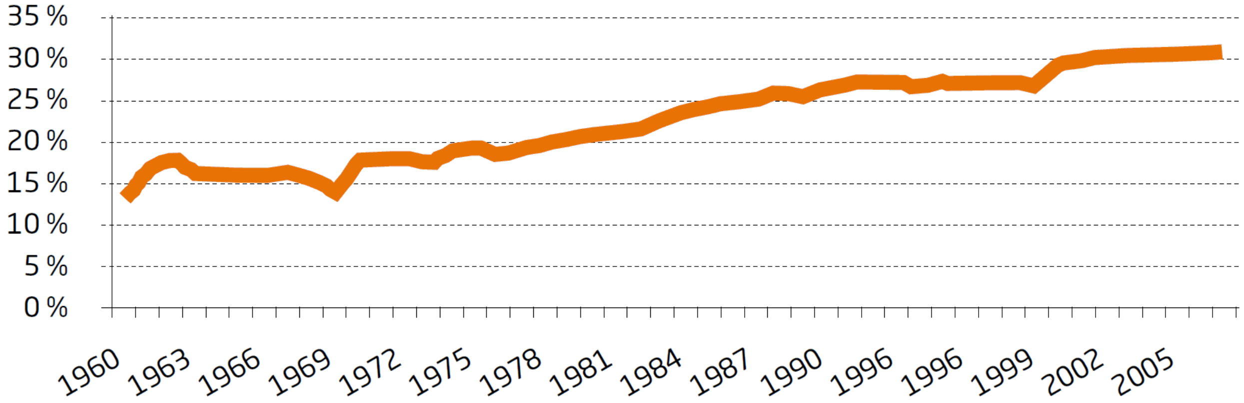


Figure 5.7: Share of electricity of the total energy consumption in industry (Finnish Energy Industries, 2013)

One third of total electricity consumption and 55% of industrial electricity consumption is consumed by forest industry. However, forest industry is undergoing a radical structural change, due changes in consumption patterns, increased competition, especially in developing markets, and increased domestic costs. These trends have resulted in moving the production of traditional forest industry production closer to developing markets. However, forest industry develops new products, such as biofuels, materials, chemicals and functional wood and fiber products. Due these uncertainties, the electricity consumption estimates have quite a high variability regarding consumption of forest industry. National Climate and Energy Strategy (2013) basic scenario estimates the electricity consumption to grow slowly until 2030, as can be seen on table 5.2. The energy consumption in metal and chemical industries are expected to grow slowly. However, also in these industries both consumption and production are expected to move closer to developing markets. National Climate and Energy Strategy expects the electricity consumption in these sectors to grow slowly until 2030. (Finnish Energy Industries, 2013; Työ- ja elinkeinoministeriö, 2013)

Table 5.2: Electricity consumption in industry per sector, according to National Climate and Energy Strategy basic scenario (Työ- ja elinkeinoministeriö, 2013)

	2010	2020	2025	2030
Pulp and paper production	20,3	21,0	21,8	22,6
Metal industry	5,4	6,9	7,1	7,0
Oil refining	1,3	1,6	1,7	1,7
Chemical industry, rubber and plastics production	5,4	6,7	7,0	7,2
Other industry and construction	9,0	10,6	11,4	12,2
Total	*41*	*47*	*49*	*51*

Heat used in industrial processes is often produced in the industrial plant. Often these same processes consume so much electricity, that it is economical to produce electricity at the same time, which has made these energy-intensive industries also power producers. (VTT, 1999) Forest industry produces more than 65% of its produced power by side stream biomass fuels from its own processes. These side streams include black liquor, barks and other waste wood. Ore and steel production produces secondary heat at high temperature combustible gases, which can be used in coke ovens batteries, Cowper stoves, slab reheating furnaces and steam boilers. The waste heat and combustible gases can be used in electricity production, and usually metal processing plant produces approximately 60% of its own electricity consumption. In chemical industry processes combustible gases are produced as side streams, such as hydrogen. Production of sulfuric acid makes high-pressure steam, which can be used to generate electricity. (VTT, 1999)

Industry accounts for approximately 35 % of TPES in Nordic countries. To meet the ambitious CO2 reduction targets, all industry sectors are required to decrease their emissions. Less than one quarter of industrial CO2 emissions is met by fossil fuels in 2050, and CCS has a critical role in eliminating the industrial CO2 emissions. Natural gas is used in chemical and petrochemical industry, iron and steelmaking industry and pulp and paper industry in the Nordic countries (IEA, 2013).

In cement sector, alternative fuels (waste fuels) and CCS have key roles in reducing the carbon emissions. Iron and steel industry is decarbonized by electrification and energy efficiency improvement, while electrolysis and hydrogen have only marginal roles. Chemical and petrochemical industries rely on bio-based feedstock, CCS and energy efficiency investments. Pulp and paper industry rely more to bio-energy, and CCS is deployed to some extent. Energy efficiency is improved in aluminum industry. These improvements require in total between 83 to 137 billion USD by 2050 (IEA, 2013).

5.4 Overview of natural gas use

The use of natural gas as fuel began in the US in 1930's. It became a major energy source in US in the 50's and in Europe in the beginning of 60's. Nowadays the natural gas provides approximately one fifth of world's primary energy supply (21,3% in 2012) (IEA, 2014). In Finland, natural gas provides 8% of TPES (Statistics Finland, 2013).

Natural gas used in Finland in 2014 originates from Russia. The use of natural gas is linked with logistics; natural gas is used commonly in industries which have an access to natural gas pipeline network, but rarely outside it.

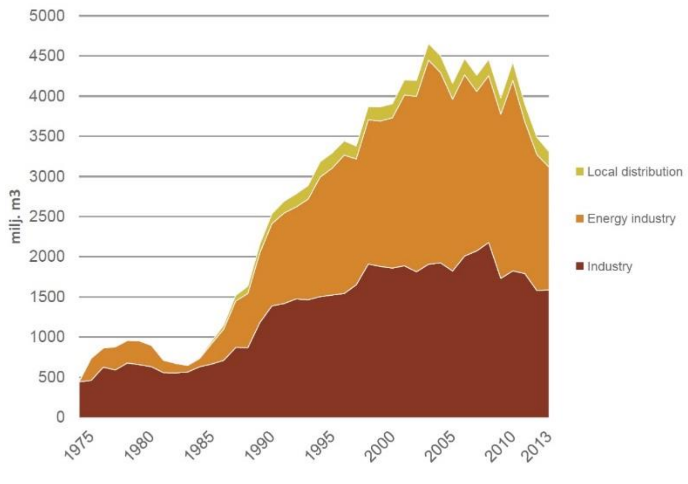


Figure 5.8: Natural gas consumption in Finland 1974-2013 (Finnish Gas Association)

Figure 5.8 illustrates the natural gas consumption from 1974 to 2013. The consumption has grown until 2000, and stayed approximately constant from 2000 to 2010. Year to year variation in this period occurred due varying heat requirement in the winters. After 2010, natural gas consumption has reduced in both energy production and industry.

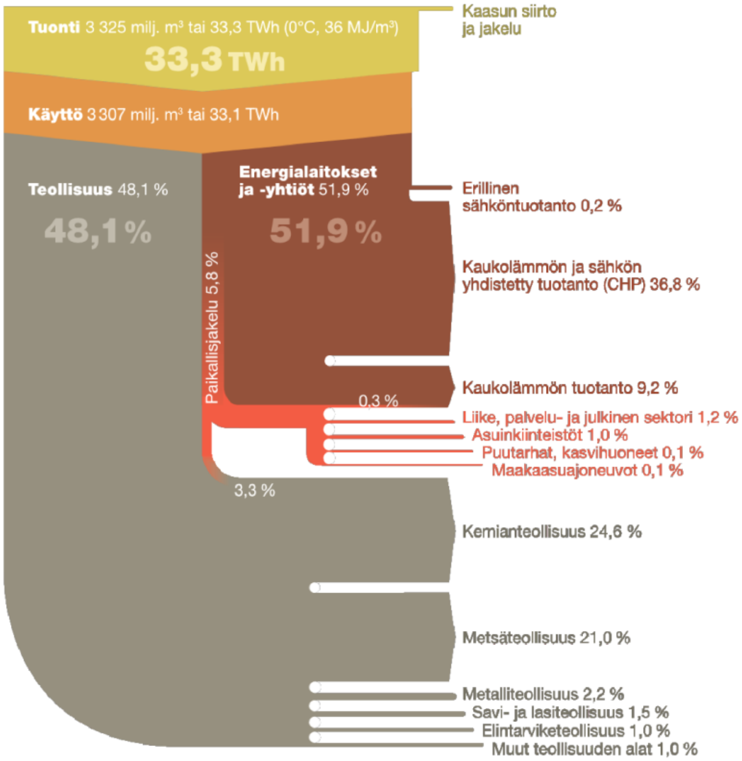


Figure 5.9: Natural Gas consumption by sector in 2013 (Finnish Gas Association)

Figure 5.9 shows the natural gas consumption by sector. 51,9% of the natural gas was consumed by energy industry in 2013. Other industries consumed 48,1 % of all natural gas. Chemical industry was the largest industrial consumer, consuming 24,6 %. Majority of chemical industry consumption (761 million m3) was consumed by petrochemical industry (500 Mm3) (NESTE). Forest industry was the second largest industrial sector, consuming 21 % of all natural gas. Metal (2,2 %), clay and glass (1,5 %) and food (1,0 %) industries had small but significant shares of consumption.

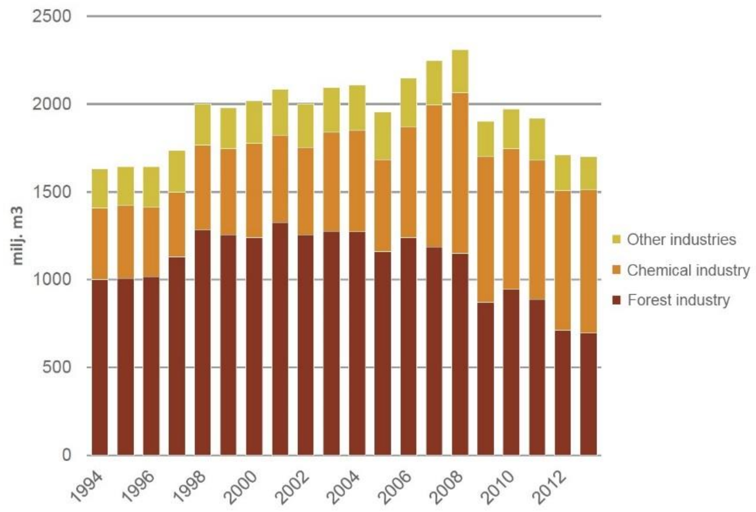


Figure 5.10: Natural gas consumption in chemical and forest industries 1994-2013 (Finnish Gas Association)

Figure 5.10 illustrates the consumption of natural gas consumption in major industry sectors from 1994 to 2013. The consumption in forest industry peaked in 2001, and has been in decline since 2006. In chemical industry, the consumption increased until 2008, and has been stable since. The consumption in other industries show little variation since 1994.

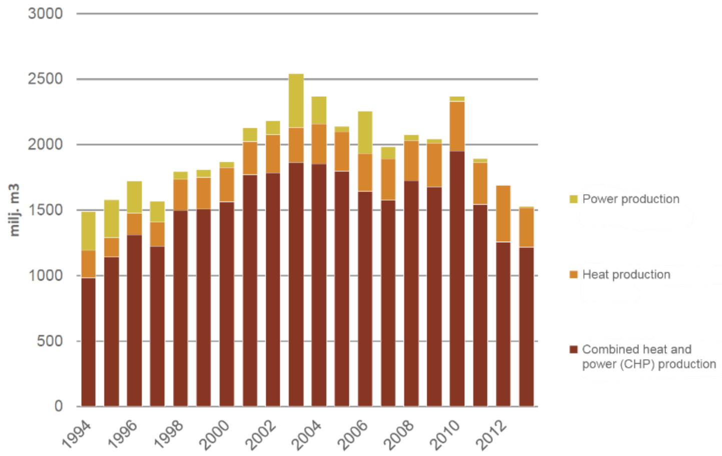


Figure 5.11: Natural gas in power and heat production 1994-2013 (Finnish Gas Association)

Figure 5.11 illustrates the natural gas consumption in power and heat production. Majority of natural gas in energy production is consumed in CHP. The use of natural gas in CHP production peaked in 2010, at approximately 2000 m3, and has since declined to approximately 1200 m3. Natural gas used to have a share in dedicated power production, but nowadays its amount is marginal. Natural gas consumption in dedicated heat production has varied from 200 to 400 m3.

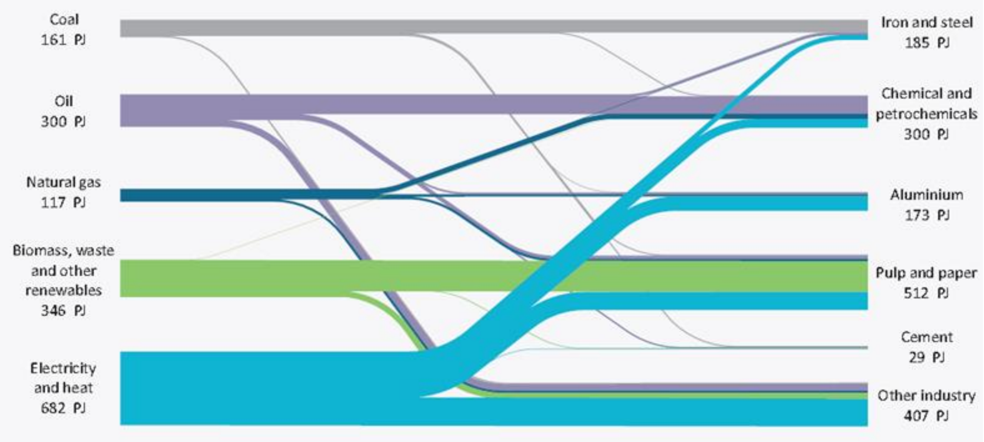


Figure 5.12: Energy flows by fuel in Nordic countries in 2010 (IEA)

Figure 5.12 represents the fuel use in different sectors in the Nordic industry. Fossil fuels (coal, oil, natural gas) represent large share of fuels, however the total share of biomass, waste and other renewables is high. Chemical, pulp and paper, and metal industries are the main energy consumers. Natural gas is used in chemical, pulp and paper, and to lesser extent, metal industry.

5.5 Natural gas logistics: pipeline network and LNG terminals

Transport of gaseous fuels require specific infrastructure. It is possible to transport gaseous fuels via pipelines in gaseous form, or by tanker trucks, rail carts or marine traffic in liquefied form. As there is no large scale LNG terminals in Finland as of 2014, natural gas is delivered by pipelines at the moment and by tanker trucks. However, to diversify the supply of natural gas the Finnish government and energy companies have ambitious plans with LNG terminals. There are seven planned LNG terminals: Finngulf in Porvoo or Inkoo, Manga in Tornio, Pori, Turku, KotkaHamina harbor, Rauma and Salo. Additionally, a pipeline connection to Estonia (Balticconnector) is planned, which would connect the Finnish and Baltic gas markets. The logistics affect the price and availability of the gas; the price of the natural gas is significantly higher where pipeline network do not reach and therefore the use of natural gas is not common in those areas. Respectively, there will likely to be new gas consumers in the areas where the LNG terminals are planned.

As of 2014, all natural gas is imported from Russia via double pipeline, crossing the border near Imatra. The distribution and sales of natural gas to consumers is done by one operator, Gasum. Gasum has made a long-term gas supply contract with Russia provider Gazprom Export to import Russian natural gas until 2026. The contract allows annual 6 billion m^3 imports. (IEA, 2013) Until 2014, Gasum was owned by four entities: Fortum (31% of shares), Gazprom (25%), the Finnish state (24%) and E.ON (20%). However, on 3.11.2014, the government of Finland decided to acquire the shares of Fortum and E.ON, giving it 75% of shares. According to Minister of Ownership Steering Sirpa Paatero the goal of the acquisition was to “strengthen the security of supply and provide competitive priced energy to Finnish industry”, as well as improve the gas markets and infrastructure for natural and biogas, and LNG. The total price of Fortum’s and E.ON’s shares was 510 million euros. (Government Communications Department, 2014) Gasum intends to diversify its supply, by increasing biogas production, and LNG terminals. The delivery of natural gas and LNG logistics are discussed in this chapter. Biogas is not on focus in this thesis.

5.5.1 Pipeline network

Main method to transport natural gas is via pipeline network. All natural gas currently (2014) consumed in Finland is delivered from Russia, via double pipeline. High-pressure transmission pipelines deliver the gas to Finland and to different towns. Finland has 1287 km of high-pressure gas pipelines. The high-pressure pipelines are constructed of steel, with polyethene plastic surfacing. The pressure in the pipeline is 30-54 bars, however nowadays some pipelines are constructed for 80 bar pressure. Figure 5.13 illustrates the transmission pipelines in Finland.

Figure 5.13: Current natural gas pipeline network (Source: Gasum)

Along the pipelines, there is a valve station every 8-32 km, in total 125 stations. Valve stations allow cutting the gas flow and emptying the pipeline by blowing out the gas in case of malfunction. Usually valve stations have link stations in vicinity, 123 in total. They allow to monitor the transmission. Transmission is centrally monitored from Kouvola. Compressor stations allow the adjustment of pressure and increasing the transmission capacity of the network. There are nine turbocompressors, with total axial power of 64 MW, in the three compressor stations: four in Imatra, three in Kouvola and two in Mäntsälä. The delivery from high-pressure transmission pipeline to customer is done via pressure reduction stations. There are 134 pressure reduction stations in pipeline network, where consumption is measured. The delivery from pressure reduction stations to customers are done via small polyethylene pipes, with 4 or 8 bar pressure, except in some areas in Helsinki, where there are steel pipes with 1,025 bar pressure. Inside the buildings the gas is delivered by steel pipes. (Gasum, ei pvm)

5.5.2 LNG terminals

Gasum; Kilpilahti, Porvoo

Kilpilahti LNG production facility is existing LNG (and liquefied biogas, LBG) land terminal in Kilpilahti, Porvoo, completed in 2010. Gasum has natural gas liquefaction plant in Kilpilahti, where natural gas is taken from the pipeline, liquefied, loaded into LNG trucks and carried to LNG consumers by Skangass (or by customer’s own vehicles). Capacity of the plant is 20 000 tons of LNG per annum, and there are three 700 m^3 LNG storage tanks (total 0,01 TWh). The site is fully automated and controlled from Gasum’s control room in Kouvola. (Gasum, ei pvm)

Gasum; Finngulf, Inkoo/Porvoo

Finngulf is a planned LNG terminal. Finngulf LNG terminal will be implemented by Finnish gas distributor Gasum. Finngulf will be located either in Inkoo or Tolkkinen, Porvoo. Purpose of the Finngulf LNG terminal is to diversify the natural gas supply, reduce Finland’s dependence of Russian gas, and open up the gas market for competition. Finngulf would provide LNG also to Estonian markets. The terminal will have a LNG vaporization process, with capacity of 5 GW (8 GW if Balticconnector gas pipeline will be constructed). The terminal will have a storage capacity of two 165 000 m^3 LNG storage tanks, and 150 000 m^3 in floating storage vessel, in total approximately 3 TWh (Gasum, Sito, 2014). The terminal has capacity to deliver 10-20 TWh of natural gas per annum, equivalent to 25-50% of annual natural gas consumption in Finland. (Mattila, 2013), and can feed 5-6 GW into pipeline network. The terminal will serve Estonian natural gas markets also. The site of the LNG terminal will be in Joddböle, Inkoo, 4 km from the center of Inkoo, and 20 km from connection to natural gas pipeline network in Siuntio. The planned submarine Balticconnector pipeline will be connected in the terminal area if both projects are realized. Another option is to locate the LNG terminal in Tolkkinen, Porvoo, 4 km from current Gasum LNG terminal in Kilpilahti. The schedule for the completion of the project is late 2018. (Kallio) (Mattila, 2013) The total cost of the Finngulf LNG terminal is expected to be 380 million euros. (Jännes, 2013) Gasum intends to make investment decision in 2014 (Kallio). Finngulf-project is eligible for EU support. EU will support one LNG terminal in Gulf of Finland from its Projects of Common Interest (PCI) fund to improve European energy infrastructure. EU has started Baltic Energy Market Interconnection Plan (BEMIP) in 2008, which aims to connect Finnish and Baltic natural gas pipeline networks to European gas pipeline network. The projects that help to achieve this goal, such as Finngulf LNG terminal and Balticconnector natural gas pipeline, will be supported from EU's PCI fund. (Liukko & Grönlund, 2014) The terminal does not fall under the remit of the national aid programme. (Ministry of Employment and Economics, 2014)

Gasum; Balticconnector submarine pipeline

Part of the Finngulf LNG terminal project plan is to connect Finland and Estonia via Balticconnector pipeline under the Gulf of Finland. The planned pipeline will have maximum capacity of 3 GW. Estonian government and Estonian gas company EG Võrguteenus prefers to construct LNG terminal in Paldinski, Estonia, and EU will not support two terminals from the PCI fund. Negotiations between Estonian and Finnish governments, Gasum and EG Võrguteenus are ongoing. (Piit, 2014) Compromize to two LNG terminals is to connect Finnish and Baltic natural gas pipeline networks via submarine Balticconnector pipeline. The aim is to start operating the pipeline in 2016. (Jännes, 2013) According to counselor of industry Herkko Piit from the Finnish Ministry of Employment and Economics, the Balticconnector would receive EU's maximum Projects of Common Interest support, 75% of the total project cost of 200 million euros. (Piit, 2014)

Manga

Manga is an LNG terminal planned in Tornio by Outokumpu, SSAB, EPV Energy, and Gasum. The purpose of the project is to deliver LNG to industrial processes, energy production and maritime shipping in the area. Manga will not have connection to natural gas pipeline network.

Project development company Manga LNG Oy has been founded to implement the project. Wärtsilä is chosen as the contractor. The terminal will have 50 000 m3 (0,3 TWh) storage tank for LNG, pipeline connection to Röyttä industrial site, which is the location of Outokumpu steel production plant, vaporizer unit, and LNG truck loading station. The construction is intended to take place in 2014-2018, and the deliveries will start in 2018. For deliveries, a joint venture shipping company is planned, with a vessel with capacity of 18 500 m^3. (Manga LNG) The total cost of the terminal is estimated to be 100 million euros. The Finnish Ministry of Employment and the Economy has approved to support the project with 33 million euros. (Työ- ja elinkeinoministeriö, 2014)

Gasum; Tahkoluoto, Pori

On September 2014, Skangass (a subsidiary of Gasum) has made an investment decision to build an LNG terminal in Tahkoluoto, Pori. The construction will take place between 2014 and 2016, and it will be the first operational LNG terminal in Finland. The terminal will serve industry in the southwest Finland, the area where the natural gas pipeline network does not reach, as well as marine and road traffic. A 12 km pipeline to local industrial area will be constructed, however the terminal will not have a connection to natural gas pipeline network. The terminal will have 30 000 m^3 (0,2 TWh) LNG storage. (Gasum, 2014) The terminal will receive 23 million euros investment support from the Ministry of Employment and Economics. (Ministry of Employment and Economics, 2014)

Gasum; Pansio, Turku

Gasum is planning to construct an LNG terminal in Pansio port of Turku. The terminal would have 30 000 m^3 (0,2 TWh) LNG storage tank and a vaporizer plant. The terminal would serve local industries with LNG, as well as marine traffic. A pipeline to local industrial area is planned. The total investment cost is estimated 60 million euros. (Gasum) Due to complaints on plans to built

Pansio LNG terminal, Gasum made an investment decision on Tahkoluoto LNG terminal in Pori. This terminal will serve some of the potential customers in Southwest Finland, and therefore the investment decision on Tahkoluoto terminal might delay the construction of Pansio terminal. Pansio terminal is not off the table however, as marine traffic in Turku port will require source of LNG in the future, and delivery by trucks from Pori will be less cost efficient. According to local newspaper Aamuset article in March 2014, the construction will start earliest in 2015. (Aamuset-kaupunkimedia, 2014)

Haminan Energia; KotkaHamina harbor, Hamina

Haminan Energia is planning to construct LNG terminal in the oil- and gas import terminal in KotkaHamina harbor. The planned terminal would include 30 000 m3 (0,2 TWh) LNG storage tank, and pipeline connection to local industries as well as natural gas pipeline network. The investment decision is expected in 2014, and construction will take place in 2015-2018. The total investment cost is estimated 100 million euros. Haminan Energia’s plan to construct 50 MW CHP plant in the harbor area is related to the LNG project. (Toikka, 2013)

AGA; Rauma

AGA is planning to build an LNG terminal in Rauma harbor. The planned terminal will have 8 LNG storage tanks, with 10 000 m3 (0,06 TWh) combined capacity. The purpose of the terminal is to deliver LNG to local industries and marine traffic. No pipelines are planned; the gas will be delivered to consumers by trucks. (AGA) The construction is expected to be finished in 2017. The Finnish Ministry of Employment and the Economy has granted 8 million euros support for AGA Rauma LNG terminal.

LNG Finland; Salo

In November 2011, newspaper Salon Seudun Sanomat reported that LNG Finland intends to build a floating LNG terminal at sea near Salo. The terminal will provide LNG to marine traffic. According to Timo Kiviaho from LNG Finland, the storage capacity will be "several hundred to several thousand of cubic meters". (Salon Seudun Sanomat, 2013) As of November 2014, no new information about the cost, capacity or technical details is publicly available about the project. However, the Finnish Ministry of Employment and the Economy will make a decision about investment support for LNG Finland's LNG terminal in 2014. (Ministry of Employment and Economics, 2014)

5.6 CO_2 emissions

Finland has ratified the Kyoto protocol, an international commitment to limit the greenhouse gas emissions to 1990 levels by 2012. European Union has set an additional "20-20-20" target, which mandates the Union to reduce greenhouse gas (GHG) emissions by 20%, generate 20% of total energy demand by renewable sources and improve energy efficiency by 20%.

In 2005, EU introduced its emissions trading scheme (EU-ETS) as a method to reduce climate emissions. EU-ETS is the main mechanism for EU to meet its Kyoto and 20-20-20 targets. In the trading scheme, allowances to emit one ton of CO2, known as carbon credits, are sold and purchased by companies within EU. Goal of the scheme is to reduce emissions in market-based manner. All sectors in this thesis, including thermal power production at installations over 20 MW, metal industry, basic inorganic chemicals, fertilizers/ammonia and cement production have been part of the EU-ETS since its first phase (2005-2007) (VTT, 2007; IEA, 2013). EU-ETS includes on average 55 % of all CO_2 emissions in the EU. This share is higher in Finland (62 %), due to cold climate and energy-intensive industries. Introduction of EU-ETS has affected the Finnish energy industry; the competitiveness of Finnish condensing power plants is poor in the years of high precipitation, and extra price on carbon has further weakened the competitiveness of these power plants. (VTT, 2007)

Finland is on track to meet its Kyoto target. Emissions, excluding land use, land use change and forestry (LULUCF), were 66,34 million tons of CO_2-equivalent (Mt CO_2-eq.), down from 70,36 Mt CO2-eq in 1990. If LULUCF is included, the reduction is even more favorable; 25,78 Mt CO_2-eq. (2009) from 55,32 Mt CO_2-eq. (1990). This is due forest growth in Finland is more rapid than its harvest. (Statistics Finland, 2009) However, it can be argued that this progress is due mitigation measures and economic downturn, rather than successful mitigation measures alone. Industry has reduced its emissions as well. The emissions from energy sector were 53,11 Mt CO_2-eq. in 2009, down from 54,48 Mt CO_2-eq in 1990. Industrial processes increased slightly, from 5,07 Mt CO_2-eq (1990) to 5,24 Mt CO_2 eq (2009), however the increase was higher by 2008, at 7,07 Mt CO_2-eq. (Statistics Finland, 2009) Measured by fuel, 43,9 % of emissions originated from oil combustion, 41,2 % from coal, 13,8 % from gas and 1,1 % from other fuels. (Statistics Finland, 2014)

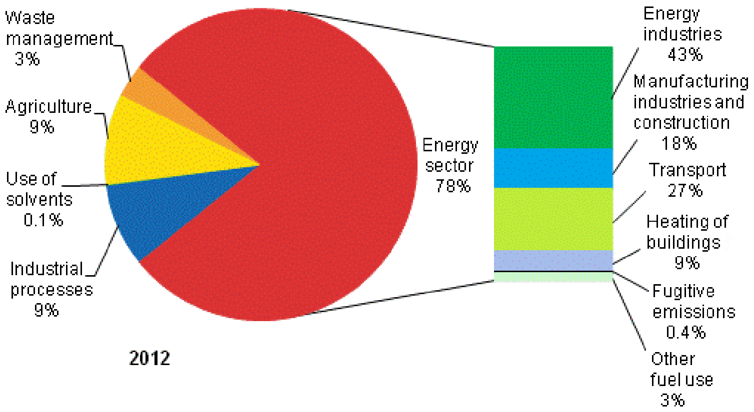


Figure 5.14: Greenhouse gases by sector in Finland in 2012 (Statistics Finland, 2012)

Figure 5.14 represents the GHG emissions by sector in Finland. Energy production is the largest emitting sector of GHGs, accounting 78 % of all GHG emissions in 2012. Electricity generation accounted 43 % of the emissions from energy acquisition. Second largest sector was transport, with 27 % share of energy acquisition. Manufacturing industries and construction accounted for 18 %, and heating 9 %. Industrial processes not related to energy production accounted for 9 % of all GHG emissions in Finland, agriculture accounted for 9 % and waste management 3 %.

Emissions from electricity generation per kilowatt hour is low in Finland (199 g/CO_2/kWh in 2011) compared to average of IEA countries (418 g/kWh). (IEA, 2013) 57 % of the greenhouse gas emissions in the Nordic countries originate from energy production. Finland and Denmark have the highest emissions from energy production, due the large share of fossil fuels in their energy production. However, the Nordic countries have ambitious plans to decrease emissions from energy production. Sweden, Denmark and Norway aim for 100 % reduction, Finland 80 % reduction and Iceland 50-70 % reduction. These reductions are mainly carried out by increasing share of wind and biomass in electricity production (IEA, 2013).

6 POTENTIAL INDUSTRIAL APPLICATIONS FOR ELECTROLYSIS GASES IN FINNISH INDUSTRY

This section introduces four industrial sectors (steel, oil refining, cement and glass sectors), where there is potential for power-to-gas generated SNG, and 1-2 example process cases in each sector. These processes were chosen for their high total emissions, the high total emissions in their industry sector (see figure 1.2), or high share of natural gas in their energy use profile. Furthermore, these sectors are known to be challenging for process electrification. First, an overview of the energy and fuel use in each industry sector is provided.

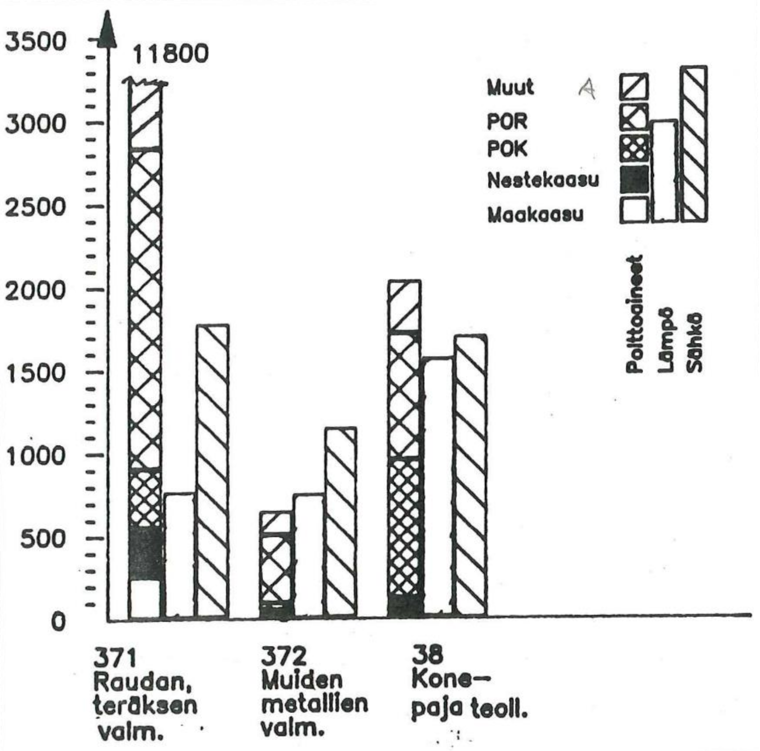
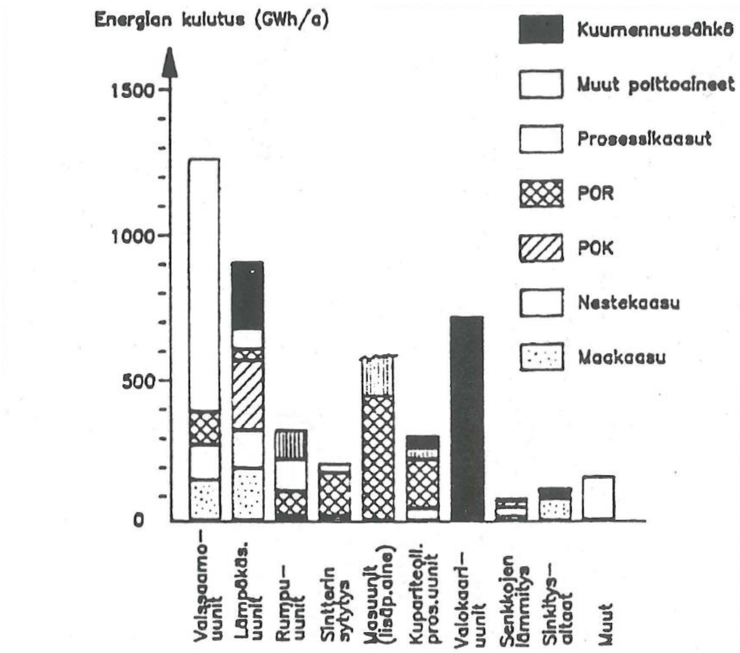


Figure 6.1: Metal industry energy consumption, by industry branch [GWh/a] (Malin, 1987)

Figure 6.1 illustrates the energy consumption in metal industry by industry branch in Finland in 1987. The iron and steelmaking are by far the most energy consuming sectors, consuming 11 800

GWh fuels/a. Next is the machine engineering, with fuel consumption of 2 100 GWh/a, and manufacturing of other metals consumes approximately 650 GWh fuels/a.



	Valssaamo-uunit	Lämpökäs. uunit	Rumpu-uunit	Sintterin sytytys	Masuunit (lisäp.aine)	Kupariteoll. pros.uunit	Valokaari-uunit	Senkkojen lämmitys	Sinkitys-altaat	Muut
Yksiköitä (kpl)	12	2300	5	3	3	10	6	100	30	
Yht.lämpöteho (MW)	320	400	50	40	60	80	100	80	30	

Figure 6.2: Metal industry energy consumption, by process equipment

Figure 6.2 illustrates the fuel consumption in metal industry by process equipment in Finland in 1987. Rolling processes are the single most consuming process machinery branch. However, majority of the energy for rolling processes is provided with blast furnace gas. Other high consuming machinery include heat treatment furnaces, electric arc furnaces and blast furnaces. Blast furnace process and electric arc furnace processes are chosen as case studies in the steel sector, described in chapter 6.1.

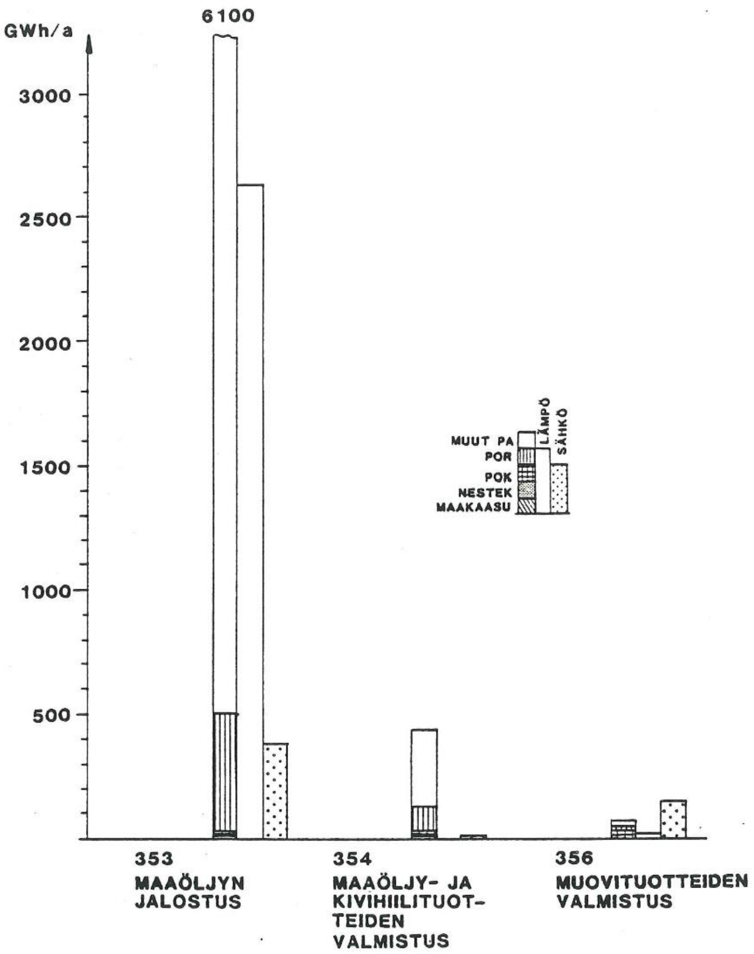


Figure 6.3: Fuel consumption in manufacturing of oil-, rubber- and chemical products in 1983 (Malin, 1988)

Figure 6.3 shows the fuel consumption in chemical industry sector in Finland in 1983. The oil refinement is responsible of the vast majority of the consumption in chemical industry. Oil and coal products have smaller energy consumption. Specific detail about energy use profile in the chemical industry sector is, that the fuel combustion, not electricity, is the main energy source in oil refining and oil and coal product manufacturing. Plastics manufacturing has high electricity than fuel consumption, but the fuel consumption is still large in the plastics sector too. All branches of the chemical industry consume natural gas, although the consumption is marginal compared to other fuels. Oil refining in chosen as the case study in the chemical industry sector, described in chapter 6.2.

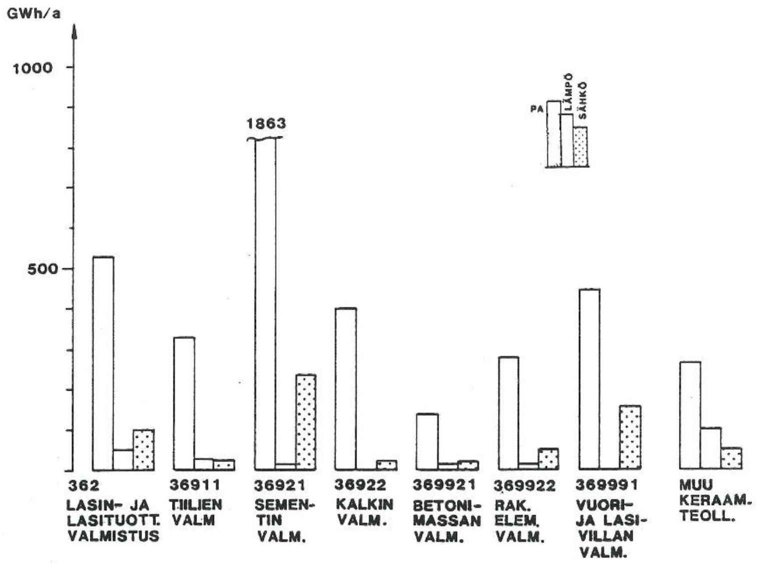


Figure 6.4: Energy consumption in manufacturing of clay-, glass- and stone products in 1983 (Malin, 1988)

Figure 6.4 illustrates the energy consumption in clay-, glass- and stone product industries in Finland in 1983. Cement manufacturing is the most energy consuming industry branch in this sector, followed by glass product manufacturing, glass and mineral wool manufacturing, lime manufacturing, brick manufacturing and construction element manufacturing. In all industry branches fuel use is the dominant energy source. Cement and glass manufacturing are chosen as case studies in this industry sector, presented in chapters 6.3 and 6.4, respectively.

Steel manufacturing has very high energy intensity and carbon intensity, due use of coal as main fuel, and use of coke as reducing agent. Ruukki steel mill in Raahe is the top carbon dioxide emitter in Finland (Ministry of Employment and the Economy, 2014), emitting 4,5-5 million tons CO_2 annually. Ovako is a smaller steel mill, which is chosen as a case study due its high share of natural gas in its energy use profile. Neste Oil refinery in Porvoo is the second with 3,7 million ton emissions (Neste Oil , 2011), however the indirect emissions from the products of the Neste Oil refineries in Finland result in emissions of 45 million tons (see section 6.2 & appendix 2). Finnsementti emits 0,9 million tons from its two cement plants, and cement industry is one of the largest emitters globally (see figure 1.2). Glass industry is known to use high share of natural gas in its energy use profile. Iittala is chosen as a case study in glass industry, even though its total emissions (11 000 tCO_2/a) are low compared to other cases, as information is available publicly unlike other glass industry companies. Another case in glass industry in the former Lahti glass factory of Pilkington Ltd., as it represents a typical case in float glass manufacturing.

Public information about energy balances of the companies is limited. A rationale to choose these case study companies in question was to select the ones which have at some point released information about their natural gas and total energy use.

6.1 Steel manufacturing

Steel is a metal alloy, with iron and carbon as main ingredients. It is commonly used in construction, manufacturing industry, power industry, agriculture, medicine and household items. It has high tensile strength and low cost. The exact composition of steel depend on material requirements of the end-use application. The world steel production exceeded 1000 million tons in 2004. The EU-27 production was 198 million tons in 2006.

Four steel mills are operational in Finland. Raahe and Koverhar steel mills are part of Ruukki/SSAB, Tornio steel mill is part of Outokumpu and Imatra Steel Oy Ab is part of Wärtsilä Corporation. Raahe and Koverhar are integrated steel mills based on blast furnace process. Outokumpu Stainless in Tornio is based on electric arc furnace process, using scrap metal and ferrochromium as raw materials. Imatra Steel is based on electric arc furnace process, using scrap metal as raw material.

6.1.1 Steel production routes

Four routes for steel manufacturing are currently used: the blast furnace/basic oxygen furnace process (BF-BOF), the direct smelting of scrap (in electric arc furnace process, EAF), smelting reduction and direct reduction. In 2006, 59,8 % of steel in EU-27 was produced in BF-BOF process, and 40,2 % is EAF process (Remus, et al., 2013). As these are only two production routes in Finnish steel industry, only BF-BOF and EAF processes are discussed in this thesis. Besides these two routes, steel can be produced by smelting reduction and direct reduction. The sub-processes of the four routes are depicted in figure 6.5.

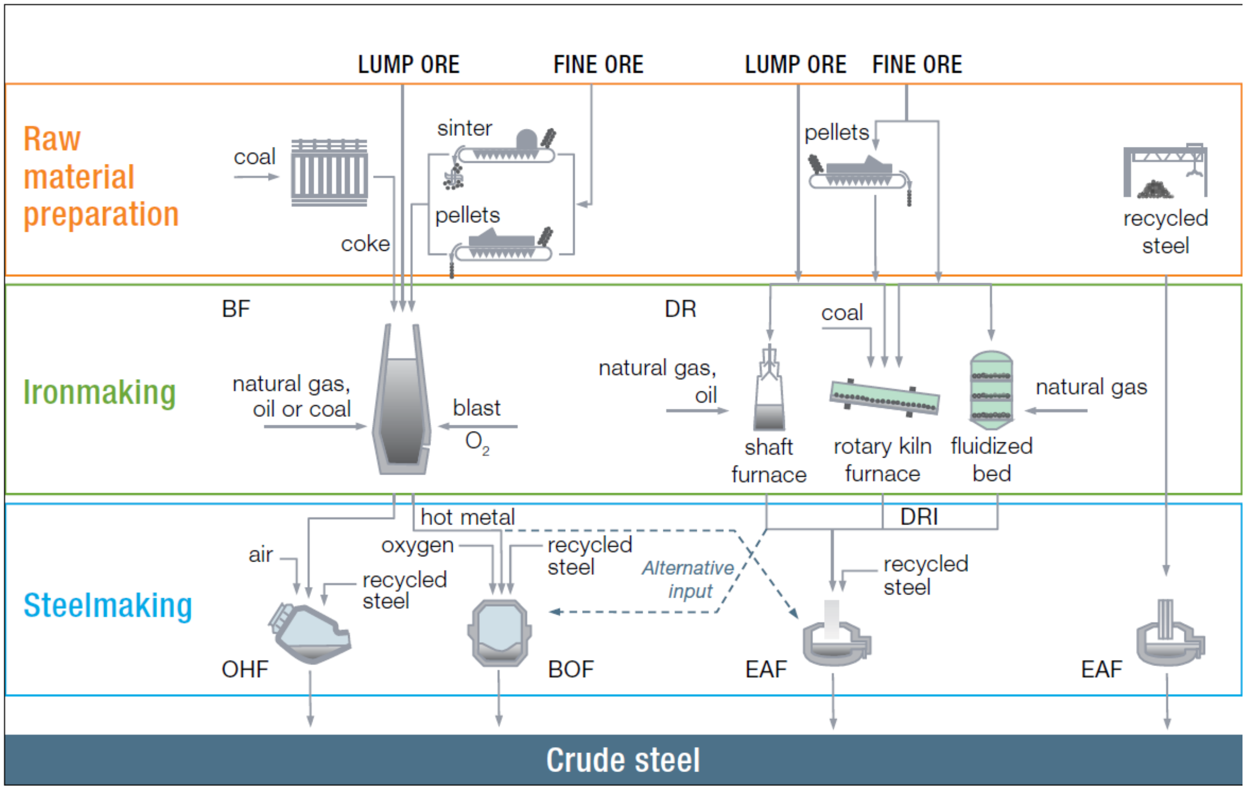


Figure 6.5: Crude steel production methods (Degner et al., 2008)

Steel is made either from virgin material or recycled steel, virgin material being more common. Both raw materials are used in steel production in Finland. The production from recycled steel is limited by availability of raw material (Yellishetty, et al., 2011). The raw materials are agglomerated and fed into blast furnace to produce hot metal. There are routes which do not require agglomeration (coal coking, iron ore sintering, pelletizing). Alternatively, instead of hot metal, direct reduced iron (DRI) can be produced which is refined into steel in an electric arc furnace (EAF). Various DRI methods are available, including shaft furnace, rotary kiln and fluidized bed technologies. 74 million tonnes of DRI was produced globally in 2012. Majority of DRI technologies (71 %) are based on gas, such as Midrex and HYL/Energiron (Midrex, 2013), rest being coal-based technologies. (Suopajärvi, 2014)

6.1.2 Blast furnace ironmaking

In blast furnace ironmaking, iron oxides are reduced and melted into hot metal by using fossil carbon (or potentially in future, biomass carbon) from coke, pulverized coal, oil, or natural gas in a blast furnace. Majority of carbon dioxide emissions in steel manufacturing results from this reduction process. The crude iron (pig iron) is produced in blast furnaces, where the raw iron is reduced into crude iron by carbon monoxide gas (CO-gas) and hydrogen gas (H-gas). These gases are formed when coke and extra heavy fuel oil are combusted in the furnace. The excess blast furnace gas is used in air preheating and electricity generation.

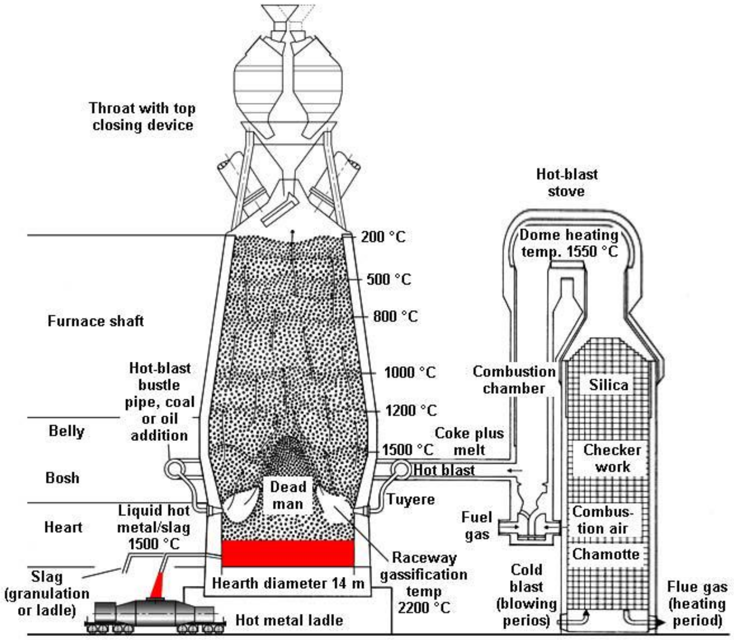


Figure 6.6: The blast furnace process schematic (Suopajärvi, 2014)

A blast furnace is a large counter-current reactor with steel stack lined with refractory bricks. The raw materials (iron, coke and limestone) are charged from the throat at the top of the furnace. Reducing agents (coal, oil or natural gas) are injected from the lower part of the furnace, with oxygen-enriched air. The blast furnace process is depicted in the figure 6.6. (Suopajärvi, 2014)

The blast furnace process

The raw materials should be prepared for the blast furnace process to preferable particle size by crushing, grinding and screening. The raw materials are then mechanically conveyed to the blast furnace. A blast furnace is a closed system, where the iron-bearing materials (iron ore, sinter and/or pellets), additives (slag formers, such as limestone) and reducing agents (coke or other fossil material) are charged continuously. The blast furnace gas (BF-gas) is prevented from exiting the furnace. A hot air blast, enriched with oxygen and other reducing agents, is injected into the blast furnace’s tuyère level. This gas mixture forms carbon monoxide (CO) with the coal-based reducing agents. Part of the coke can be replaced by coal, oil or natural gas, which are also injected directly to tuyère level. The CO reduces iron oxides into metal iron. The liquid iron is collected into the hearth along with the slag, and both are cast on regular basis. The liquid iron is transported to casting, the slag is processed to be applied in concrete or road material manufacturing. The coke oven gas is collected from the top of the furnace, and used in heat and electricity generation at the other processes of the steel mill.

The oxygen is removed from the iron oxides in stages. Firstly, hematite (Fe_2O_3) is reduced into magnetite (Fe_3O_4), which is further reduced into wüstite (FeO). The first two reactions occur by indirect oxidation with reducing gases. The wüstite reaction occurs in the lower part of the reactor by indirect reduction with CO gas, or by direct reduction with solid carbon. The liquid metal in the blast furnace is periodically cast. One of the tap holes in the side wall of the hearth is opened. The hot metal flows through refractory or low cement runners and is poured into ladles at the granulation plant or to an open pit. The tap hole at the hearth is closed again.

Most common auxiliary fuel is pulverized carbon. Significant substitution of coke by pulverized coal is possible, however it affects the chemical properties of the blast furnace process; the ash decreases the heating value of coal. Extra-heavy fuel oil is used as auxiliary fuel as well, which was common practice in the 1960's. The oil crisis in the 1970's switched fuel oil use towards use of coal. Ruukki steel mill has achieved the consumption of 358,0 kg coke and 100,5 kg oil/t hot metal (Luengen, et al., 2011). Natural gas substitution has been practiced in the markets where natural gas is inexpensive; in the USA, steel mills have reached 155 m^3/t hot metal natural gas consumption (Babitch, et al., 2002). Higher injection rates lead to local supercooling of the furnace hearth. Plastics and coke oven gases have also been used in the blast furnaces (Babitch, et al., 2002). (Suopajärvi, 2014)

6.1.4 Electric Arc Furnace Steelmaking

The direct smelting of iron-containing materials such as scrap, is usually performed in electric arc furnace (EAF). Nowadays in EU 41,8 % of all steel production is performed in EAF plants (Stahl, 2008). In Italy (61 % of all steel) and Spain (77 %), EAF route is significantly higher than BF-BOF process (Remus, et al., 2013). Steel manufacturing in electric arc furnaces is mature and cost-effective steel manufacturing method. It uses scrap metal as raw material, which do not require chemical reduction process. Therefore, the primary energy consumption and CO_2 emissions are lower than in ore-based steel manufacturing. However, the electricity consumption is approximately two times higher than in ore-based production (Ohlström & Savolainen, 2005).

Feedstock in EAF is mainly ferrous scrap, consisting for example scrap from steel production, cut-offs from steel product manufacturers (e.g. automotive industry) and end-of-lifecycle steel products. Direct reduced iron (DRI) is also used as a feedstock, as it contains low amount of gangue and undesirable metals (e.g. copper), and has more stable price than scrap. Ferroalloys are used to control the iron content of the finished steel.

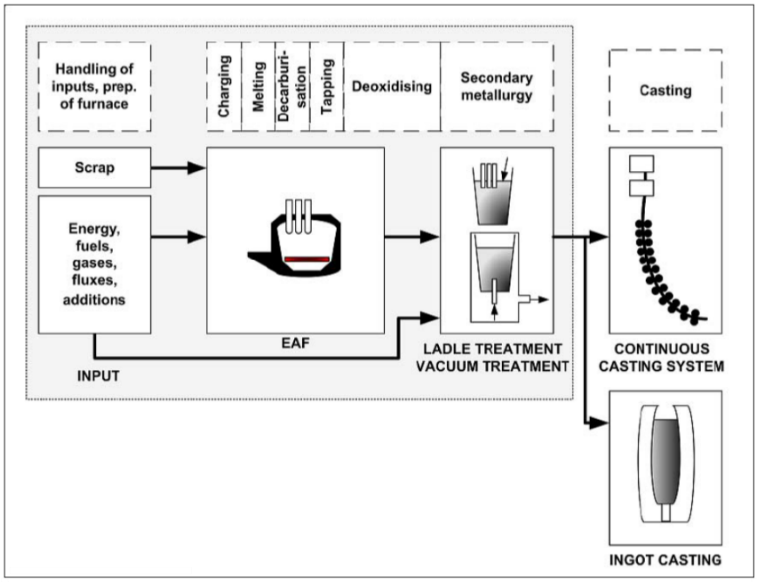


Figure 6.7: Electric arc furnace steelmaking process schematics (Rentz, 1997)

Figure 6.7 illustrates the electric arc furnace steelmaking process. The EAF process starts with scrap preheating, which is done outside the EAF, usually in scrap charging baskets, in a charging shaft added to EAF, or in a specially designed scrap conveying system allowing continuous charging during the melting process. The preheated material in then charged into the EAF, usually with lime or dolomite. Carbon-based materials are also charged, to adjust the quality of the finished steel. The furnace electrodes are raised into top position, and the roof is opened. Normally 50-60 % of the scrap is charged initially with the first charge basket; then the roof is closed and the electrodes are lowered into the scrap. An electric arc struck 200-300 mm above the scrap, melting the scrap. Initially, the power is kept low to prevent damage to the furnace walls and the roof. When the electrodes are submerged into molten metal, the power can be increased to complete the melting process. After the initial charge is melted, the remainder of the scrap is added. Oxygen lances and/or oxy-fuel burners are sometimes used to complete the melting. Natural gas and oil are used as fuels. Oxygen is used for both metallurgical reasons and to improve efficiency. The slag is removed from the steel after the melting process by tapping, either from the EAF or at the deslagging station. The slag can be used in construction industry. Secondary metallurgy is performed after the tapping. This is usually done in ladle furnaces, which have an arc-heating unit to control the temperature of casting process. The treatment includes the addition of deoxidizing agents and alloying elements to adjust the chemical composition of finished steel. In some processes, vacuum treatment I used to adjust the concentration of such elements as hydrogen, nitrogen and oxygen in the finished steel. Inert gases (Ar or N_2) are used for homogenization of the steel. In stainless steel making, argon oxygen decarburization (AOD) converter or vacuum oxygen decarburization (VOD) treatments are used. After the secondary metallurgy, the molten steel is casted.

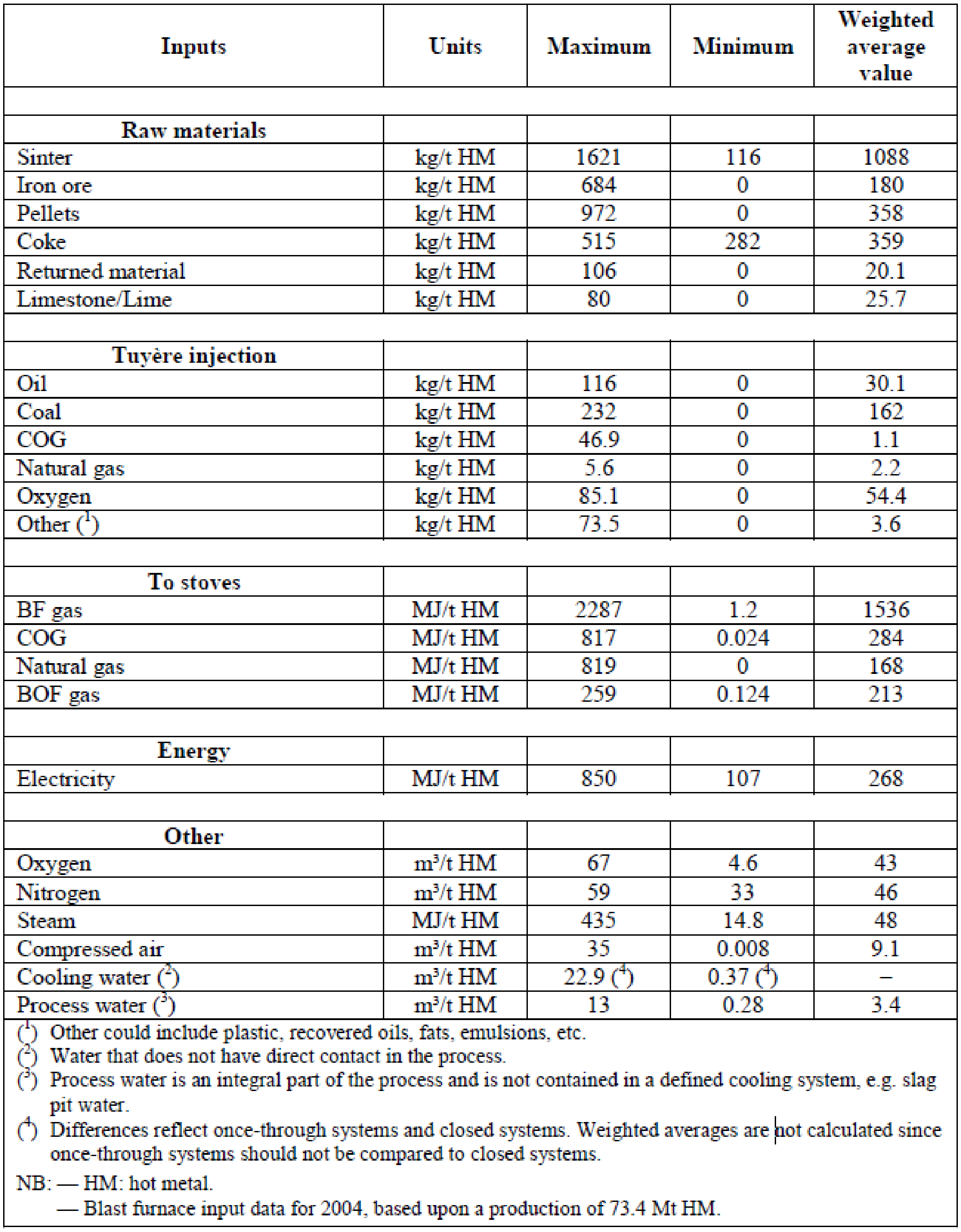
6.1.3 Energy use in steel industry

Steelmaking is very energy-intensive industry. The melting point of iron is 1510 °C and therefore the steel production requires large amount of energy, as reducing agents and electricity. Besides this, the iron has to be reduced which requires reducing agents, usually fossil fuels. Estimates of specific energy consumption (SEC) of BF-BOF method range from 5 to 6,4 MWh/ton of steel (World Steel Association, 2011) (IEAGHG, 2013), while the best practice SEC is 5,7 MWh/t (Worrell, et al., 2008). World Steel Association claims that the energy consumption could be as high as 8,6 MWh/t of crude steel. Producing steel from recycled steel consumes considerably less energy; the best practice SEC is 2,2 MWh/t (Worrell, et al., 2008). The World Steel Association (World Steel Association, 2011) estimates the energy consumption of EAF steel production is 2,5-3,5 MWh/t crude steel. (Suopajärvi, 2014)

Reducing agents for steel manufacturing are mainly carbon-based fossil materials (coal, oil, natural gas), however biomass-based reducing agents are studied (Suopajärvi, 2014). Majority of the CO_2 emissions result from the use of coke and injected reducing agents in the blast furnace. IEA (IEA, 2007) estimates the CO_2 emissions in BF-BOF process is 1,7 tCO_2/t of hot rolled coil, depending on the input materials and the efficiency of the process. Typical amount of reducing agent in European blast furnaces is 300-350 kg of coke and 150-200 kg of coal per ton of hot metal produced (Luengen, et al., 2011). DRI-EAF route based on the use of natural gas produces 1,1

tCO_2/t crude steel. DRI-EAF route based on the use of coal produces 2,5 tCO_2/t crude steel. CO_2 emissions from recycled steel material are approximately 0,4 tCO_2/t crude steel (IEA, 2007). (Suopajärvi, 2014) Blast furnace gases needed for the reduction reaction are generated by burning coke (or other carbon-based fuel) and injected into the furnace. Coke is the primary fuel, and CO from coke is the primary reducing agent in the blast furnace process. Coke is mechanically strong, porous material that has three functions in the blast furnace: (1) it acts as a reducing agent; (2) it provides energy for the process; (3) it serves as a support medium for the burden material (Xu & Cang, 2010). The amount of coke used per ton of steel is 300-350 kg, depending on the other reducing agents used in the process (Luengen, et al., 2011). Danloy et al. (Danloy, et al., 2008) and Hooey et al. (Hooey, et al., 2010) claim that using other reducing agents the amount of coke could be reduced to 200 kg/t hot metal. Other reducing agent are used in the blast furnace process in cases when they are cheaper than coal. (Suopajärvi, 2014). CO or H_2 can be used as reducing agent in the reduction of iron ore into iron (Laplace Conseil, 2013). Steel manufacturing from ore consume approximately six times more energy than steel manufacturing from scrap, due chemical energy requirement of reduction process. However, due efficient use of blast furnace gases, the primary energy consumption is two-fold and carbon dioxide emissions are four-fold in steel manufacturing from iron ore, compared to manufacturing from scrap metal.

Table 6.1: Input data from blast furnaces in different EU members states (Eurofer, 2009) (Poland, 2007)



Inputs	Units	Maximum	Minimum	Weighted average value
Raw materials				
Sinter	kg/t HM	1621	116	1088
Iron ore	kg/t HM	684	0	180
Pellets	kg/t HM	972	0	358
Coke	kg/t HM	515	282	359
Returned material	kg/t HM	106	0	20.1
Limestone/Lime	kg/t HM	80	0	25.7
Tuyère injection				
Oil	kg/t HM	116	0	30.1
Coal	kg/t HM	232	0	162
COG	kg/t HM	46.9	0	1.1
Natural gas	kg/t HM	5.6	0	2.2
Oxygen	kg/t HM	85.1	0	54.4
Other (1)	kg/t HM	73.5	0	3.6
To stoves				
BF gas	MJ/t HM	2287	1.2	1536
COG	MJ/t HM	817	0.024	284
Natural gas	MJ/t HM	819	0	168
BOF gas	MJ/t HM	259	0.124	213
Energy				
Electricity	MJ/t HM	850	107	268
Other				
Oxygen	m^3/t HM	67	4.6	43
Nitrogen	m^3/t HM	59	33	46
Steam	MJ/t HM	435	14.8	48
Compressed air	m^3/t HM	35	0.008	9.1
Cooling water (2)	m^3/t HM	22.9 (4)	0.37 (4)	–
Process water (3)	m^3/t HM	13	0.28	3.4

(1) Other could include plastic, recovered oils, fats, emulsions, etc.
(2) Water that does not have direct contact in the process.
(3) Process water is an integral part of the process and is not contained in a defined cooling system, e.g. slag pit water.
(4) Differences reflect once-through systems and closed systems. Weighted averages are not calculated since once-through systems should not be compared to closed systems.

NB: — HM: hot metal.
— Blast furnace input data for 2004, based upon a production of 73.4 Mt HM.

Table 6.1 describes the average inputs per ton of hot metal in EU blast furnaces. As can be seen, natural gas is used for tuyère injection (on average 2,2 kg/t hot metal, or 1,1 % of carbon-bearing material used for tuyère injection), and in stoves (168 MJ/t hot metal, or 7,6 % of fuels used in stoves). Besides methane, also oxygen is used in blast furnace process.

6.1.4 Potential for fuel substitution to SNG

Two steel mills in Finland are connected to natural gas network (Ovako in Imatra and Ruukki in Hämeenlinna). Malin (1987) studied the natural gas consumption in Finnish metal industry in 1987. Malin estimated the consumption on natural gas in Ovako Steel in Imatra 23 Mm^3/a (230 GWh/a). Ovako used natural gas for one (of two) electric arc furnaces, scrap preheating, crucible drying, continuous casting machine, rolling processes and heat treatment. The steel production in Ovako mill was 200 000 t/a. If the energy consumption is assumed 3,5 MWh/t crude steel (World Steel Association, 2011), the energy provided by natural gas provided 33 % of total energy consumption (694 GWh) of the Ovako Imatra steel mill in 1987[2].

Two steel mills in Finland are not connected to natural gas network (Ruukki in Raahe and Outokumpu in Tornio). Malin (1987) estimated the coke consumption to be 5000 GWh/a, heavy fuel oil consumption 1800-2000 GWh/a, extra heavy fuel oil 460-570 GWh/a, light fuel oil 230 GWh/a, LPG 20 GWh/a, and tall-oil pitch 70 GWh/a. Therefore, oil and gas products share of total energy consumption was 34 % (2660 GWh/a). Sintering plant of the plants used heavy fuel oil, blast furnaces extra heavy fuel oil and COG, steel melting light fuel oil and LPG, rolling processes heavy and light fuel oil, and power plant heavy and extra heavy fuel oil as well as COG. However, the completion of ManGa LNG terminal in Tornio will likely significantly increase the natural gas consumption in Outokumpu and Ruukki steel mills.

Nogami, Yagi, Kitamura and Austin (Nogami, et al., 2006) studied the effect of metallic charging, top gas recycling and natural gas injection to mass and energy balances of blast furnace ironmaking method by kinetic-based numerical simulations. The coke was the primary fuel in the study, and coal and natural gas were used as auxiliary fuels.

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[2] Data about Ovako presented here is from 1987. At that time, the capacity of Imatra Steel mill was 200 000 t/a (Malin, 1987). Nowadays the capacity of the plant is 300 000 t/a (Ovako). Therefore, significant changes has been made in the plant and the data presented here is not up to date.

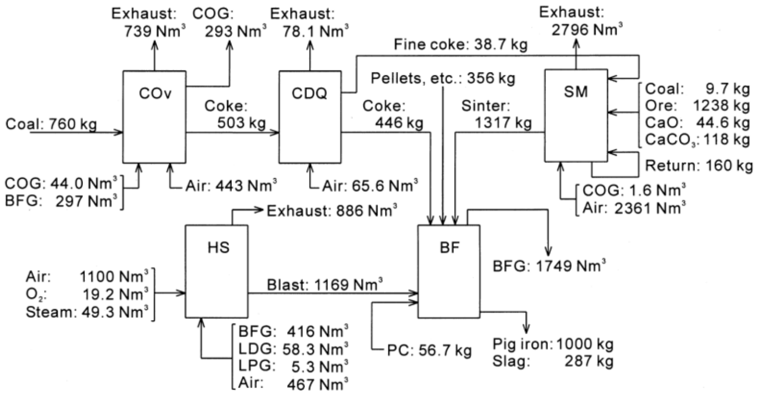

Figure 6.8: Material flow of the standard ironmaking system (Nogami, et al., 2006)

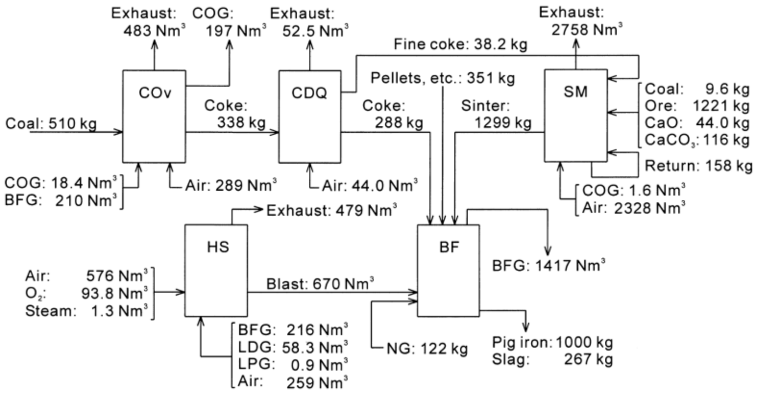

Figure 6.9: Material flow of the natural gas injection ironmaking system (Nogami, et al., 2006)

Figure 6.8 illustrates the material flow of the standard ironmaking system, and figure 6.9 illustrates the material flow of the natural gas injection system in Norigami et al. (2006) study. The natural gas consumption increased to 122 kg/t pig iron from 0 kg in the standard system, while coal consumption decreased from 760 kg to 510 kg/t pig iron. Oxygen consumption increased from 19,2 Nm^3 to 93,8 Nm^3. The oxygen can be provided by power-to-gas system.

Table 6.2: Fuel inputs to evaluated systems (t/hot metal basis) (Nogami, et al., 2006)

Operation	Coal [kg]	NG [Nm3]	LDG[3] [Nm3]	LPG [Nm3]	Oxygen [Nm3]
Standard	826,3	0,0	58,3	5,3	24,7
Metallic recharging	391,1	0,0	58,3	2	11,3
Top gas recycling	676,1	0,0	58,3	0,0	202,6
Natural gas injection	520	161,5	58,3	0,9	93,8

Table 6.3: Energy inputs to evaluated systems [MJ/t hot metal] (Nogami, et al., 2006)

Operation	Coal	NG	LDG[2]	LPG	Elec.	Total
Standard	27011	0	379	628	378	28395
Metallic recharging	12785	0	379	236	257	134
Top gas recycling	22100	0	379	0	627	22732
Natural gas injection	16997	5885	379	104	470	23713

Tables 6.2 and 6.3 describe the fuel and energy inputs of the four systems. The fuel inputs in table 6.2 are converted to energy inputs in table 6.3. In natural gas injection, the pulverized coal injection to tuyère layer is replaced with natural gas. The amount of natural gas input is lower than replaced coal input in standard system.

Figure 6.10 shows the energy balances of the studied systems. All systems showed decrease in energy input, especially metallic charging. Decrease in natural gas injection is small. In natural gas injection system the energy output (as coke oven gas and blast furnace gas) is almost as large as in standard system. Figure 6.11 shows the CO_2 emissions of the four systems. Natural gas injection and metallic charging systems show decrease in CO_2 emissions.

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[3] LDG=Converter off gas

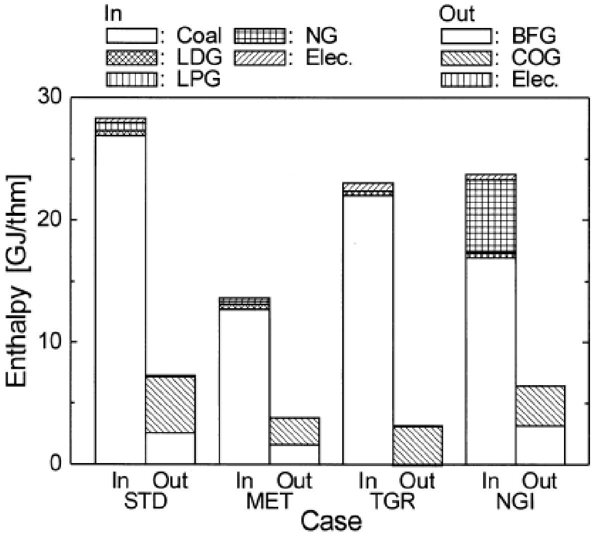


Figure 6.10: Energy balance of four evaluated ironmaking systems (Nogami, et al., 2006)

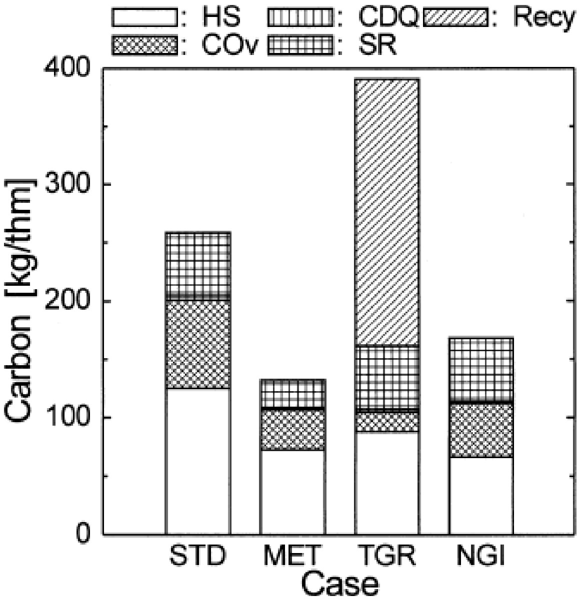


Figure 6.11: CO2 emission from four ironmaking systems (Nogami, et al., 2006)

However, Nogami et al. accounted the emissions from natural gas in their study. As power-to-gas generated methane is zero-carbon, further emission reductions can be reached. Model Nogami et al. used is not available, but further 15-20 % CO_2 reduction from zero-emission SNG could be reached (see Appendix 1).

6.3 Oil refining

Crude oil is viscous, lipophilic liquid at ambient temperature, which is combustible and has high energy content. Crude oil is former organic matter, which has been geochemically processed over millions of years. It is a mixture different hydrocarbons (mainly paraffin, naphthalene and aromatic hydrocarbons), and trace amounts of sulfur-, nitrogen- and oxygen based compounds, as well as some metals, such as vanadine and nickel. Additionally, there are some physical impurities, such as salts, water and sediments which originate from the source or marine transport. Oil is refined into fuels for road, air and marine transport, combustion fuels for energy and other industries, raw materials for the petrochemical and chemical industries. The refinery process includes numerous different sub-processes.

Oil is commonly used as transport fuel and to fuel industrial processes, and to some extent in electricity and heat production (mainly at peak power plants nowadays). Oil provides 42 % of the EU's total energy demand, and 94 % of the transport fuels. Therefore, the EU's refinement industry has critical strategic importance. The sector has suffered from overcapacity since the first oil crisis, and distilleries have been closed, but the total oil processing capacity has been growing in recent years due to optimization of the capacity of current refineries. Currently there are approximately one hundred oil refineries around the EU countries. In this chapter, the refineries of Neste Oil in Porvoo and Naantali are chosen and descrobed as case studies, as described in book Suomen Kemianteollisuus by (Riistama, et al., 2003).

6.3.1 Refinery process

Oil refining is a distillation process and physical and chemical treatment of the distillates to different commercial oil products. Distillation is the key process, and other chemical sub-processes include cracking, alkylation and polymerization, which increase the amount of valuable light runnings (e.g. gasoline), and reforming and hydrogen treatment, which improve the qualities of the different distillates. Reforming increases the octane number of the gasoline, which improves the compression ratio of the gasoline. Hydrogen treatment is used to remove sulfur and heavy metals, as well as cracking the complex hydrocarbons to shorter ones. The type of wanted end-products, the crude oil quality and requirements from the authorities influence the configuration of the refinery, and the configuration of individual refineries differ significantly. Figure 6.12 depicts the simplified schematics of Neste Oil Porvoo refinery process, which includes over 30 different sub-processes. The main sub-processes are described in more detail in this chapter.

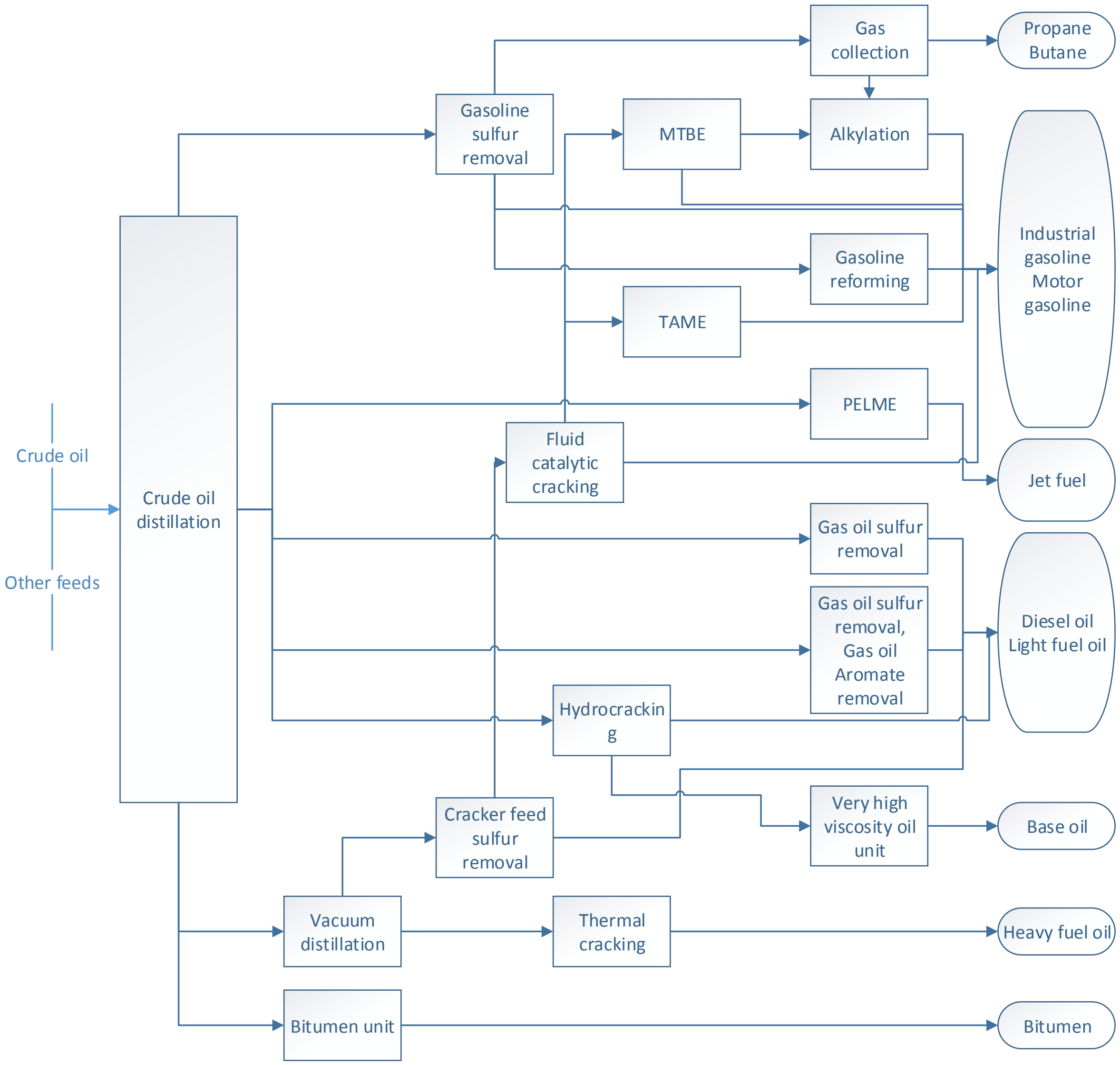


Figure 6.12: Simplified schematics of Neste Oil Porvoo refinery process

Salt removal and crude oil distillation

Crude oil usually includes salt, either from the source or from marine transportation. The removal of the salt is the first step of the refinery process. Crude oil is pre-heated to 120-140 °C. 3-5 % water is mixed with the oil. Electric current is applied to the oil. The salt mixes with the water, separating the salt from the oil, and the electric current makes the water droplets to descent deeper through the crude oil. The water settles into the bottom, from where it is collected and fed to wastewater treatment. The oil is flown to distillation process.

Before distillation, the oil is heated to 350-380 °C and pressurized to 0,1 bar overpressure. The oil is then fed into distillation column, which has dozens of column trays to separate different distillates. Motor gasoline and lighter distillates are gaseous, ascending to the top of the column. Heavier distillates are collected from the middle of the column. These are called middle runnings. Distillation produces the following runnings:

- ***Gases*** are collected, and separated to *combustion gases* (hydrogen, methane, ethane) and *liquefied petroleum gases* (propane, butane)
- Majority of the ***straight-run gasoline*** is fed to sulfur removal and then to reformation process. Some is used as *industrial gasoline*.
- ***Middle runnings*** include *petroleum* (170-230 °C) and *gas oils* (220-380 °C). Gas oils are used for *diesel oil* and *light fuel oil* production after the sulfur removal.
- ***Heavy gas oil*** (360-480 °C) is fed into catalytic cracking process.
- Undistilled ***bottom oil*** (over 450 °C) is fed to vacuum distillation.

Vacuum distillation column (bottom oil unit) is distillation process in 70 mbar pressure and approximately 400 °C. The products are vacuum gas oil, which are fed to catalytic cracking, and undistilled bottom oil, which is partly refined into heavy fuel oil by thermal cracking or dilution, and partly bitumen.

Cracking

Cracking is the process of cutting the complex hydrocarbons into shorter ones. The main cracking methods are thermal cracking and catalytic cracking. Neste refinery in Porvoo uses hydrocracking and fluid catalytic cracking (FCC), and Neste refinery in Naantali uses thermoform catalytic cracking (TCC). Inputs are vacuum gas oil and heavy gas oil. Inputs should include low amounts of metals to preserve the catalysts.

FCC unit includes reactor and regenerator in parallel. FCC process occurs in 500 °C temperature and 1,5 bar pressure by aluminum oxide-silica (zeolite) catalyst. The cracking reaction occurs in a riser pipe where the vaporized input oil and the catalyst are broken into lighter components, including LPG, gasoline and diesel. Catalyst is separated from the vaporized oil products in the reactor using cyclones and re-fed into regenerator, where the catalyst is separated from collected petroleum coke using air blast. Cracked oil vapors are fed into distillation column and distilled into products. Light gas contains ethane, which is fed to ethane plant (Borealis Polymers Oy in Porvoo case). Liquefied gases are polymerized or distilled into C_3 and C_4 –fractions. C_3 is fed into propene unit (of Borealis) and C_4 to MTBE-unit. Gasoline distillate is distilled into light gasoline (C_5-C_7) and fed to the TAME-unit. Heavy gasoline is desulfurized and used as high octane motor gasoline component. Middle runnings are desulfurized, and bottom oil is mixed with heavy fuel oil.

Thermofor catalytic cracking (TCC) unit includes reactor and catalyst regeneration kiln are on arranged as column. The catalyst is elevated to the top of the column with pressurized air. The catalyst then ascends through the reactor, reacting with preheated (440 °C) oil. Cracked product is separated from the catalyst on the bottom of the column with steam and fed to distillation. Heavy gas oil is re-fed to the process to improve gasoline recovery. Catalyst is covered with petroleum coke, which is combusted in the regeneration kiln. Catalyst is then cooled, and blown to the top of the column by pressurized air.

Hydrogen cracking occurs in high overpressure and 400 °C temperature, cracking vacuum distillates to lighter compounds by hydrogen gas. No olefin compounds remain after the process, and sulfur and nitrogen compounds are reduced into hydrogen sulfide and ammonia. The process occurs in two series-connected reactors. The sulfur and nitrogen reduction occur in the first reactor, and cracking in the second, catalyzed by zeolite-based nickel-tungsten catalyst. The hydrogen cracking process creates combustion gas, gasoline, petroleum, gas oil and bottom oil. The products are separated from bottom oil by distillation. The hydrogen gas is produced from natural gas by steam reforming.

Reforming

Reforming is a process where low-octane (octane number 50-60) gasoline is processed into high octane (octane number 90-100) motor gasoline. The input is gasoline from crude oil distillation and hydrogen cracking. The following chemical processes occur in the reforming, which all increase the octane number of the gasoline:

- Aromatization of naphthalene
- Cyclic ring formation
- Isomerization of naphthalene
- Isomerization of paraffin
- Hydrogen cracking

Neste Oil’s refinery process uses aluminum oxide platinum as catalyst. The reforming process occurs in 500-540 °C temperature and 5-9 bar pressure in four kilns on top of each other. The catalyst is fed from the top reactor, and descended though the four kilns. Coke is formed on the catalyst, which is combusted in a regenerator. Hydrogen is separated from the product, and the product is fed into distillation where benzene is separated and transported to benzene plant at Borealis factory. Product gasoline is used in motor gasoline production. Hydrogen is used in cracking process and sulfur removal. Other gases are processed in gas removal, to separate combustion gas from liquefied gases.

Alkylation

Alkylation is a process of converting liquefied gas runnings (n-butylenes and isobutane) into high octane motor gasoline component, with octane number 95-96. Alkylene is therefore optimal component for lead-free fuel manufacturing. Isobutane and butylene originate from FCC-cracking unit, as well as butadiene plant of Borealis, after reacting with methanol in MTBE-unit. Alkylation reaction occurs in pipe reactor 30 °C temperature and 6 bar pressure. The reaction products are fed to the clarifier, where hydrogen fluoride is separated from reaction products and fed back into the reactor after cooling. The reaction products are then fed into separation column, where alkylate is separated from isobutane, which is returned into the alkylation reactor, and propane, which is fed into propane separation column and propane purification. Alkylation unit also includes hydrogen fluoride and waste gas and wastewater purification processes.

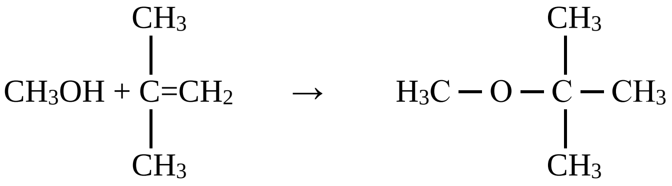
Sulfur removal and sulfur production

Sulfur should be removed from the distilled gasoline and middle runnings from crude oil distillation and catalytic cracking before reforming. Sulfur removal occurs in hydrogen pressure (15-30 bar for gasoline, 50 bar for middle runnings) and 300-380 °C temperature, in presence of cobalt, nickel or molybdenum catalyst. Sulfur compounds are reduced to hydrogen sulfide, nitrogen to ammonia, oxygen is removed, and metal platinum catalyst are absorbed on the top of sulfur removal catalyst. The distillation of the product oil produces sulfur-free gasoline, solvents, jet fuel and gas oil. The gas oil is used as a component for diesel oil and light fuel oil.

Liquid gases from FCC-unit contain hydrogen sulfides and mercaptan. Mercaptan is corrosive and foul smelling. Mercaptan is oxidized with oxygen from air into disulfides by copper catalyst solution. Liquid and gaseous products from absorbed into 25 % amine solution (e.g. di-isopropanolamine and dietanoliamine) in 5-10 bar pressure, to remove hydrogen sulfide. Absorption solution is regenerated by heating in atmospheric pressure, when hydrogen sulfide is removed from amine, and fed into Claus furnace to produce elemental sulfur.

Oxygenate production

Oxygenates are hydrocarbons that include oxygen, and part of Neste Oil's gasoline. Oxygenates are produced in methyl-tert-buthyl ether unit (MTBE). Inputs for MTBE are methanol and C_4-gases. These inputs react in 70 °C temperature and 15 bar pressure with acidic ion exchange resin as a catalyst. The MTBE reaction is:



$$CH_3OH + \underset{\underset{CH_3}{|}}{\overset{\overset{CH_3}{|}}{C}}{=}CH_2 \rightarrow H_3C-O-\underset{\underset{CH_3}{|}}{\overset{\overset{CH_3}{|}}{C}}-CH_3 \qquad (13)$$

Tert-amyl-methyl ether (TAME) unit is another oxygenation unit in Neste Oil's Porvoo refinery. TAME uses methanol and olefinic light gasoline, including C_5-C_7 hydrocarbons, from FCC. Undesired diolefins are first reduced to olefin. The etherification reaction occurs in similar conditions than MTBE. Methanol reacts with isoamylen and isohexene, forming oxygen-bearing ethers. The bottom oil, which includes hydrocarbons from FCC process, is used as gasoline component. Methanol is enriched, and returned to the process.

Hydration of aromatic compounds

Hydration process adds hydrogen to aromatic compounds. The hydrogen addition increases the quality of the solvents, decrease the emissions from gasoline and removing problematic benzene from motor gasoline. Neste Oil's Naantali refinery manufactures solvents and kerosene, using gasoline as input. These are first desulfurized. The aromatic compounds react with hydrogen in high pressure and temperature in Arosat reactor, transforming benzene ring of aromatic compounds to cyclohexane ring.

Neste has also engineered its own NExSAT hydration technology, based on nickel-based catalyst. Solvent producing unit uses distilled and desulfurized middle runnings as inputs. Products are solvents free of aromatic compounds. Porvoo refinery has aromatic removal unit, which processes light gas oil and hydrogen cracked petrol, producing aromatic free middle runnings, which are used as components for reformed diesel fuels. Porvoo also has hydration unit for benzene, where the benzene in desulfurized light gasoline is hydrated into cyclohexane. Benzene-free light gasoline is a component for low-emission motor gasoline.

Other refinery units and hydrogen production

Plane kerosene is processed in *Merox process*, where mercaptan-sulfur compounds are oxidized with air into disulfides. The kerosene is then washed with water, dried and filtered in multiple stages before fueling into airplanes. Some chemicals are also added, e.g. to improve the electric conductivity of the kerosene. Liquefied olefin gas compounds C_3 and C_4 are transformed into gasoline and middle runnings by *catalytic polymerization*. Polymerization occurs in presence of phosphorous acid catalyst. Remaining liquefied gases are separated by distillation. The phosphorous acid is absorbed into sand in Naantali refinery. Porvoo refinery hexane separation unit processes the hexane from *gasoline sulfur removal process*. The process is based on molecular sieves. N-hexane is absorbed into the molecular sieve mass, while iso- and cyclohexane pass the mass. The sieve is regenerated by hydrogen. N-hexane is used in ethene plant, and isohexane in motor gasoline production after benzene hydration.

Hydrogen unit produces the hydrogen required in cracking, aromate hydration, diolefin selective hydration, and sulfur and metal removal processes. Porvoo refinery applies natural gas and LPG steam reforming. If LPG is used as input, sulfur is first removed. Then the input is heated to 850 °C temperature, when the hydrocarbons react with steam, in presence of nickel catalyst:

$$C_nH2_{n+2} + n\ H_2O \rightarrow n\ CO + (2n + 1)\ H_2 \qquad \Delta H > 0 \qquad (14)$$

To increase the amount of hydrogen and to remove carbon oxides, the carbon monoxide is converted into catalytic carbon dioxide:

$$CO + H_2O \leftrightarrow CO_2 + H_2 \qquad \Delta H = -42\ kJ \qquad (15)$$

High temperature converter (350-400 °C) iron oxide is used as a converter, while low temperature converter (200-250 °C) use copper oxide. Sodium carbonite-glycine solution is used to wash the CO2 from the gas. Methanation is used to reduce carbon monoxide and carbon dioxide to methane in presence of the nickel catalyst:

$$CO + 3\ H_2 \rightarrow CH_4 + H_2O \qquad \Delta H = -205\ kJ \qquad (16)$$

$$CO + 4\ H_2 \rightarrow CH_4 + 2\ H_2O \qquad \Delta H = -165\ kJ \qquad (17)$$

Final product includes 97 % hydrogen and rest is methane. Carbon dioxide is collected from the process by AGA Oy.

6.4 Cement manufacturing

Cement is a fine, non-metallic, inorganic powder, which forms a paste that harden over time. This hydraulic hardening in primarily due to the formation of calcium silicate hydrates as a result of the reaction between mixing water and the constituents of the cement. Cement is a basic construction material, used in most modern constructions. The use of cement dates back to antiquity in Europe, and the Portland cement, the most common cement type in concrete construction today, was patented in 1824.

The cement markets are usually local. According to (Schorcht, et al., 2013), the transport of cement is usually economically viable for up to 200-300 km. Finnsementti Oy provide 80% of the cement sold in Finland (the company imports approximately 1/3 of the rest). Demand for cement is closely related to construction markets, and during 1990's the demand for cement fluctuated steeply. There are 268 cement clinker production plants in the EU-27. Additionally, there are two clinker plants without mills, and 90 grinding plants without kilns. Typical kiln produces 3000 tonnes clinker per day. In Finland, Finnsementti Oy manufactures cement in its factories at Lappeenranta and Parainen and is the only cement manufacturing company in Finland at the moment. Finnsementti is part of international CRH group.

Main ingredient in cement clinker is limestone (mainly calcium carbonate, $CaCO_3$). Other ingredients used in cement manufacturing are other stone material, air cooled blast furnace cinder, fly ash and black rust. Gypsum ($CaSO_4$), ferrosulfate and aggregate material (e.g. blast furnace cinder) are added after clinker burning. Typical chemical composition of Portland cement includes tricalcium silicate (40-80 %), dicalcium silicate (10-50 %), tricalcium aluminate (0-15 %), tetracalcium aluminoferrite (0-20 %), calcium oxide (0-3 %), magnesium oxide (0-5 %), dipotassium sulfate (0-2 %) and disodium sulfate (0-1 %) (Schorcht, et al., 2013).

6.4.1 Cement manufacturing

There are four main process routes for the cement manufacturing:

- In the *dry process*, the raw materials are ground and dried to raw meal in the form of a flowable powder. The dry raw meal is fed to the preheater, precalcirner kiln, or a long kiln.
- In the *semi-dry process* the dry raw meal is pelletized with water and fed into grate preheater before the kiln or to a long kiln equipped with crosses.
- In the *semi-wet process* the slurry is first dewatered in filter presses. The resulting filter cake is extruded into pellets and fed either to grate preheater or directly to a filter cake dryer for raw meal production.
- In the *wet process* the raw materials are ground in water to form pumpable slurry, which is then fed directly into the kiln or first to slurry dryer.

The process of Finnsementti is chosen as a case study in this thesis, as the company is the sole cement producer in Finland. Finnsementti has factories in Lappeenranta and Parainen, which both use the dry process. Parainen has one kiln and Lappeenranta two. The Parainen factory produced 769 kt/a and Lappeenranta 425 kt/a in 2002 (Ohlström & Savolainen, 2005).

Raw material grinding

Finnsementti uses domestic limestone as a raw material. Limestone is ground at the limestone quarry and transported to cement factory. At the factory, grinded limestone is stored in silos from where it is moved to grinding. 80-90 % of raw materials used in Finnsementti’s cement manufacturing process are domestic (Ohlström & Savolainen, 2005).

Portland-clinker burning

Ground limestone is fed into preheating system, after which it is dropped into a rotary kiln. The cement clinker is burned in 100 meter long rotary kiln. In clinker burning, the ground raw meal is fed into a rotary kiln where it is calcinated and sintered to produce cement clinker. Limestone-, silica-, aluminum- and iron compounds are transformed into calcium compounds and sinter into cement clinker at 1400 °C temperature. The flame temperature should be at about 2000 °C, and the sintering zone should be around 1400-1500 °C. The clinker should be burned in oxidizing conditions. Therefore, an excess of air is required in the sintering zone of the rotary kiln.

The rotary kiln is a steel tube, with a length to diameter ratio of 10:1 to 38:1. The tube has two to seven (or more) support stations and is in 2,5 % to 4,5 % inclination. The kiln rotates 0,5 to 5,0 revolutions per minute. The slope and rotation of the tube makes the material to be transported slowly along the tube. The tube has to withstand very high temperatures, and therefore it is often lined with heat resistant bricks and the interior is equipped with chains, crosses and lifters to improve heat distribution. (Schorcht, et al., 2013) The cement is cooled with air to 200 °C temperature in the end of the kiln. The clinker resembles gravel at this phase. Clinker cooler have important function; to lower the temperature of the material for the downstream processes, and to recover as much heat as possible to the heating process. The heat is recovered by air preheating in the main and secondary firing of the kiln.

Cement grinding

Portland cement is produced by intergrinding clinker with sulfates, such as gypsum and anhydrite. Cements may include other aggregate material as well, such as granulated blast furnace slag, fly ash, pozzolanic ash and limestone. Addition of gypsum adjusts the bonding time of the cement. The quality of the cement can be adjusted by adjusting the ingredients of the clinker, fineness of the grinding and proportions of the aggregate materials. (Ohlström & Savolainen, 2005) The type of the grinding process depends on the type of cement being produced. The mills commonly have closed circuit, meaning they can separate cement with the required fineness from the material being ground, and return the coarse material to the mill.

6.4.2 Energy use and CO_2 emissions from cement manufacturing

Cement production process is flexible regarding fuels use. Different fuels are used in cement kiln firing, commonly coal, petroleum coke, fuel oil, waste fuels and natural gas. Table 6.4 represents fuel use in cement kilns in EU-27. Coal and petroleum coke are the most common fuels used in cement kilns in EU-27. The natural gas accounts for 1 % of fuel use. Besides fossil fuels, cement industry uses waste fuels and biomass.

Table 6.4: Fuel consumption expressed as percentage of heat generation by the cement industry in the EU-27 (CEMBUREAU, 2008)

Type of fuel [%]	2006
Coal (fossil)	18,7
Petroleum coke and coal (fossil) [4]	15,9
Fuel oil, including HVFO (fossil) [5]	3,1
Lignite and other solid fuels (fossil)	4,8
Natural gas	1
Waste fuels	17,9

Well-optimized cement production process consumes 2900-3300 MJ/t clinker. The energy consumption typically accounts for 30-40 % of the production cost (Schorcht, et al., 2013, pp. 100-101). The energy use accounts for 40 % of the emissions. Finnsementti’s factories in Parainen and Lappeenranta used coal, petroleum coke, ground rubber and waste oil fuels to heat its process in 2002. According to Finnsementti’s product manager Iiro Aro (Aro, 2014), it is possible to apply natural gas as fuel at Lappeenranta cement factory, but it is not economically viable at current

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[4] reported by EU-27 members.

[5] HVFO: high viscosity fuel oil

economic situation (2014). Ohlström & Savolainen (Ohlström & Savolainen, 2005) estimates, that changing fuel to natural gas in Lappeenranta factory would reduce annual emissions by 75 kt/a, from 366 kt/a in 2002.

Cement clinker is in contact with flue gas in the rotary kiln. Therefore, changing fuel affects the quality of the cement as well. Controlling the quality of the cement might result in additional expenses. (Ohlström & Savolainen, 2005, p. 113) However, in white cement production a non-emission fuel, such as natural gas, is required to avoid the contamination of the clinker, affecting the white clinker process. (Schorcht, et al., 2013, p. 27)

Majority of CO_2 emissions (60 %) originate from limestone, which cannot be reduced with current technology. The calcium carbonate ($CaCO_3$) burning (calcination) releases 0,44 tons of CO_2 and 0,56 tons ofcalcium oxide (CaO) for every ton of $CaCO_3$:

$$CaCO_3 \rightarrow CaO + H_2O \qquad (18)$$

In concrete drying process, some CO2 is bound to concrete. Some procents of CO2 is immediately absorbed into the concrete in skin drying, in reactions (19) and (20):

$$CaO + H_2O \rightarrow Ca(OH)_2 \qquad (19)$$

$$Ca(OH)_2 + CO_2 \rightarrow CaCO_3 \qquad (20)$$

The drying process takes very long time (approximately 50 years), and there is no estimation of total amount of CO_2 binding.

6.5 Glass industry

Glass is usually transparent material, consisting high proportion of silica (SiO_2). However, there is no common definition for glass, and the term is used for an inorganic material in a glassy state, which may be likened to a solid, but which has properties of highly viscous fluid, with neither crystalline structure nor distinct melting point. Structurally glass is similar to liquids, but in ambient temperature they behave like solids. The term glass is limited to inorganic substances and organic materials, such as transparent plastic, is not called glass.

Glass industry has several branches, including plate glass, container glass, household and art glass, and glass wool manufacturing. The EU identifies the following glass industry sectors:

- Container glass
- Flat glass
- Continuous filament glass fibre
- Domestic glass
- Special glass (without water glass)
- Mineral wool (with two divisions, glass wool and stone wool)
- High temperature insulation wools (excluding polycrystalline wool)
- Frits

The total production in 2005 in EU-25 was approximately 37,7 million tonnes (Scalet, et al., 2013). The glass industry in EU is diverse, both in the products made and the manufacturing techniques employed. The products vary from bulk consumer goods to construction materials and to crystal decanters and goblets. Manufacturing methods vary from manual methods (blowpipes and cutting) to completely automated product lines. The majority of products are made of soda-lime glass, consisting mainly of silicon dioxide (SiO_2), however other types of glass are used for some purposes (crystal, heat resistant, etc.).

Most float glass is used in building industry, which accounts 75-85 % of the output of plate glass industry. The most of the remainder in used in automotive industry. In Finland, Pilkington manufactures clear and green float glass for automotive industry, and some microfloat glass for laboratory use (Scalet, et al., 2013). Other companies manufacturing glass products in Finland are Iittala (container glass), Saint Gobain Isover (glass wool), Paroc (glass wool), Ahlström Kuitulasi (glass wool) and Karhulan lasi (glass containers). In this thesis, domestic and art glass process of Iittala, and float glass process of Pilkington are described. Pilkington Lahti factory is since closed, but it is described as a case study nevertheless, as float glass is one of the largest branches of glass industry, and Lahti glass factory can be considered a typical float glass manufacturing process.

6.5.1 Iittala

Iittala Group manufactures domestic and art glass in its Iittala. The main products are colored glass and crystal glass, which are manufactured by both automated methods and handicraft. These products accounted 60 % of the production. The factory was started in 1881, and employed 200 people in 2007 (Hämeen ympäristökeskus, Ympäristönsuojeluosasto, 2007). Iittala has previously owned Nuutajärvi and Humppila glass factories, but does not operate these factories anymore.

Process description

The Iittala domestic glass manufacturing includes manufacturing and melting the batch (raw material), glass forming and quality control, finishing and packing. Iittala glass factory has two manufacturing lines, automated Vesa line, and Vihtori where glass is made manually. The automated Vesa line includes one batch melting furnace, four glass manufacturing machines and four cooling furnaces. The manual Vihtori line includes one batch melting furnace, one day batch furnace, four large and three small crucible ovens and three cooling furnaces.

Automated line is continuous, where one product is usually manufactured for one day. Process can be adjusted during manufacturing if needed. Manual line is a hybrid of continuous and batch process; the production is flexible and the products can be made for short duration. Colored batches are used to manufacture colored glass.

Iittala glass manufacturing process has five glass forming lines.

Glass batch used to be manufactured in Nuutajärvi and transported to Iittala factory in 2007. However, the Nuutajärvi glass factory is not part of Iittala group anymore. Glass batch is stockpiled for couple of days. The factory stockpiles also propane, argon, acetylene, oxygen and hydrogen for maintenance purposes (Hämeen ympäristökeskus, Ympäristönsuojeluosasto, 2007).

Batch melting furnace

Batch melting furnaces are recuperative furnaces. The recuperators recover the heat from flue gases, which have 1300 °C to 1700 °C temperature. The batch is fed into the oven continuously, to keep constant load at the furnace. In many types of glass manufacturing, batch melting is a continuous process, which is operated for months (or years) without pauses; when the batch melting process is stopped and restarted, the first batches have usually to be discarded due varying quality (Ohlström & Savolainen, 2005). Iittala process is more flexible, and the production can be changed daily (Hämeen ympäristökeskus, Ympäristönsuojeluosasto, 2007).

Crucible ovens

In crucible ovens, air preheating is done in the bottom of the oven and the melting crucibles are on the top of the oven. Day batch furnaces are similar in design and have higher capacity. Day batch furnaces are loaded once a day.

Cooling furnaces

Glowing homogenous glass material is fed to forming machines via feeder channel. After forming process the temperature of the glass should be lowered slowly, to reduce crystallization and tension forming. Cooling furnaces are used to provide stable reduction of temperature. Cooling furnaces can be either batch or continuous, and operate at temperature range of 450-700 °C. (Malin, 1988)

6.5.2 Energy use and potential for power-to-gas

The glass making requires high temperature and is therefore energy intensive. The combustion therefore creates high amount of thermal nitrous oxides (NO_x) as well as carbon dioxide emissions. European Environmental Agency (EEA - NEC, 2008) estimates the total CO_2 emissions of EU-27 glass industry 22 million tons, representing 0,8 % of total EU emissions. The 15 % of total energy is consumed as electricity, 30 % as fuel oil and 55 % as natural gas. Natural gas is used as the main fuel in glass making as it can provide high enough temperature with minimal soot emissions, which could compromise the quality of the glass batch. Heat is transferred from flame to molten glass via gas- and particle radiation (when using natural gas as fuel). The radiation can be increased by letting the gas and combustion air mix slowly, which causes methane to crack, forming soot particles, which increase radiation. (Malin, 1988) Heat is transferred by convection from electric heaters.

Energy use in Iittala

Iittala glass factory uses natural gas in its furnaces and heat plant, and light fuel oil as auxiliary fuel. Some cooling furnaces use electricity. The plant is connected to natural gas network and Gasum has a natural gas station in the factory premises. Iittala's natural gas and light fuel oil consumption is presented at the table 6.5.

Table 6.5: Fuel consumption of the Iittala glass factory (Hämeen ympäristökeskus, Ympäristönsuojeluosasto, 2007)

	Process and volume [m^3/a]	Total energy [GJ/a]
Natural gas	Melting furnace Vihtori: 2 097 900 Melting furnace Vesa: 2 706 300 Heat plant and other processes: 779 800 Total: 5 700 000	199 000
Light fuel oil	Auxilary fuel	4 128

The factory has 2,1 MW power plant, which produces heat for spaces and water. Glass melting consumes 57 % of total energy use. Usually the glass melting consumes 75 % of total energy consumption of the factory (Hämeen ympäristökeskus, Ympäristönsuojeluosasto, 2007).

6.5.2 Former Pilkington Ltd. Lahti glass factory

Pilkington is a global glass manufacturer. Pilkington Plc. acquired Lahti glass factory in 1994, and the factory was closed in 2011. Other Pilkington's glass factories in Finland are in Laitila, Tampere, Ylöjärvi and Espoo. The company mainly produces plate glass and armor plate glass in Finland for vehicle and spare part markets. Lahti factory produces plate glass (float glass). Pilkington Marine (in Laitila) produces glass for cruise ships. Pilkington's Lahti glass factory process is described here even though the factory is not operational anymore, as it represents typical float glass process, and its process and energy use is available from the literature.

Glass mass is manufactured by melting the raw materials in 1600 °C temperature. In the melting process, the impurities and gas bubbles are removed from the glass mass. Heat is transferred from flame to molten glass via gas- and particle radiation (when using natural gas as fuel). The radiation can be increased by letting the gas and combustion air mix slowly, which causes methane to crack, forming soot particles, which increase radiation. (Malin, 1988) Heat is transferred by convection from electric heaters. Glowing homogenous glass material is fed to forming machines via feeder channel. After forming process the temperature of the glass should be lowered slowly, to reduce crystallization and tension forming. Cooling furnaces are used to provide stable reduction of temperature. Cooling furnaces can be either batch or continuous, and operate at temperature range of 450-700 °C. (Malin, 1988)

Pilkington manufactures glass from:

- 58 % Quartz sand
- 18 % Lime Na_2CO_3
- 16 % Dolomite
- 2 % Feldspar
- 5 % Ground limestone
- 1 % sodium sulfate, Na_2SO_4

Pilkington also uses 30 % recycled glass from its own sites (no external sources for recycled glass are used).

Melting capacity of the factory was 80 kt/a and net production capacity 60 kt/a. Glass types were continuously made for several months, usually four months at once. When the type was changed, the new mass was crushed for some time after manufacturing, which was the reason that net production was significantly lower than the production capacity. Approximately 75 % of the manufactured glass products were usable. 35 % of produced glass was clear, 53 % green and 12 % microfloat glass. Thickness of the glass was between 1-3 mm. (Ohlström & Savolainen, 2005)

Energy consumption varies depending on the type of glass manufactured. Melting furnace can be operated with natural gas and oxygen burners, without electricity. However, this increases the total energy consumption of the process. The specific energy consumption is lower with electricity than natural gas, it reaches the middle of the glass mass better, and the glass mass can be maneuvered with the electric heating. Lahti glass factory consumed approximately 16 – 18 million nm^3/a. (Ohlström & Savolainen, 2005)

Lahti glass factory used mainly heavy fuel oil until 1987, when it was renewed to use natural gas. The advantages of using natural gas in glass melting are clean combustion, adjustability and possibility to burn with high temperature atmosphere. Glass manufacturing process is continuous and technical changes in the furnace are difficult. Therefore, fuel substitution is challenging and costly. In long term, electrification of the process may be possible. Waste heat recovery could be possible, either to be used in the factory or in district heating network. Lahti glass factory used to feed 21 GWh/a into district heating network. (Malin, 1988) (Ohlström & Savolainen, 2005)

The raw materials were melted in 1600 °C temperature. The melting furnace used both natural gas and electricity as fuels, natural gas being the main energy source. The power of the melting furnace was 26,5 MW. 6,9 MW was regenerated from the melting furnace flue gases. 40 % of all electricity was used in the melting furnace’s main boost. Second largest electricity consumer was the tin bath. Electricity would have been more efficient than natural gas, but using natural gas with oxygen was economically optimal. Expenses in glass production were roughly divided to 1/3 to salaries, 1/3 to raw materials and 1/3 to energy consumption, and therefore the energy expenses were significant cost factor for the Lahti glass industry. (Ohlström & Savolainen, 2005)

7 DISCUSSION AND CONCLUSIONS

In this chapter, the potential for SNG is estimated for the processes described in the chapter 6. Moreover, the CO_2 emission reduction potential is assessed in the sector at EU or global level.

This thesis work assesses the technical potential for SNG in Finnish process industry. It should be noted however, that the natural gas economics at the time of the writing (2015) does not favor the use of gaseous fuels. Instead, coal is still the primary fuel in e.g. steel manufacturing. To promote the zero-emission fuels, such as power-to-gas generated SNG, either the carbon emissions should be priced significantly higher, or the whole industrial energy production should follow another type of energy pricing model than currently.

All the processes described in chapter 6 use natural gas as fuel, or the (partial) fuel substitution to natural gas is possible. Electrification can improve the energy efficiency and reduce the carbon emissions of some of the processes, such as ironmaking (EAF) or glass manufacturing. Cement clinker process is challenging to electrify, as radiation, not convection, is the primary heat transfer method in the rotary cement kiln. As hydrogen is used as a raw material in oil refining process, its substitution is not possible with current technologies, and therefore there is potential for renewable hydrogen in the future also in fossil fuel manufacturing, although reduction of fossil fuels is preferable.

7.1 Steel manufacturing

According to Danloy et al. (2008) and Hooey et al. (2010) the coke used in blast furnace process as reducing agent could be replaced with other reducing agents, such as natural gas, from 300-350 kg/t hot metal to 200 kg/t hot metal. Moreover, use of reducer material of biological origin is studied in the steelmaking process (Suopajärvi, 2014). By combination of bio-coke and SNG reducer materials, CO2-neutral steelmaking process is theoretically technically possible.

Figure 7.1 depicts the power-to-gas integrated into blast furnace-basic oxygen furnace (BF-BOF) steelmaking process, and direct reduced iron (DRI) processes. In BF-BOF process the gas is injected to tuyère layer of the blast furnace, acting as a reducer. In DRI process, the gas is injected into shaft furnace, or fluidized bed, whichever is used for the iron reduction.

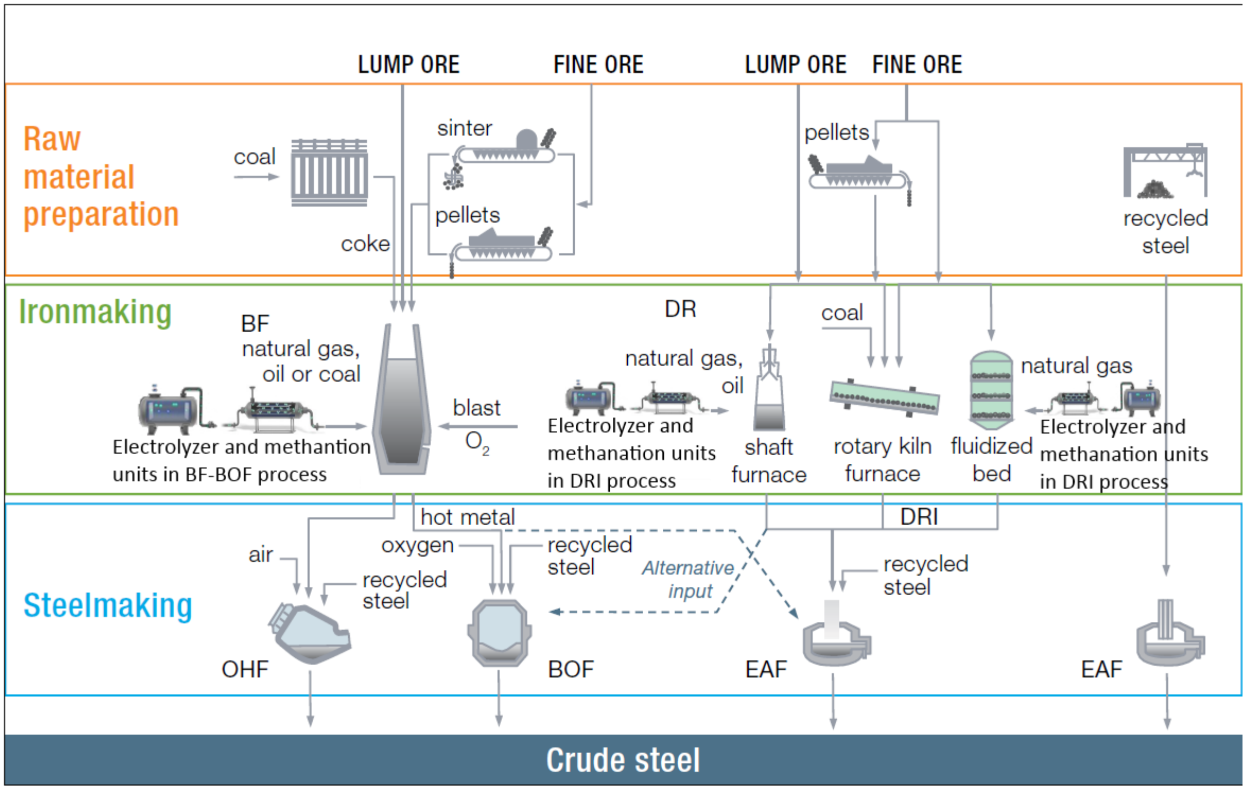

Figure 7.1: Possibilities for power-to-gas integration into different steel making processes (World Steel Association, 2011)

Approximately 70 % of the steel is produced via BF-BOF route. If the emissions could be reduced by 50 % by using SNG injection (see appendix 1), and as the BF-BOF process accounts for 88 % of global steel industry emissions, use of SNG (CO and H_2 gases) as reducing agent in BF-BOF process could theoretically reduce global steel industry emissions by approximately 40 %. As steel industry causes 7 % of global CO_2 emissions, this reduction is significant in effort to reduce global CO_2 emissions (Laplace Conseil, 2013).

7.2 Oil refining

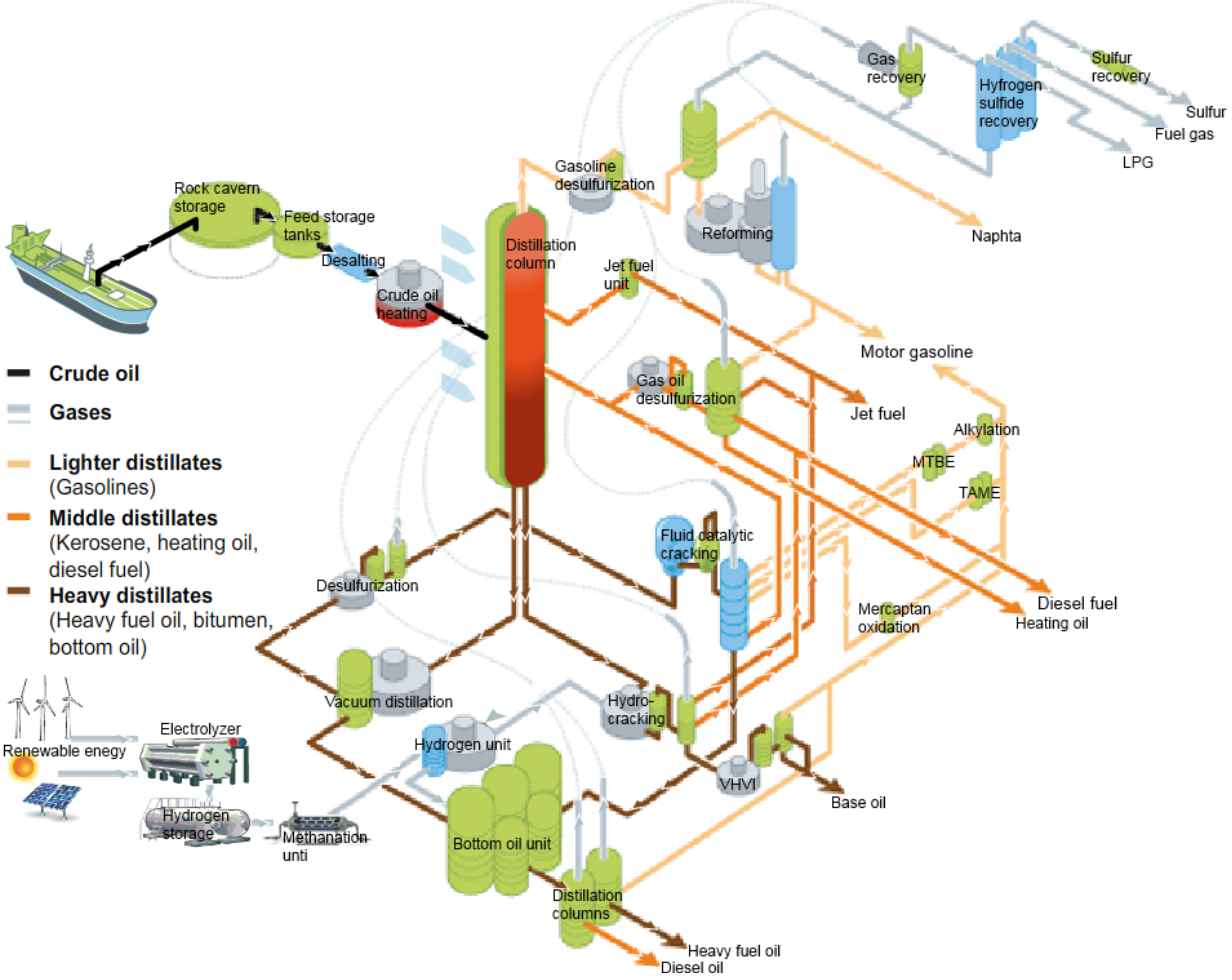

Figure 7.2: Neste Oil refinery process, with power-to-gas integrated (Neste Oil)

Figure 7.2 depicts the electrolyzer unit in Neste Oil’s refinery process. Hydrogen is used as raw material in the oil refinery process. Neste oil utilizes natural gas in cracking of complex hydrocarbons into lighter ones in thermal- and hydrocracking, in removal of sulfur, nitrogen and metals from crude oil, and in hydration of aromatic compounds (Suominen, 2014; Riistama, et al., 2003). The annual consumption of natural gas of Neste Oil operations in Finland is 0,5 billion normal cubic meters (Neste Oil , 2011), having 5,4 TWh in energy content, or 15 % of total annual natural gas consumption in Finland (3,3 billion Nm^3).

Neste Oil's direct emissions were 3,7 million tons CO_2 and indirect emissions 0,165 million tons CO_2 in 2011 (Neste Oil , 2011) (“gate-to-gate” emissions). Furthermore, the end use of Neste Oil’s fuel products resulted in 46,6 million ton emissions (“tank-to-wheel” emissions) (see Appendix 1). The emission reduction from SNG use would result in reduction of 1 million tons CO_2 per year, resulting in 2 % reduction of total CO_2 emissions from the Neste Oil’s refineries and end-use emissions. Natural gas is used as raw material in oil refining process, and therefore the CO_2 reductions of SNG use is compared to emissions from product fuels here. It should be noted however, that some chemicals (ethane, propene, benzene) are directed to the Borealis petrochemical plant next to Neste Oil’s Porvoo refinery. Mass balances of these plants are intertwined, and therefore the 1 million ton CO_2 reduction potential is for Neste Oil’s refinery and Borealis petrochemical plant together. The hydrogen is used as a raw material in oil refining process. Therefore, the investment for power-to-gas technology is limited to electrolyzer and hydrogen storage. The hydrogen unit in Neste Oil’s refinery already has a methanation unit.

Neste Oil has developed renewable diesel fuel production process called NEXBTL. The raw material for NEXBTL diesel is vegetable oil and waste animal fats. The diesel requires no modification or special precautions to the engine. NEXBTL has been produced in Neste Oil Porvoo refinery since 2007, and is nowadays produced in Neste Oil's refineries in Singapore and Rotterdam as well. Neste Oil claims the NEXBTL diesel cuts carbon dioxide emissions by 40 to 60 percent, taking into account the entire production and transportation chain. (Institut für Energie und Umwelt (ifeu), 2006)

7.3 Cement manufacturing

Cement production is energy-intensive process, as large mass of cement requires high temperatures (2000 °C), to convert the raw materials to cement clinker. The process is flexible regarding the fuel use, and therefore large variety of fuels are currently used in cement production (see table 6.4). However, according to Finnsementti’s Esa Suhonen (2015), electrification of cement clinker burning process is not possible with current technology due insulation properties of clinker mass. Currently, natural gas accounts for 1 % of fuels used in cement industry in EU-27.

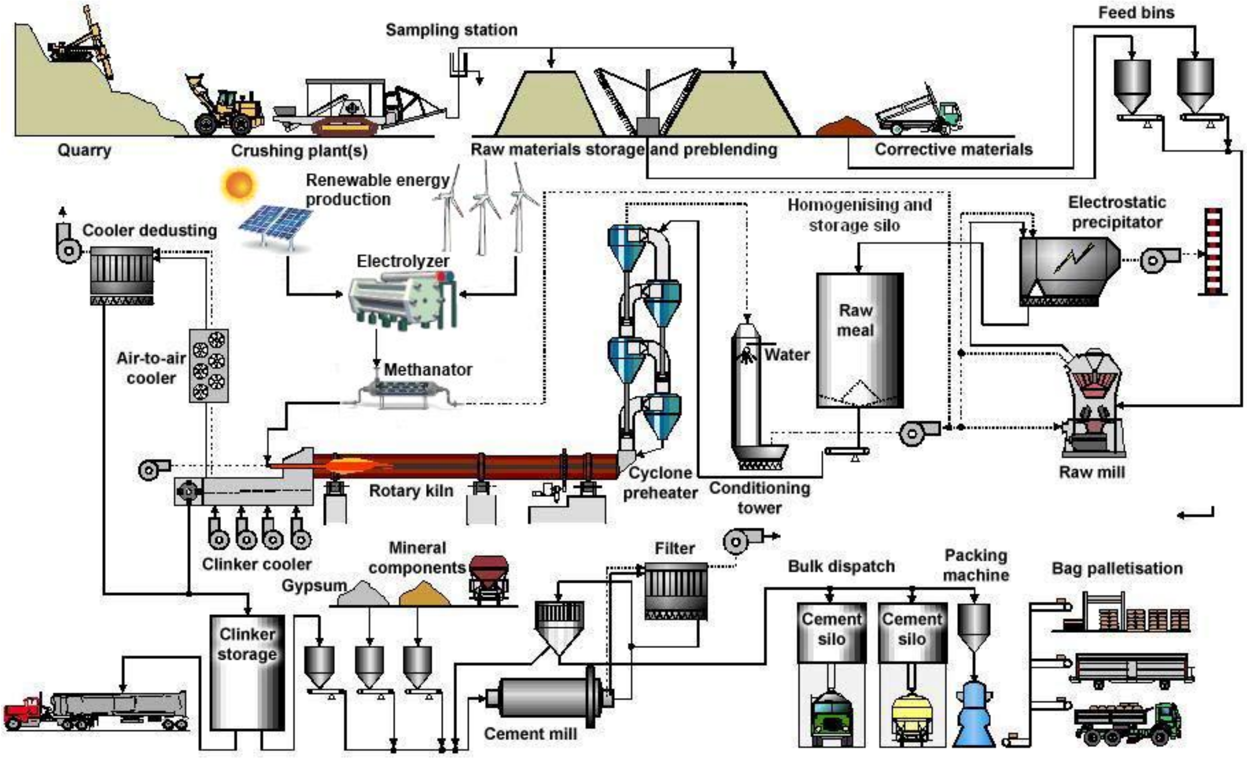

Figure 7.3: Cement manufacturing process, with power-to-gas integrated (Schorcht, et al., 2013)

Figure 7.3 illustrates the integration of power-to-gas process to cement manufacturing process. Fuel switching in cement industry requires both burner optimization and changes in chemical composition of the cement clinker. Methane has weaker heat radiation than solid and liquid fuels, and therefore the flame should be closer to the material. Furthermore, the flue gases are in direct contact with material, and thus affects the chemical composition of finished clinker. These effects should be addressed if fuel switching to gaseous fuels is implemented.

The cement clinker burning process requires fuel combustion, and the process is flexible regarding fuel use. Due this flexibility, theoretically all fuel emissions from cement clinker burning could be eliminated by switching to zero-emission fuels such as SNG, biomass or waste fuels, reducing total emissions from cement manufacturing by 40 %. However, the economic situation should be changed significantly to favor the non-emission fuels. Remaining 60 % of the emissions from fuel use originate from raw material and therefore cannot be eliminated with current technology.

7.4 Glass manufacturing

The glass making requires high temperature and is therefore energy intensive. The combustion therefore creates high amount of thermal nitrous oxides (NO_x) as well as carbon dioxide emissions. European Environmental Agency (EEA - NEC, 2008) estimates the total CO_2 emissions of EU-27 glass industry 22 million tons, representing 0,8 % of total EU emissions. The 15 % of total energy is consumed as electricity, 30 % as fuel oil and 55 % as natural gas.

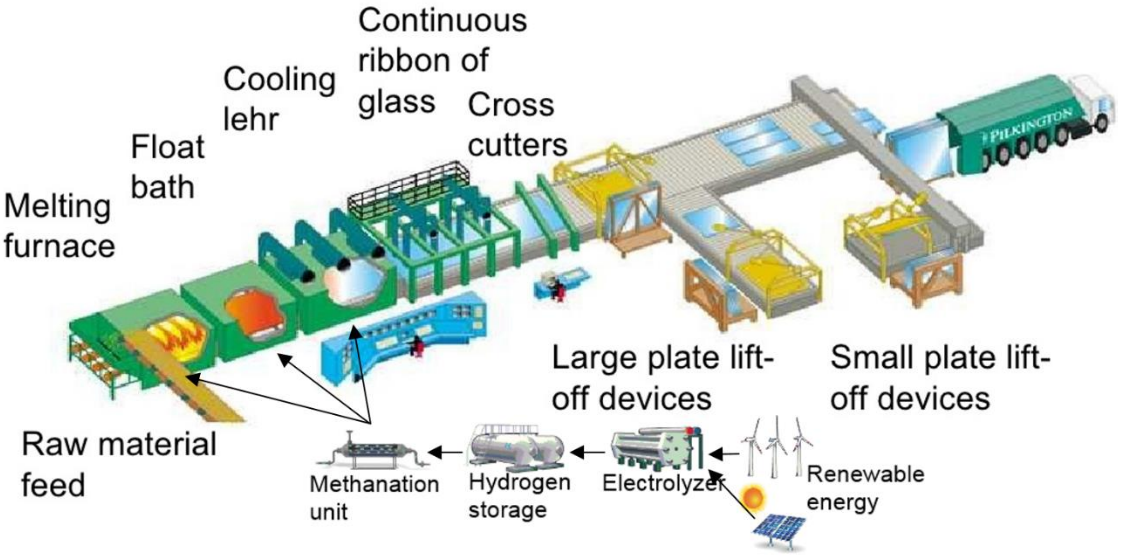

Figure 7.4: Pilkington's float glass manufacturing process, with power-to-gas integrated (Pilkington)

Pilkington's Lahti glass factory used to emit approximately 34,4 kt/a from natural gas and 9,5 kt/a from raw materials, which accounted for 80 kt/a glass batch melting and 60 kt/a net production. Figure 7.4 illustrates the integration of power-to-gas generated SNG into the float glass manufacturing process. The melting furnace, float bath and cooling processes all used to consume natural gas in a typical float glass process.

Iittala household and art glass factory has significant potential to reduce emissions by SNG fuel switching, as almost all of the emissions originate from primary fuel, the natural gas, excluding the emissions from used electricity. Total emissions from Iittalan Lasi factory are on average 11 000 tCO_2/a. The natural gas consumption causes 97 % of CO_2 fuel emissions. Therefore, the emission reduction from switching to SNG is 97 % (see appendix 3), excluding emissions from consumed electricity and raw materials.

Theoretically, most of the fuel emissions from glass industry (22 million tons in EU-27) could be eliminated by fuel switching and electrification, as the most common fuels are fuel oil and natural gas which both can be replaced with process electrification and/or power-to-gas generated SNG. However, some of the emissions from glass industry originate from raw materials when the carbon dioxide in lime, dolomite and calcium carbonate is released. This accounted for 22 % of the total CO_2 emissions from Lahti glass factory (Ohlström & Savolainen, 2005).

7.5 Result table

Table 7.1: CO2 emission reduction potential by SNG substitution in studied sectors

Industry	Scope/ Process	Max. technical emission reduction potential by SNG substitution	Number of industrial plants in EU-27*	Industry capacity in EU-27 [Mt]*	Industry annual emissions in EU-27 [Mt CO_2]*	CO2 emission reduction estimation [Mt CO_2]
Steel manufacturing	BF-BOF	Up to 33% [1]	100	100	253	up to 30…50
	EAF	0…0,33 % [2]	230	70		process dependent[2]
Oil products	Final fuel and plastic consumption	2 % [3]	100	755 [4]	2415	up to 50
Cement	Portland-clinker burning	1 % [5]	268	268	205	2…8 [5]
Glass	All EU glass processes	55 % [6]		38	22	5

*Source: EU BAT reference scenarios

[1] Coke replacement with SNG as reduction material, as in Nogami et al. study, plus coal replacement as fuel. Further emission reductions possible by synergies with bioreducer technology.

[2] Natural gas is used in rolling processes in steel plants. Percentage is relation of natural gas use in total energy use of EAF steel plant.

[3] CO_2 from final product consumption. Further emission reductions possible by synergies with biofuels.

[4] EU total oil consumption.

[5] 1 % equals to current share of natural gas in EU-27. Technical potential for significant increase. Increase to 10 % SNG fuel share would result in 8 Mt CO_2 emission reductions.

[6] EU-27 natural gas fuel share in glass industry.

8 SUMMARY

Power-to-gas is a process of transforming electricity to hydrogen gas by electrolysis, and optionally into methane by methanation. The hydrogen or methane gases can then be stored and utilized in energy production, either in power generation, mobility or industrial sector. This thesis assessed the potential for power-to-gas generated SNG and hydrogen in Finnish process industry. First, the storage options for renewable energy were introduced, qualities of methane and hydrogen were described, the power-to-gas technology was introduced, and an overview of Finnish and Nordic energy sector and natural gas use was described. Then the key natural gas utilizing industry sectors were identified, which were steel, oil refining, cement manufacturing and glass industries.

Technical potential for SNG were discovered in all studied sectors, and SNG technology can have significant contribution in decarbonizing the industrial energy generation. Economic potential and business models should be assessed the next, as well as calculating the detailed mass and energy balances of the studied processes. The studied industrial sectors have key processes that may be difficult to electrify, due requirements for combustion (such as high enough temperature), or process optimization. Therefore carbon neutral fuels, such as SNG or biomass, are required in these sectors. SNG and hydrogen gases may be essential in decarbonizing the studied industrial sectors.

8.1 Answers to research questions

This thesis aimed to answer the following questions. The research questions are revised in this chapter.

Which are the largest industrial processes currently that apply natural gas as fuel in Finland?

Main industrial consumers of natural gas in Finland are the pulp- and paper industry, chemical industry, and steel industry sectors. Glass industry sector was included in this study due its high natural gas share. Ironmaking, oil refining, cement manufacturing and glass manufacturing processes were described in detail in the chapter 6. There are large number of natural gas consuming industrial applications in the industries in question as well as other industries that were excluded from this thesis.

Which industrial processes apply hydrogen as fuel or raw material in Finland?

Oil refining and chemical industries uses natural gas as raw material for hydrogen. In oil refining, the hydrogen is used for cracking, metal, sulfur and nitrogen removal. Oil refining is the largest consumer of natural gas at the moment in Finland. Hydrogen is also used as a raw material for large number of other chemical processes which were excluded from this study. Hydrogen is not used as fuel at significant amounts at the moment.

Is there potential for electrification of the processes that currently apply natural gas as fuel?

In this thesis, electrification potential was discovered in steel industry and glass industry. Steel can be made from virgin material via blast furnace-basic oxygen furnace process route, which currently uses coal as fuel and reducing material, or from scrap via electric arc furnace process route. Switching to electric arc furnace process would reduce energy intensity of the industry. However, using scrap as raw material may lead to quality issues, and therefore a share of iron should come from virgin material for high quality special purpose steel. Oil refining is a chemical process, which uses hydrogen as raw material and therefore electrification of the process is not possible. However, the electrification of the end use (mobility) is possible and the technology is available. Cement manufacturing is not possible to electrify with current technology. Glass industry processes often use natural gas and electric heaters in parallel. The electric heating could be increased, if it is technically and economically favorable.

Is there potential for other processes to use gaseous fuels that apply other fuels at the moment?

Most industrial processes which require high heat input could apply natural gas as fuel if it would be economically feasible. However, process adjustment might be required, due changed heat transfer, reduced particle emissions and possible effect to raw material due absence of flue gases. At the moment, the costs of different fuels do not favor the use of natural gas.

In steel industry, partial substitution of coal in iron ore reduction process is possible with hydrogen and/or carbon monoxide, in both blast furnace/basic oxygen furnace (BF-BOF) process and direct reduction processes, as concluded by Nogami et al. Further substitution for renewable fuels is possible using bioreducer technology. Cement manufacturing with using SNG as fuel is possible, but process optimization is required. In glass production, natural gas is already used as fuel to large extent.

How much the fuel substitution to SNG would reduce emissions of the studied processes?

Emission reduction potentials were estimated in chapter 7 and summarized in the table 7.1

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APPENDIX

Appendix 1: Estimation of CO2 from blast furnace fuels in Nogami et al. study

Emissions from direct combustion, with NG [CO_2eq]				
Operation	Coal	NG	LPG	Total
Standard	1865,951	0	8,12278	1874,073
Natural gas injection	1174,264	328,2003	1,37934	1503,844

This table represents the emissions from direct fuel use. However, the material and energy flows in BF-BOF ironmaking is much more complex than direct combustion. Therefore, this table shows 20% reduction in emissions per ton hot metal when changing to natural gas injection, but Nogami et al. estimated 33 % reduction.

Emissions from direct combustion, with SNG [CO_2eq]				
Operation	Coal	NG	LPG	Total
Standard	1865,951	0	8,12278	1874,073
Natural gas injection	1174,264	0	1,37934	1175,643

As power-to-gas gas is carbon neutral, emissions from natural gas use are eliminated. This leads to 17 % further CO2 reduction per ton hot metal.

Appendix 2: Estimation of indirect emissions from Neste Oil’s product fuels (tank-to-wheel emissions)

Product (Neste Oil , 2011)	Production [ton/a]	Emissions per ton [tCO2eq] (UK Department of Energy & Climate Change, 2012)	Total emissions [tCO2eq]
LPG	457300,00	3,00	1373681,641
Gasoline	4265500,00	3,15	13423528,5
Diesel and light fuel oil	8389800,00	3,19	26746682,4
Heavy fuel oil	1066184,00	3,23	3441641,952
Bitumen	490100,00	0,00[6]	0
Sulfur	133300,00	0,00[5]	0
Solvents	267400,00	3,19[7]	852471,2
Other	231200,00	3,19[6]	737065,6
Total			**46575071,29**

Appendix 3: Estimation of CO2 reduction potential in Iittala glass factory

Average annual CO2 emissions [tCO2/a]	Natural gas consumption [m3/a]	Emissions per m3 natural gas [t/m3]	Total emissions from natural gas [tCO2/a]
11000	5700000	0,002028	11559,6
	Light fuel oil consumption [m3/a]	Emissions per m3 light fuel oil [t/m3]	Total emissions from light fuel oil [tCO2/a]
	120	2,642191	317,0629

Light fuel oil (auxilary fuel) accounts 2,7 % of annual emissions. Therefore, the emission reduction from NG → SNG switch is 97%.

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[6] Non-fuel raw material. Estimated zero emissions due end use.

[7] Estimated the same as light fuel oil