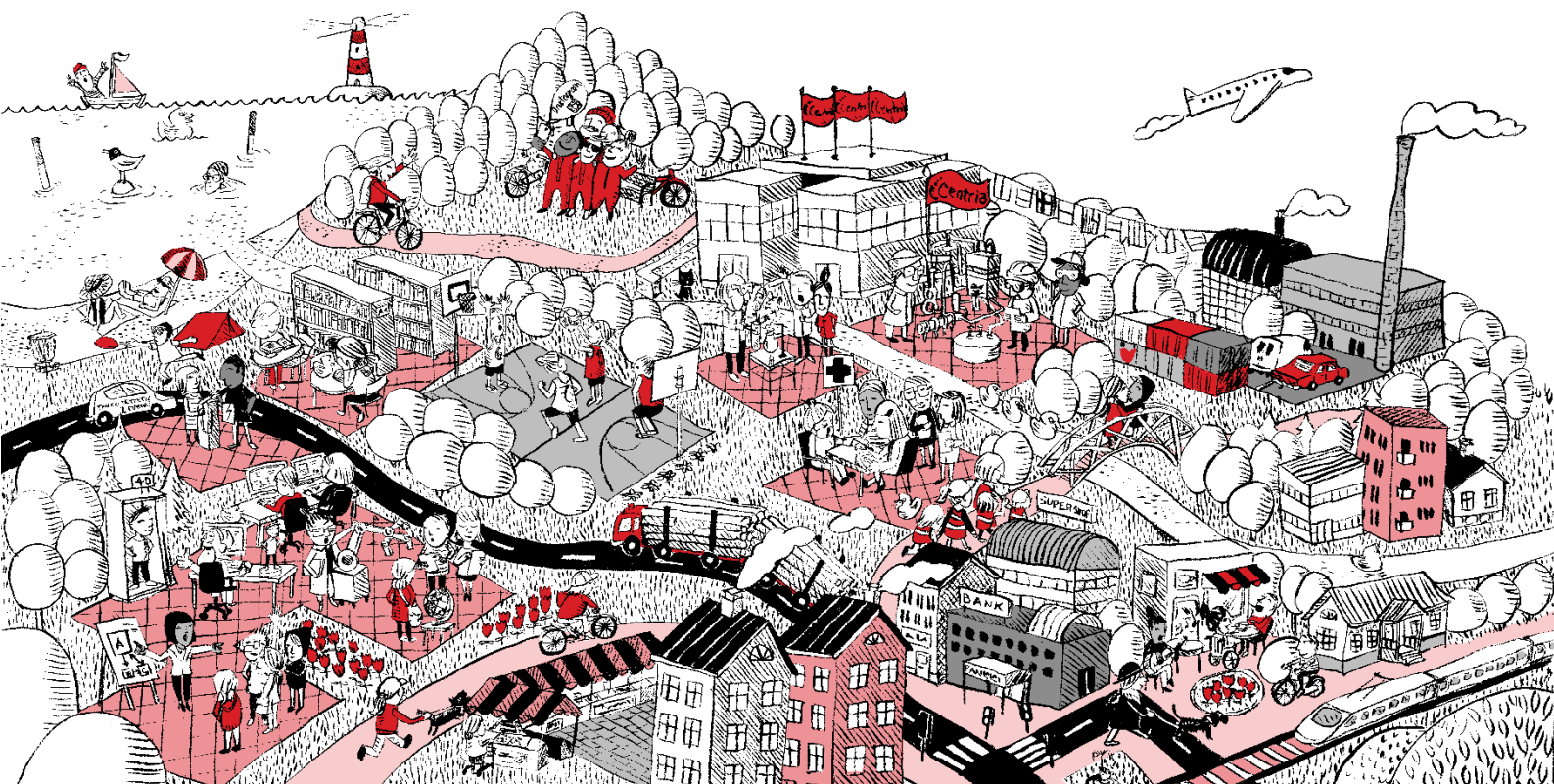


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HEAT BALANCE OF THERMOCATALYTIC DECOMPOSITION PROCESS

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ABSTRACT

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Instructor representing commissioning institution or company Laura Rahikka		
<p>This thesis was commissioned by Hycamite TCD Technologies, located in Kokkola, which focuses on using a technology, thermocatalytic decomposition process, to break down methane into hydrogen and carbon. The goal was to study the heat balance of their pilot plant and to form an Excel file and a descriptive figure of the balance.</p> <p>The theoretical part focuses on different methods of heat transfers and heat exchangers, on energy balance of an open system and different parts of it. There is also a short section describing the optimizing of a system. The paper has the description of the calculations performed on the practical section of the thesis.</p> <p>The practical part of the paper was completed mainly on Excel as this thesis is heavily practical in the mathematical way. The picture was drawn on AutoCAD 2020. Both can be found in the confidential sections in the Appendices. The background data for the calculations are confidential as well.</p>		
Key words carbon, heat exchangers, heat transfer. hydrogen, methane, thermocatalytic decomposition process, thermodynamics		

CONCEPT DEFINITIONS

GHG

Greenhouse gas, a substance, which increases the greenhouse effect in the atmosphere

IEA

International Energy Agency

TCD

Thermocatalytic decomposition

ABSTRACT
CONCEPT DEFINITIONS
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1 INTRODUCTION

The topic of this bachelor's thesis came from a start-up located in Kokkola, Hycamite TCD Technologies, the CEO of the company, Laura Rahikka, was the instructor representing commissioning company. From Centria University of Applied Sciences, Staffan Borg was the supervisor. The goal was to collect data of the TCD process and to study the heat balance of the system, and to create an Excel file, which could be used to model the changes in different process modes.

As the background data and conclusions have information of the company's own data, this thesis does include confidential sections in Appendices, which are not public in the online version of the paper but shared with both supervisor and instructor. These include a table with the data of the system, the Excel with all the calculations and a picture, where all the data is connected into a simplified flow sheet of the process.

The second chapter of the thesis describes the background of the thesis, including some information about hydrogen production in general and why is it necessary. It also describes the TCD process and its chemical reaction. The chapter closes by a description of the commissioning company, Hycamite TCD Technologies. From there, the next 3 chapters are introducing theory behind the thesis, chapter 3 being about heat transfer, its three different methods and most typical types of heat exchangers. Chapter 4 deals with energy balance in a system, and chapter 5 focuses on ways to optimize a system's heat balance in order to make it more profitable and environmentally sustainable.

The sixth chapter deals with the practical part of the thesis. The chapter itself only deals with the formulas used in the calculations, and the Excel file and the picture drawn are part of the confidential section. The last, seventh, chapter brings the paper into a conclusion and gives some suggestions for the future.

2 BACKGROUND

While hydrogen is thought to become even more important in the future, due to its versatile possibilities in fields such as energy and chemical, it is currently not made in a sustainable matter. Most of the current hydrogen is made from natural gas, 59%, and coal, 19%, the latter used mainly in China. Rather than recovering the GHGs, these side streams are released into the atmosphere, as shown in Figure 1. This is because the price of the hydrogen rises while Carbon Capture, Utilisation and Storage, CCUS, is used, because its more complicated and energy intensive process. Because of this, in 2020, with a production of 90 Mt, the amount of GHG released was as high as the emissions of Indonesia and Britain combined, 900 Mt. (International Energy Agency 2019, 37; 2021, 108.)

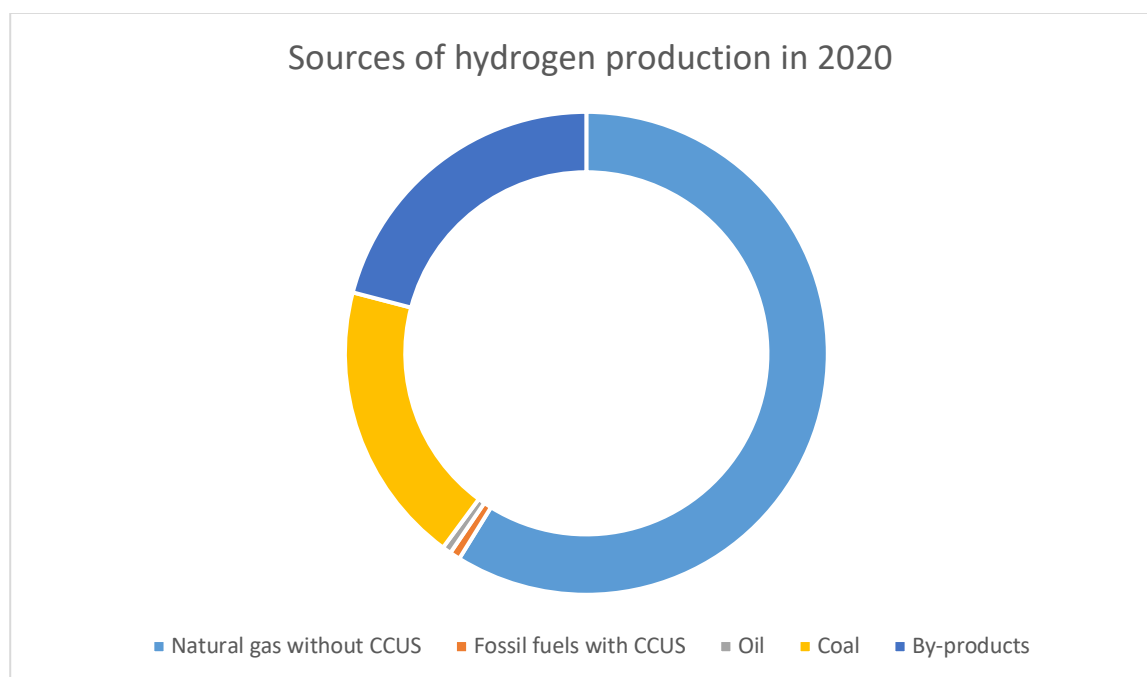


FIGURE 1. Hydrogen production by sources in 2020 (adapted from International Energy Agency 2021, 108)

Out of the 90 Mt of produced hydrogen, most is used in the industrial fields. 45 Mt of H₂ is used in the chemical field, and 5 Mt in steelmaking in a single process with direct reduced iron process. In the chemical field, hydrogen is mainly used in producing ammonia and methanol. Most of the remaining hydrogen is used as feedstock and reagents, in refining field. In the IEAs scenario for net zero by 2050, demand for hydrogen is set to rise as high as 530 Mt, with demand in industry rising to 140 Mt. With

the use of hydrogen rising in transportation, the demand of the field could rise from just 20 kt to 100 Mt. (International Energy Agency 2021, 43-45.)

Hydrogen can be categorized by the source and the type of production (Figure 2), giving the produce a colour code. The most common type of hydrogen is grey, made from the fossil fuels, with no CCUS used in the process. In blue hydrogen, while the source is fossil fuel, the CO₂ will be abated, making it a more sustainable one. The cleanest form is green hydrogen, which is made from renewables, such as biogas, giving no direct emissions. Between blue and green categories, there is turquoise hydrogen, which like green hydrogen has no direct emissions, which uses fossil fuels as the source. (Lumbers, Agar, Gebel & Platte 2021.)

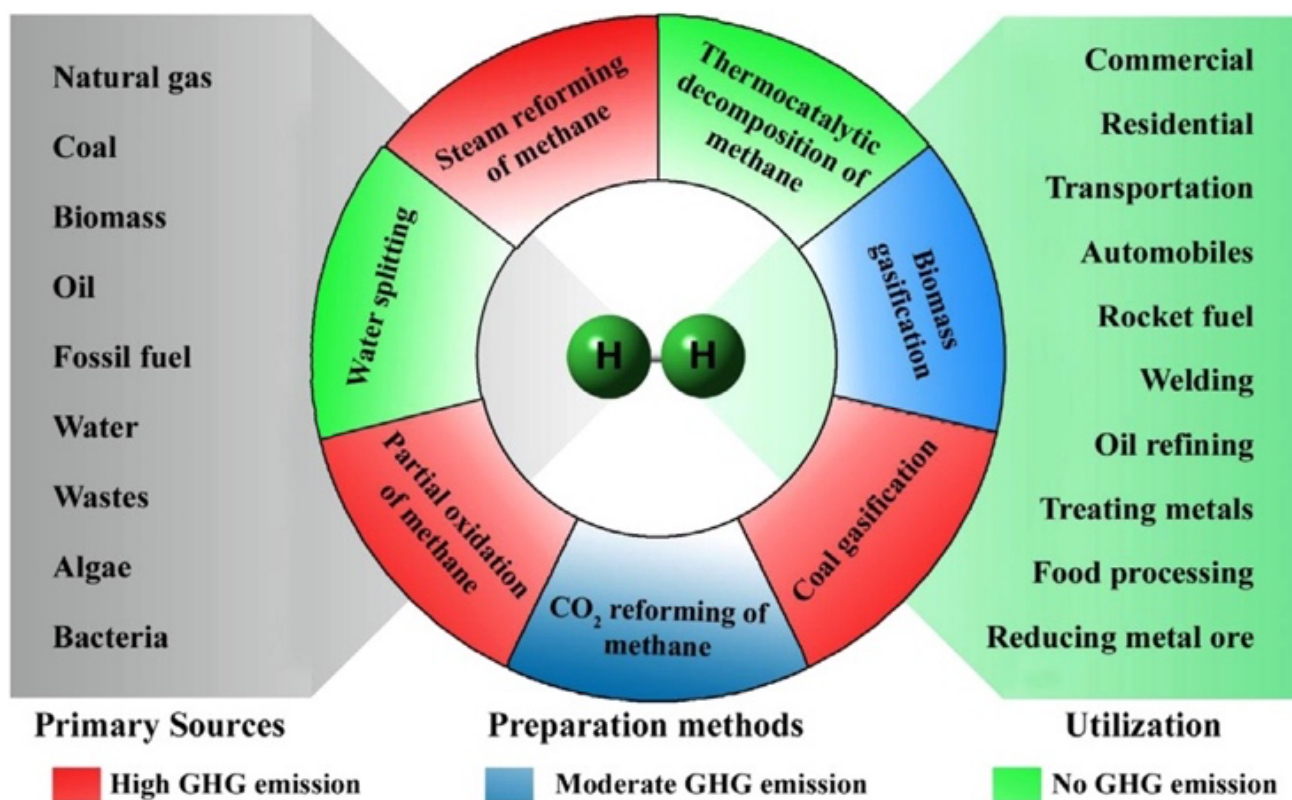


FIGURE 2. Primary sources, preparation methods and utilization of hydrogen (Ashik, Wan Daud & Abbas 2015)

2.1 Thermocatalytic decomposition of methane

Thermocatalytic decomposition of methane, TCD, is a way of producing GHG free hydrogen. This is done by breaking the bonds between carbon and hydrogen by introducing heat into the system, resulting into the formation of hydrogen gas and solid (nano) carbon, as shown in Formula 1. The bonds within the methane molecule are strong, 440 kJ/mol, making it quite inactive, and making the required temperature as high as 1470 K, with the absence of a catalyst. For this reason, to decompose methane, a catalyst is used. (Ashik, Wan Daud & Abbas 2015.)



TCD has five steps, the first one being the adsorption of methane into the catalyst. In the second step, the bonds between carbon and the hydrogen begin to break, one bond at the time, before on the third step the formed hydrogen atoms form the usual hydrogen molecules, which then leave the catalyst in the gas form. In the final two steps, the formed carbon forms nanofibers, which grow on the catalyst, making the structure of the fibers, as the form of the fibers can be affected by the shape of the catalyst. (Ashik et al. 2015.)

The catalyst is usually a metal based, even while using a carbon-based catalyst would be more readily available, cheaper and they have many advantages, such as higher stability in high temperature. Still, iron-based catalysts could be more cost-effective, due to larger catalytic activity and the tendency to produce more valuable forms of carbon, instead of amorphous types, produced in use of carbon-based catalyst. The issue with the use of catalytic reaction is the deactivation of it, mainly occurring as the formed carbon particles surround the active part of the catalyst, fouling it. While regeneration can be used, there is a rise of emissions, making it more cost-effective to prevent the fouling by optimizing the reaction conditions to suit the used catalyst as well as possible. (Lumbers et al. 2021.)

2.2 Hycamite TCD Technologies

Based in Kokkola, Finland, Hycamite is a start-up company, which was launched in 2020. Their mission is to provide a technology, which can be used to produce emission-free hydrogen from natural gas, and produce pure carbon as a side stream, which can also be profitable. It is also possible to start using biogas as a source, making the process a carbon sink (Hycamite 2022). As of early 2022, the

company has started to test the technology in a pilot-sized plant and plans to start to build a demo-sized plant to Kokkola Industrial Park (KIP), starting on summer 2022 (Keskipohjanmaa 2022).

Currently the company is working on three projects, the first one being Thermal Decomposition of Methane, in which they partner with University of Oulu, and has been supported by the European Social Fund and the European Regional Development Fund. The project focuses on developing TCD equipment for the use of small and medium sized companies. Second project, supported by the same funds is Carbomite, which focuses on building a small-scale pilot plant to KIP. Lastly, the third project, supported by Business Finland, aims at international markets. The company is currently planning a demo-scale plant, which will have a yearly capacity of 2000 tons of hydrogen, and which already has the investment decided on. (Hycamite 2022.)

3 HEAT TRANSFER

Energy has many different forms, such as heat, kinetic and chemical. Heat is a form of energy that can transfer to and from a non-insulated system, with no work done or matter transferring. A related term, specific heat, tells how much energy, heat, needs be transported into the substance for the temperature to rise by one degree. The SI unit of heat, like other forms of energy, is joule, named after James. P. Joule, who discovered that heat was not a caloric substance, contrary to the theory of the time. (Çengel 2006, 4.)

Thermodynamics is a field dealing with the connections between heat and matter, and it is the base of many branches of engineering, mainly chemical and mechanical engineering. Even while the applications are different according to the branch, all thermodynamic formulas follow four universal laws, named with a number, from zeroth to third. As the entire universe influences everything in it, it is practical to form a boundary, inside which the laws are then applied. Inside the boundary, a thermodynamic system is formed, and outside it lies the surroundings. A system can be closed, open or insulated, depending on the type of the boundary. (Wijeyesundera 2011.)

The first law of thermodynamics determines that energy cannot be made or destroyed, it can only change form. This means that in a steady system, the energy that enters a system is equal to the energy leaving it, though it can be in a different form. According to the second law, heat can only naturally move from a higher temperature to a lower one (Wijeyesundera 2011). There are three different ways that heat travels, which are conduction, convection, and radiation. All three modes still require a temperature difference, and free heat transfer stops after an equilibrium has been reached, as continuing would break the second law of thermodynamics due to the lack of driving force. (Çengel 2006, 17.)

3.1 Conduction

Conduction can happen in all states of matter, requiring a medium of matter to take place. In the method, heat transfers through matter, in a solid as vibrations and movement of free electrons, and in fluids as diffusion and inner collisions of the particles forming gas or liquid. The amount, rate, of conduction depends on the thickness, the area of the material and the temperature difference, which affects the transfer, and lastly the material itself. Each material has a temperature dependant constant of thermal conductivity, k , which tells, how well it can work as a conductive material, the constant being

higher in materials as metals, and lower in insulators. Rate of heat transfer by conduction can be calculated using formula 2, where ΔT is the temperature difference between the thickness, Δx , of the material and the area of transfer is marked as A . (Çengel 2006, 18.)

$$Q_{cond} = -kA \frac{\Delta T}{\Delta x} \quad (2)$$

3.2 Convection

If two mediums are interacting with each other, and the other one is a moving fluid, on top of conduction, the fluid movement of the medium will moreover improve the transferring of heat and higher speed of the fluid increases the rate of convection. If the flow is increased by an external source such as a pump, the convection is said to be forced. Natural/free convection happens without the effect, only working with density difference caused by the temperature difference, which then must be significant enough to overcome resistance which the air causes. Convection also has a role during the change of state, such as boiling. (Çengel 2006, 26.)

The rate of convection is calculated by using Newton's law of cooling (Formula 3), in which h is a convection heat transfer coefficient, which is not a property of the fluid, but the value is measured in an experiment, with typical values varying from 2-25 W/m²K in free convection in gasses to as high as 100000 W/m²K during boiling. As in conduction, the area of the transfer is also taken into account, this time as A_s rather than simply A . The way of calculating the temperature difference is by finding out the temperature at the surface, T_s and far from it, T_∞ , as the temperature is often going to be higher at the surface of the mediums, though this is not always the case. (Çengel 2006, 26.)

$$Q_{conv} = hA_s(T_s - T_\infty) \quad (3)$$

3.3 Radiation

Unlike conduction and convection, radiation does not require intervening medium, and thus is the only form of heat transfer which can travel in the vacuum, such as space. All material above 0 Kelvin, absolute zero, emit radiation which is formed by photons, electromagnetic waves. Emission of heat hap-

pens by thermal radiation in every part of the material. The part, which is most important is the emission which happens on the surface due to the rays emitted by the inner parts will be absorbed inside the material. The theoretical maximum of radiation can be calculated using Stefan-Boltzmann law, formula 4, where σ is the Stefan-Boltzmann constant, $5.670 \cdot 10^{-8} \text{ W/m}^2\text{K}^4$, and area, A_s , and temperature, T_s , are measured from surface. (Çengel 2006, 28.)

$$Q_{emit,max} = \sigma A_s T_s^4 \quad (4)$$

This law only works on a theoretical blackbody, and all real surfaces emit a smaller number of photons. In this case, emissivity of the surface is considered by multiplying the formula with ε which varies between 0 and 1 depending on the material and temperature of the surface. Depending on the material, radiation can be as little as just a few percentages of the maximum amount, meaning that if the system also has forced convection with a high amount of heat transfer radiation might be irrelevant in some cases, while in some systems it might have a significant amount of system's thermodynamics. (Çengel 2006, 28.)

3.4 Heat Exchangers

The basic concept of heat exchangers is to put two fluids of different temperature in contact with each other, separated by a solid wall. As the modes of heat transfer show, the heat will then move from the hot fluid to colder one. Heat exchangers are categorized into two main types, recuperators and regenerators. Recuperators have the usual two fluids and heat transfer happens on a surface, with no moving parts. (Vepsäläinen, Pitkänen & Hyppänen 2012, 135.)

For heat exchangers, the theoretical overall heat transfer coefficient, U ($\text{W/m}^2\text{K}$), can be calculated by using Formula 5, where h_o and h_i are the heat transfer coefficients of the outer and inner part of the inner pipe and λ tells the thermal conductivity of the material between the fluids. A_i and A_o are the area of the inner pipe, A_i in the inside and A_o outside, and A_m is the heat transfer area, which tells the logarithmic mean of the calculated areas (formula 6), and the last geometric part of the equation, s , is the thickness of the inner tube. A_1 can be either one of the areas, depending on the need. (Green & Perry 2008, 11-4.)

$$\frac{1}{UA_1} = \frac{1}{h_o A_o} + \frac{1}{h_i A_i} + \frac{s}{\lambda A_m} \quad (5)$$

$$A_m = \frac{A_o - A_i}{\ln\left(\frac{A_o}{A_i}\right)} \quad (6)$$

When the overall heat transfer coefficient is known, the heat transfer rate of the system can be calculated by using Formula 7, in which A describes the area of heat transfer, and (ΔT_{lm}) is logarithmic mean temperature difference (formula 8), which is calculated using $(\Delta T)_1$ and $(\Delta T)_2$, the temperature difference at the ends of the exchanger. (Green & Perry 2008, 11-4.)

$$q = UA_m(\Delta T_{ln}) \quad (7)$$

$$(\Delta T_{ln}) = \frac{(\Delta T)_1 - (\Delta T)_2}{\ln\left(\frac{(\Delta T)_1}{(\Delta T)_2}\right)} \quad (8)$$

3.4.1 Concentric tube heat exchangers

Concentric tube (or double pipe) heat exchanger is the simplest type of heat exchanger, consisting purely out of two pipes, one within the other, where two fluids are made to flow either parallel or at counterflow to each. These modes of flow are described in Figure 3. Because of its simplicity, it is also the cheapest one for small scale needs. (Vepsäläinen et al. 2012, 135.)

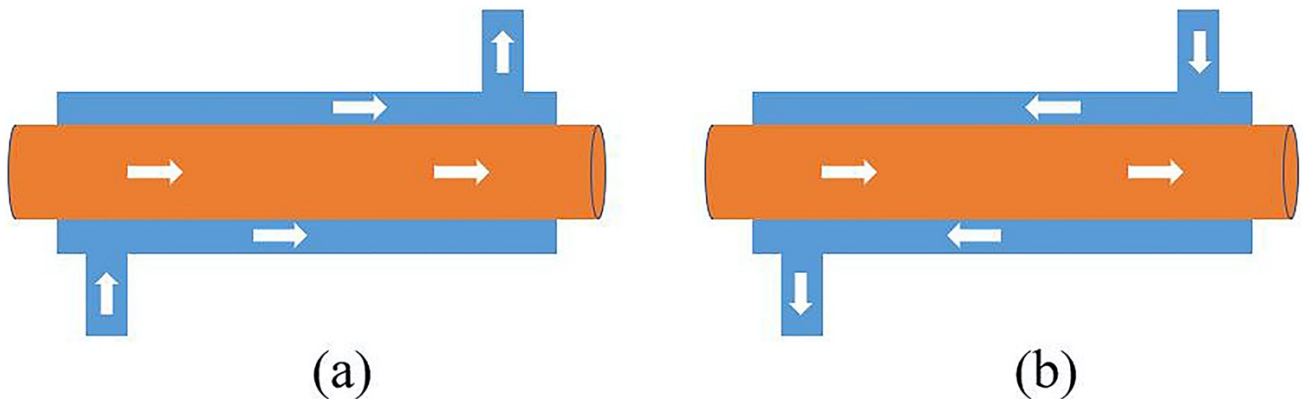


FIGURE 3. a) Parallel flow and b) Counter flow (Li, Wang, Han, Li, Yang, Guo, Liu, Zhang, Zhang & Jiang 2021)

3.4.2 Plate heat exchanger

Plate heat exchanger, PHE, consists of metal plates, which are held together by a frame which can be opened to inspect the gaskets within. The plates form two different channels for the two fluids, by adding gaskets between the plates, meaning that they are in no point in direct contact with each other. Each plate has a hot fluid and a cold fluid on each side, making a large heat transfer area. As seen in Figure 4, the plates are also corrugated, which creates turbulence in the fluids which further heightens the heat transfer coefficient. (Alva Laval 2022.)

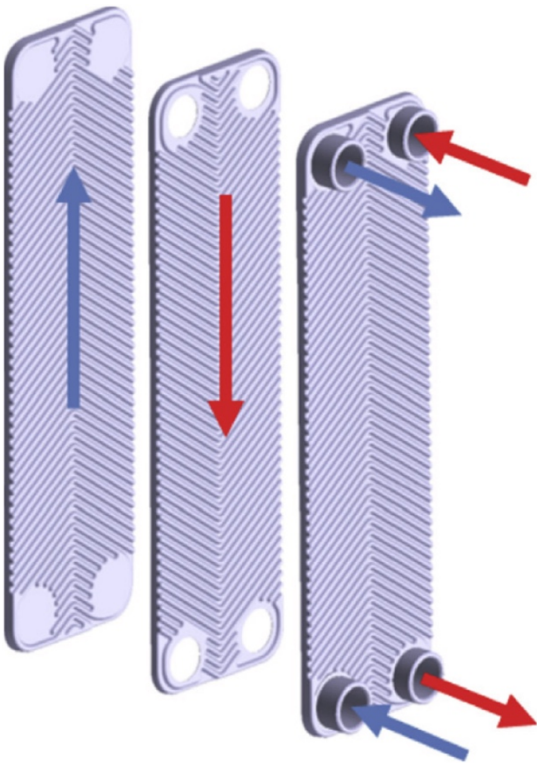


FIGURE 4. The flow of fluids in the PHE exchanger. (Zhang, Zhu, Mondejar & Haglind 2019)

While plate heat exchangers have many advantages, such as low cost, reliability and they are easy to modify, by adding or taking off plates their disadvantages are poor effectiveness in higher pressure, causing them to leak (Alva Laval 2022). PHE exchangers show most effective results for heat transfer in liquid-to-liquid applications, for the cold and hot streams are then both in almost the same pressure (Çengel 2006, 612).

3.4.3 Shell-and-tube type of exchanger

The most used heat exchanger in industrial scales is the shell-and-tube exchanger contains within it several small sized tubes, in which one of the fluids flows. In the surrounding shell the other fluid flows. The common structure of shell-and-tube type is shown in Figure 4. The exchanger also has baffles, which force the fluid in the shell to move around the tubes, making the heat transfer more efficient. These types of exchangers tend to be relatively heavy and with great size making them unusable in some application. (Çengel 2006, 611).

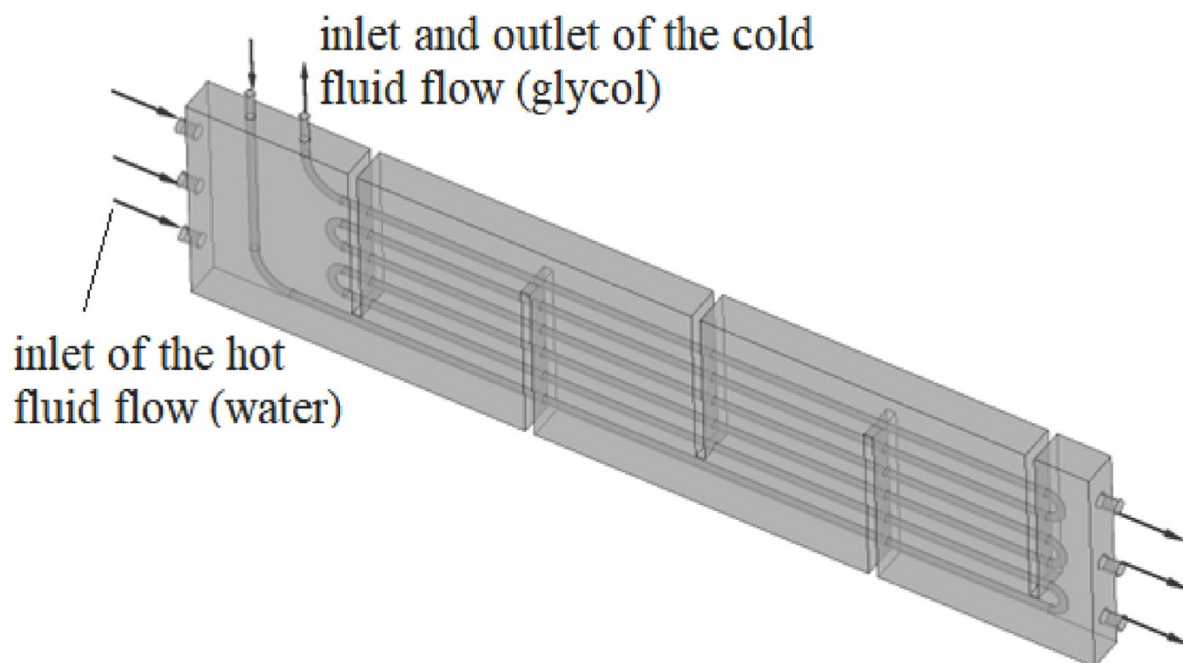


FIGURE 5. Structure of shell-and-tube type of heat exchanger. (Slimene, Poncet, Bessrour & Kallel 2022)

4 ENERGY BALANCE

As the first law of thermodynamics states, energy cannot disappear, only transfer form. This means that all energy entering a system must be equal to the amount of energy leaving it. By finding out the difference between energy flows, a need for heating (or cooling) can be calculated. In a closed system, where there is no mass flow, the energy flow can be calculated from formula 9 (Hirvijoki 2021). As is seen in the formula, only the heat added to the system (ΔQ) and the work done by the system (ΔW) are considered.

$$\Delta U = \Delta Q - \Delta W \quad (9)$$

In an open system, where there is not only flow of energy, but of matter (FIGURE 6), the change of inner energy depends also on the energy content of the matter, which can change in a system (Hirvijoki 2021). In the following part, the mass of the matter has been considered by the change of mass, Δm , while later in the calculations mass flow is used instead.

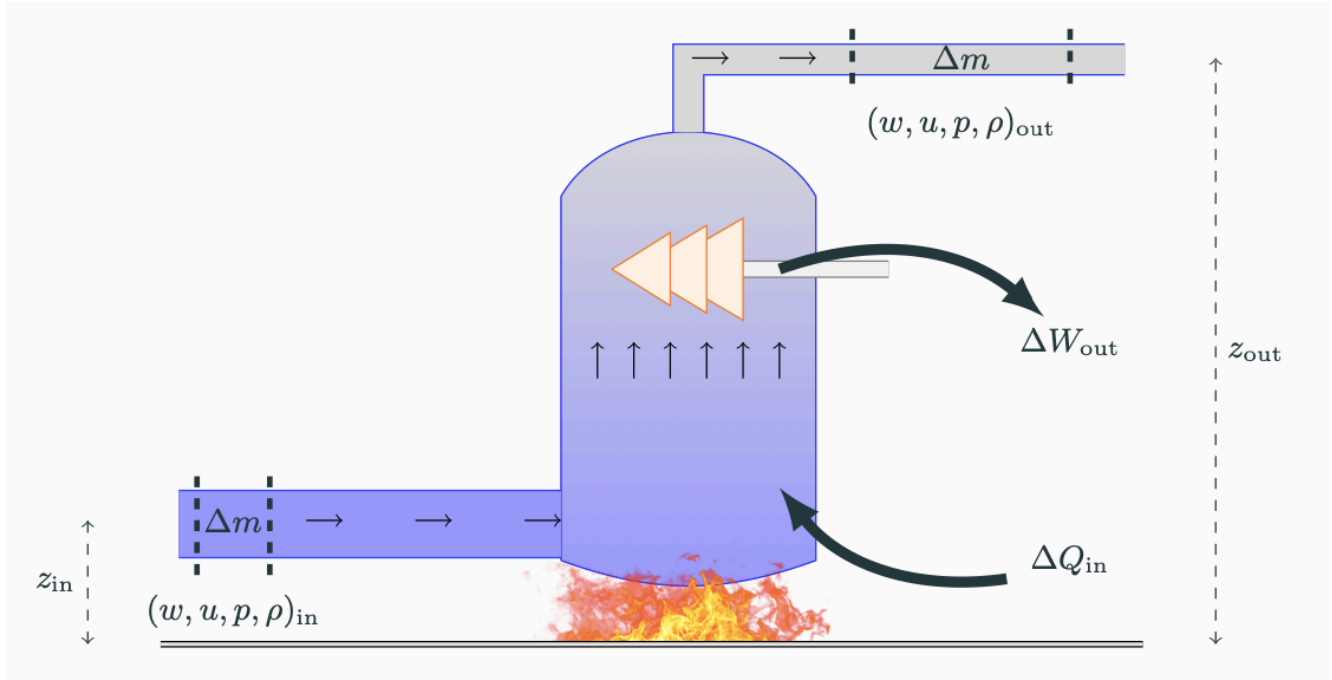


FIGURE 6. Energy flow through an open system (Hirvijoki 2021)

4.1 Potential and kinetic energies

Potential energy depends on the mass, as seen in Formula 10, but also the distance of the entry/exit point of the matter from the ground or other zero position decided on, and the gravitation. As the mass is in movement, it also has kinetic energy (Formula 11), which stays stable if the speed does not change. In the formulas, Δm defines the change of mass in kilograms, z is the height of the mass from a selected 0-point, and w is the speed of the mass, while g is the gravitational acceleration, circa 9,81 m/s². (Hirvijoki 2021.)

$$E_{pot} = \Delta m g z \quad (10)$$

$$E_{kin} = \frac{1}{2} \Delta m w^2 \quad (11)$$

4.2 Inner energy

The change of inner energy of matter, ΔU , can be calculated for a reaction, if the reaction and the change of enthalpy, ΔH is known. As the change of enthalpy is commonly told in a specific temperature, it can first be changed into another temperature using Formula 12, where Δc_p is calculated using the specific heat capacities of the material, before and after the chemical reaction. The Formula 12 only works in steady pressure, when only the temperature difference is taken into account. (Lampinen & Seppälä 2008, 161–163.)

$$\Delta H(t, p) - \Delta H(T_0) = \Delta c_p (T - T_0) \quad (12)$$

After finding out the change of enthalpy in the desired temperature, this information can then be used to calculate the ΔU by using the Formula 13, if one or more of the reacting substances are in a gaseous form. In it, $\Delta n(g)$ is a term, which determines the change of moles in gas form which with the R (gas constant, circa 8,314 J/Kmol) and temperature can be seen as the amount of enthalpy, energy, which is used by the gas in order to gain volume, as work. (Lampinen & Seppälä 2008, 161–163.)

$$\Delta H = \Delta U + \Delta n(g)RT \quad (13)$$

Once the change of inner energy is known, in kJ/mol, the amount of energy needed to enter the system can be calculated by Formula 14, in which u is ΔU , but in J/kg, so it needs to be modified by calculating the mass of a single mole of the material. (Lampinen & Seppälä 2008 161–163.)

$$E_{in} = \Delta mu \quad (14)$$

4.3 The energy balances

In an open system, the energy balance can be calculated by using Formula 15, in which h_{out} and h_{in} tells the enthalpy of the mass flow, bringing the inner energy into the equation. P_{out} is the amount of work done by the system in watts. In the beginning of the chapter, formula 9 was introduced. When compared these two equations together, it can be seen that they are otherwise identical, except for the mass flow in the open system. (Hirvijoki 2021.)

$$m (g (z_{out} - z_{in}) + \frac{1}{2} (w_{out}^2 - w_{in}^2) + h_{out} - h_{in}) = Q_{in} - P_{out} \quad (15)$$

5 THE SYSTEM OPTIMIZING

As some parts of a process require the addition of heat and some need to be cooled down, it is possible to regenerate the energy from one stream to another using heat exchangers. By doing so, and not simply relying on heating and cooling from outside sources, such as hot and cold water, the energy consumption of the entire process can be limited to minimal, which helps to lower the overall cost and environmental impacts of the plant. (Smith 2016, 457.)

The first step to optimizing the heat exchangers is to recognize the energy streams of the process, by for example creating a table, where the need for heat or cooling has been calculated and the streams named and/or numbered according to the flow sheet's information to make them easily understandable (Smith 2016, 458). An example of a table is given in Table 1.

TABLE 1. An example of stream data for heat exchangers (adapted from Smith 2016, 458-459)

Stream	Type	Supply Temperature	Target Temperature	ΔH	Heat capacity flowrate CP
1	Hot	200 °C	40 °C	20 MW	0,125 MW/K
2	Cold	20 °C	160 °C	10 MW	0,071 MW/K

The right column shows the CP, heat capacity of the stream, and it can be either calculated by dividing the enthalpy change by temperature change, or by multiplying the mass flow rate by the specific heat capacity of the fluid. If put in a plot, this would be the slope of the curve. (Smith 2016, 458.)

As heat transfer requires a difference in temperature for the exchange to be effective, a minimal difference is chosen by considering the cost of heating from outer source, and the cost of transferring heat within the system. Calculating this requires the knowledge of both the price of steam and cooling water, as well as the price of heat exchangers suitable for the requirements of the process. When the minimal temperature is known, the streams can be drawn in a temperature-enthalpy graph with the cold stream ending at the minimal temperature difference point. On its left side, the amount of cooling water can be seen, and on the right side, at the end of the hot stream, is the needed steam. (Smith 2016, 458.)

In the usual case of there being more than 1 of each type of flows the streams are listed, and instead of drawing them separately they are connected as a composite hot stream and cold stream. They are then added to a graph, finding the pinch point (Smith 2016, 461). This pinch technique (FIGURE 7) is useful and often valuable, but since the process at hand only has single streams, pinch technique was not used.

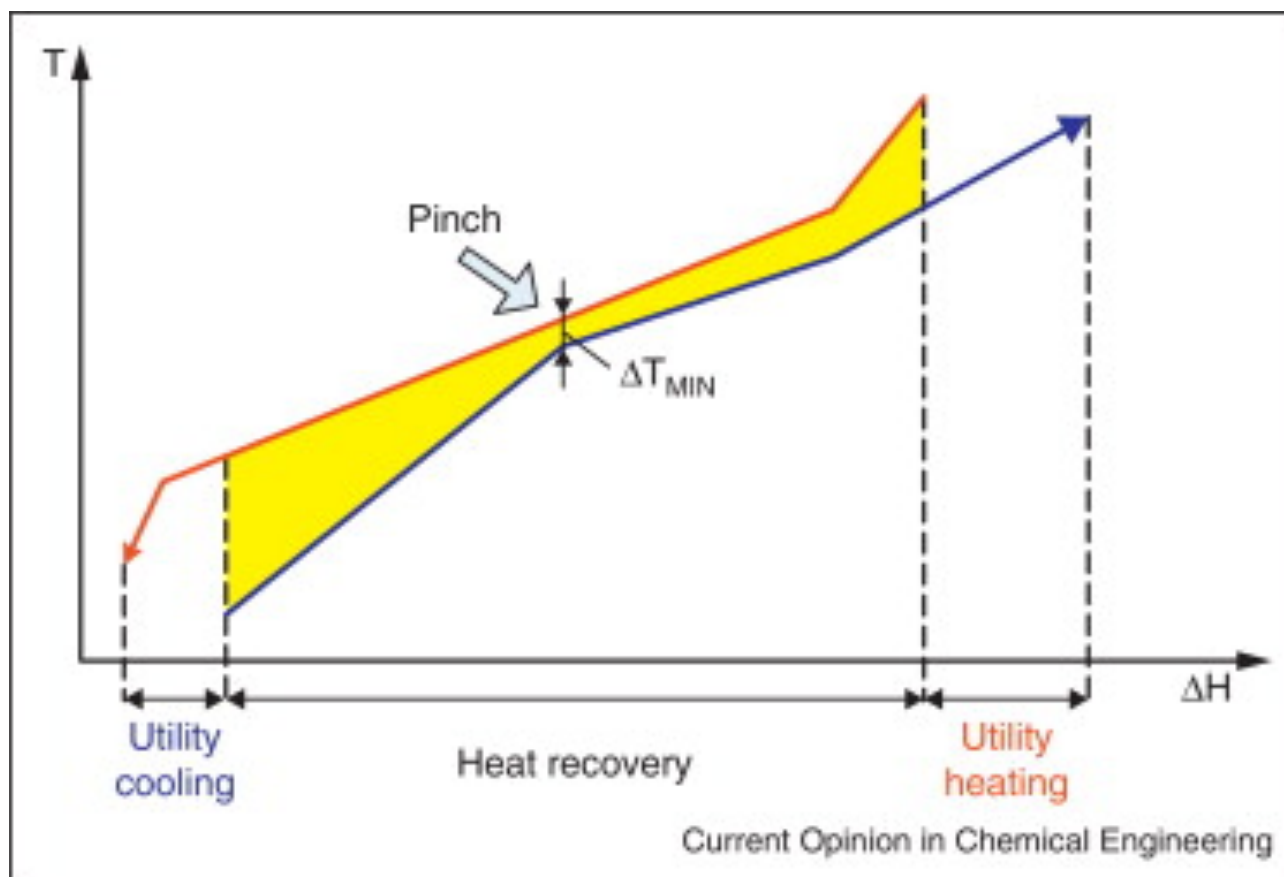


FIGURE 7. Example of the pinch technique (Klemeš & Kravanja 2013)

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APPENDIX 1

Collected data

APPENDIX 2

Heat flow

APPENDIX 3

Excel