MSc Program
Renewable Energy Systems

# Carbon capture and utilisation in the cement industry: A technological and economical assessment of carbon capture and electrolysis technologies and its implications on energy demand.

A Master's Thesis submitted for the degree of
“Master of Science”

supervised by
Ao.Univ. Prof. Dipl.-Ing. Dr. Reinhard Haas

Mag. Peter Martin Eiler

00550665

Vienna, 27.04.2022

# Affidavit

I, **MAG. PETER MARTIN EILER**, hereby declare

1. that I am the sole author of the present Master's Thesis, "CARBON CAPTURE AND UTILISATION IN THE CEMENT INDUSTRY: A TECHNOLOGICAL AND ECONOMICAL ASSESSMENT OF CARBON CAPTURE AND ELECTROLYSIS TECHNOLOGIES AND ITS IMPLICATIONS ON ENERGY DEMAND.", 70 pages, bound, and that I have not used any source or tool other than those referenced or any other illicit aid or tool, and
2. that I have not prior to this date submitted the topic of this Master's Thesis or parts of it in any form for assessment as an examination paper, either in Austria or abroad.

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## Abstract

To reduce global warming, the reduction of greenhouse gas emissions is crucial. In many cases, this cannot be achieved directly through the substitution of fossil fuels by renewable energy sources. Carbon capture and utilization projects can provide a viable solution, in particular for the cement industry, where carbon dioxide emissions are immanent to the production process of clinker.

The objective of this master thesis is to investigate and discuss the carbon capture and hydrogen production technology, as proposed by the carbon capture and utilization demonstration project ‘Carbon-to-product-Austria’. To achieve this, the cement production as well as carbon capture and hydrogen production technologies will be evaluated in order to derive technological and economic implications for the potential implementation in the entire cement industry in Austria. Furthermore, the electricity demand for the production of renewable hydrogen via water electrolysis will be calculated to allow a comparison with the current national renewable energy strategy in Austria.

The existing research projects support the technology choices made in this demonstration project, as monoethanolamine-based amine scrubbing is the only carbon capture technology available, that provides a sufficient technological maturity to be applied in an industrial scale demonstration project. However, existing alternative technologies provide viable alternatives, especially from an economic perspective, once the technological maturity is further progressed. The available hydrogen production technologies via water electrolysis also differ in their respective technological maturity, but can provide viable applications, depending on the respective production requirements and locally available electricity and heat sources.

The energy required for the renewable hydrogen demand of such carbon capture and utilization pathways cannot be covered by the existing renewable energy targets for the year 2030 as stated by the current Austrian climate and energy strategy. Therefore, an evaluation of a potential increase of the renewable energy targets and further renewable energy development in Austria need to be considered, or alternatively ways to import the renewable hydrogen from suitable geographies need to be discovered.

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# 1. Introduction

## 1.1. Motivation

To reduce global warming, the reduction of greenhouse gas (GHG) emissions is crucial. This can be achieved through renewable energy sources, for example by the substitution of fossil fuel fired power plants by renewable electricity sources in the electricity sector, and in the transport sector, with battery electric or fuel cell electric vehicles instead of gasoline or diesel driven combustion engines. But besides these sectors, there are also industrial processes, that emit GHG emissions, that cannot be reduced by means of a fuel switch. The cement industry is a good example for this issue, as only the emission caused by the kiln fuel can be avoided by switching to a renewable energy carrier. But there is also carbon dioxide (chemical formula $CO_2$) as a by-product of the chemical conversion in the clinker production, where limestone, which has the chemical formula $CaCO_{3,}$ is converted to lime, which has the chemical formula CaO. To effectively lower global GHG emission, technological possibilities to avoid or mitigate $CO_2$ emissions immanent to the clinker process need to be further developed and exploited.

One way to achieve this, is with the help of carbon capture and usage (CCU) technology pathways, where $CO_2$ emissions are captured to be further used in power generating or industrial processes: Lafarge, Verbund, OMV and Borealis are conducting a joint carbon capture and utilisation project at a cement production site in Mannersdorf. The goal of this project is to capture $CO_2$ from the flue gas emissions of the largest cement plant in Austria, and transforming the $CO_2$ with the help of green hydrogen eventually into renewable plastics, i.e. polymers (polypropylene and polyethylene), as well as secondary products. The companies are currently developing a demo scale project, using 10,000 tonnes of $CO_2$ per year, to test the technical and economic aspects. For the operation of the demo project, 10% of the total $CO_2$ emissions, i.e. 1,500 $t_{CO2}$ per year of green hydrogen will be processed. If successful, an expansion of the demo project to a full-scale plant design, to capture and utilise the total $CO_2$ emissions at the cement plant in Mannersdorf is envisioned, if technically and economically feasible.

The motivation behind this master thesis is to discover and assess the envisioned technology pathway of the ‘Carbon-to-product-Austria’, project, which acronym is C2PAT, based on existing research projects. The focus is put on technologic and economic aspects, specifically to discuss further implications in project implementations and cost reduction potentials provided by existing research projects.

## 1.2. Research objective

The objective of this master thesis is to investigate and discuss the carbon capture and hydrogen production technology, as proposed by the C2PAT demonstration project, as introduced in chapter 2.3, based on technological and economic criteria. This will be performed based on existing research projects on carbon capture and utilisation projects in the cement industry, specifically to gain insights and draw conclusions regarding efficiency and applicability of each process step compared to alternatives technologies. In an economic assessment, the impact of carbon capture technologies on the production costs of cement will investigated as well as hydrogen production costs depending on the respective energy supply.

The focus of this thesis will be put on the two first process steps, where the main feedstock required for the further processing, i.e. $CO_2$ and renewable hydrogen ($H_2$), are provided. These are of special interest, as they provide the basis not only for the C2PAT demonstration project, but also for other carbon utilisation pathways, that can alternatively be pursued in the cement industry.

The technology routes for carbon capture and hydrogen production as envisioned for the C2PAT will be discussed and assessed in regard to technological and economic fit, especially focussing on efficiencies and cost assumptions of each technology compared to alternative carbon capture and electrolysis technologies. Challenges and implications for the application of further carbon capture and utilization projects for the decarbonisation of the cement industry in Austria will be deducted, based on the results. Hence, the main research question of the proposed thesis is as follows:

> *“What are the technological and economic implications from existing research projects on carbon capture and renewable hydrogen production technologies, for the application in carbon capture and utilization pathways in the cement production, as envisioned in the C2PAT demonstration project?”*

Subsequently the following secondary research question shall be discussed based on the results of the main research question:

> *"What are the implications of the results for a potential decarbonisation of the cement industry in Austria via the suggested technology route in terms of energy demand?"*

As the focus of this thesis is on carbon capture and hydrogen production, further research and discussion of the subsequent process steps involved in the C2PAT demonstration project, that are only outlined later in this work, is suggested.

## 1.3. Methodical approach

Please note that the formulas introduced in this description of the methodical approach will be derived and explained in detail including the respective definitions and variables in the following chapters as indicated below:

In a first step, the cement production will be investigated to generate a common understanding and to assess the $CO_2$ emissions per ton of cement produced in the production process, differentiated into $CO_2$ from the combustion process (rotary kiln) and the $CO_2$ from the chemical transformation from $CaCO_3$ to CaO. To achieve this, the most important formulas for the assessment of total $CO_2$ emissions per ton of clinker will be derived and explained in chapter 3.2:

$$CaCO_3 + heat \rightarrow CaO + CO_2 \tag{1}$$

$$EF_{clinker} = 0.646 \times 0.785 = 0.507 \tag{2}$$

Based on these findings, the results will be compared with the total cement production in Austria and the total $CO_2$ emissions of the Austrian cement industry will be projected.

In a second step, the planned technology route of the C2PAT project will be assessed. This involves an examination of the carbon capture technology applied, as well as alternative technologies available. For the technological assessment the CO2 capture ratio (CCR) will be examined in chapter 4.1.3, as well as specific primary energy consumption for $CO_2$ avoided (SPECCA):

$$CCR = \frac{\dot{m}_{CO2,capt}}{\dot{m}_{CO2,gen}} \tag{3}$$

$$SPECCA = \frac{q_{clk,eq} - q_{clk,eq,ref}}{e_{clk,eq,ref} - e_{clk,eq}} \tag{4}$$

For the economic assessment, the concept of cost of clinker (COC) and the cost of avoided $CO_2$ (CAC) will be introduced in chapter 4.1.4, to show the economic impact of different technology selections, based on the results of a process simulation for a reference cement plant:

$$COC = C_{cap} + C_{fuel} + C_{RM} + C_{el} \tag{5}$$

$$CAC = \frac{COC + COC_{ref}}{e_{clk,eq,ref} - e_{clk,eq}} \tag{6}$$

Subsequently, the production of renewable hydrogen via different water electrolysis technologies will be examined, thus for the technological evaluation, the concept of process efficiency is introduced:

$$\eta_{system,LHV} = \frac{energy\ content\ of\ product\ flow}{total\ electrtical\ power\ input} \tag{7}$$

The final economical assessment will be evaluated based levelized cost of hydrogen' (LCOH), as derived from a comparative analysis for a similar project scope regarding hydrogen production (for a detailed explanation of the formula including all relevant variables, see 4.2.5):

$$LCOH = \frac{LHV}{\eta_{system,LHV}} \left( \left( \frac{i * (1 + \frac{i}{100})^n}{(1 + \frac{i}{100})^n - 1} + O\&M \right) \frac{CAPEX}{FLH} + P_{el} \right) \tag{8}$$

The following process steps will examined with the focus to obtain the most suitable ratio of $CO_2$ and $H_2$ needed for the further Fischer-Tropsch process, based on the fundamental chemical reaction for the production of synthesis gas (syngas) via reverse water gas shift (rWGS) reaction:

$$CO_2 + H_2 \rightarrow CO + H_2O \tag{9}$$

In a conclusion, the implications of the results of existing research projects will be summarized to conclude future cost reduction potentials and hydrogen and further electricity demand for the decarbonisation of the Austrian cement industry via the described technology route.

# 2. Background information

## 2.1. Climate targets

The International Panel on Climate Change (IPCC) states, that the anthropogenic greenhouse gas emissions have already led to an increase of 1.0°C of global warming compared to pre-industrial levels, with a range of +/- 20%. Thus global warming is likely to increase above 1.5°C between 2030 and 2052 if the current rate of increase is to continue (IPCC, 2019, p. 4).

The Paris Agreement addressed this issue globally in 2015 and is considered to be the first legally binding international treaty on climate change. The 196 international parties involved reached a common agreement to:

> *“Holding the increase in the global average temperature to well below 2°C above pre-industrial levels and pursuing efforts to limit the temperature increase to 1.5°C above pre-industrial levels, recognizing that this would significantly reduce the risks and impacts of climate change.” (United Nations, 2015, p. 4)*

As a consequence of this treaty, many countries adopted their climate change related targets in an effort to meet the stated common goal. The European Climate Law today states a reduction of greenhouse gases of 55% compared to the emission level of 1990 and strives for climate neutrality in the European Union by 2050 (EU, 2021, pp. 2-5). The level of the Austrian government is even more ambitious, as it is committed to carbon neutrality already by 2040 (BMK, 2020). Austria’s renewable energy strategy

In 2018, Austria released the climate and energy strategy called ‘#mission2030’. This document was the basis for the National Climate and Energy Plan (NECP), as transmitted to the EU in 2019. Besides the target to increase the share of renewable energy of the gross final energy consumption from approx. 33% in 2017 to a range from 46% and 50% in 2030, the NECP states the objective to supply 100% of the total electricity consumption from renewable energy sources by the year 2030, based on a net-balancing method of calculation, while taking the forecasted electricity consumption in 2030 into account (BMNT, 2019). This target was confirmed by the subsequent administration, and put into a legal framework called ‘Erneuerbaren Ausbau Gesetz’ (EAG), where a total expansion of 27 TWh was stipulated until the year 2030. The law also specifies, that 11 TWh ought to be from PV installations, 10 TWh from wind power, 5 TWh from hydro power and 1 TWh from bio mass plants (RIS, 2021). Based on the scenario with additional measure (WAM) the total electricity demand in Austria will increase from 274 PJ in 2020 to 307 PJ in 2030, the electricity demand of the industry sector is to grow from 100 PJ to 114 PJ (Umweltbundesamt, 2020).

## 2.2. Cement industry and the role of carbon capture

To achieve these ambitious climate targets, global greenhouse gas emissions must reach net-zero or even negative values. So far, there was a lot of focus on the decarbonisation of the electricity production, transport and buildings, but energy intensive industries, that contribute approximately one third of greenhouse gas emissions globally, have not been in the focus of decarbonisation measures so far. What is more, especially heavy industries, i.e. production of steel, aluminium and non-ferrous metals, cement, glass, chemicals and plastics, as well as pulp and paper etc., will be the hardest sectors to reach carbon neutrality. This is amongst other factors, because industrial plants, once built, have an average lifetime of around 20 years, display a high greenhouse gas intensity and many of the established production processes have already reached their practical efficiency limits. Furthermore, a growing global demand for the products from these industries will even lead to a further increase of production capacities (Ahman, et al., 2018, pp. 960-961).

For the cement industry, there are several potential pathways for greenhouse gas reduction. One is to reduce the share of clinker in the final cement, where a reduction of typically around 25% and higher seems possible, but is subject to local construction regulations. A substitution of fossil fuels for the operation of the kiln also contributes to a greenhouse gas reduction, but is reliant on the availability of non-fossil based fuels. However, as described later on, the clinker production process has immanent greenhouse gas emissions, as $CO_2$ is a by-product of the calcination of limestone to lime. Therefore, carbon capture storage and utilisation is an appropriate technological pathway for the cement production, as it prevents not just greenhouse gas emissions from the burning of fuels, but also $CO_2$ from the calcination process to enter the atmosphere (Ahman, et al., 2018, pp. 966-967).

According to the IEA, carbon capture, utilisation and storage (CCUS) comprises different technological solutions to capture $CO_2$ from the atmosphere or large point sources, for example flue gases from industrial production sites. Once the $CO_2$ is captured, it can be used as feedstock in various industrial process or be compressed and transported to geological sites, where it can be injected in underground formations, such as depleted oil and gas fields and kept for permanent storage (IEA, 2020, p. 19; Ghiat & Al-Ansari, 2021).

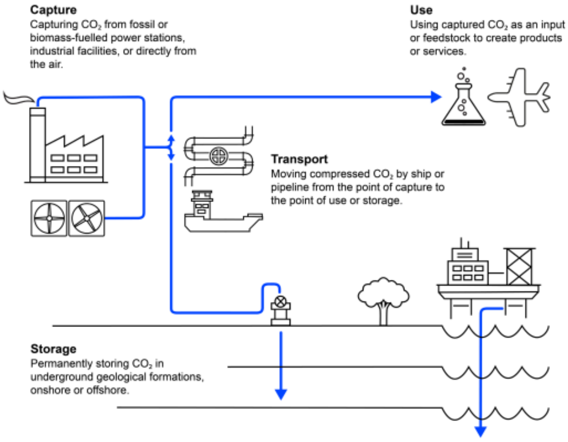

*Figure 1:Schematic illustration of CCUS (IEA, 2020, p. 20)*

When differentiating between the different routes illustrated above, it is referred to carbon capture and storage (CCS) when the $CO_2$ is permanently stored away. Carbon capture in utilisation (CCU) depicts applications, where the captured $CO_2$ is further processed together with additional feedstock, for example hydrogen, to chemicals and fuels etc. (IEA, 2020, p. 20). For the scope of this thesis, only CCU-technologies will be further investigated, as the application of CCS technologies is legally restricted in Austria since 2011 and was upheld in 2019. The law was established because of technological and ecological risks, that cannot yet be properly assessed (RIS, 2011; Parlament der Republik Österreich, 2019).

## 2.3. C2PAT demonstration project

The project 'Carbon-to-product-Austria' (C2PAT) is a joint initiative of Lafarge, OMV, Borealis and Verbund, with the common goal to reduce $CO_2$-emissions from the cement production focussing on Austria's biggest cement plant in Mannersdorf in Lower Austria, operated by Lafarge. The purpose of this undertaking is to develop a "cross-sectoral value chain for carbon capture" (Borealis, 2020) by transforming captured $CO_2$ from the cement plant in combination with green hydrogen, i.e. hydrogen produced from renewable electricity via electrolysis, to renewable-based hydrocarbons, which will be further processed renewable-based plastics, such as ethylene and propylene, before further processing to polyethylene or polypropylene at the polymerisation plant of Borealis. The schematic design of the C2PAT demonstration project is outlined in Figure 2.

The project is currently in the development phase, where the project consortium is discussing and evaluating the technical details, the business model and preparing the project for later commissioning. Based on these results, a demonstration project is scheduled to start operation in 2025 at the cement plant in Mannersdorf and the production sites of OMV and Borealis in Schwechat respectively, which are only 20 km away. The scope of the demonstration project is to capture 10,000 t/a of $CO_2$ from the flue gas of the cement plant, but the ultimate target is to scale up this process and implement a carbon capture and utilisation route for the total of 700,000 t of $CO_2$ emitted annually at the cement plant in Mannersdorf by the year 2030 (Holcim, 2020; OMV, 2021).

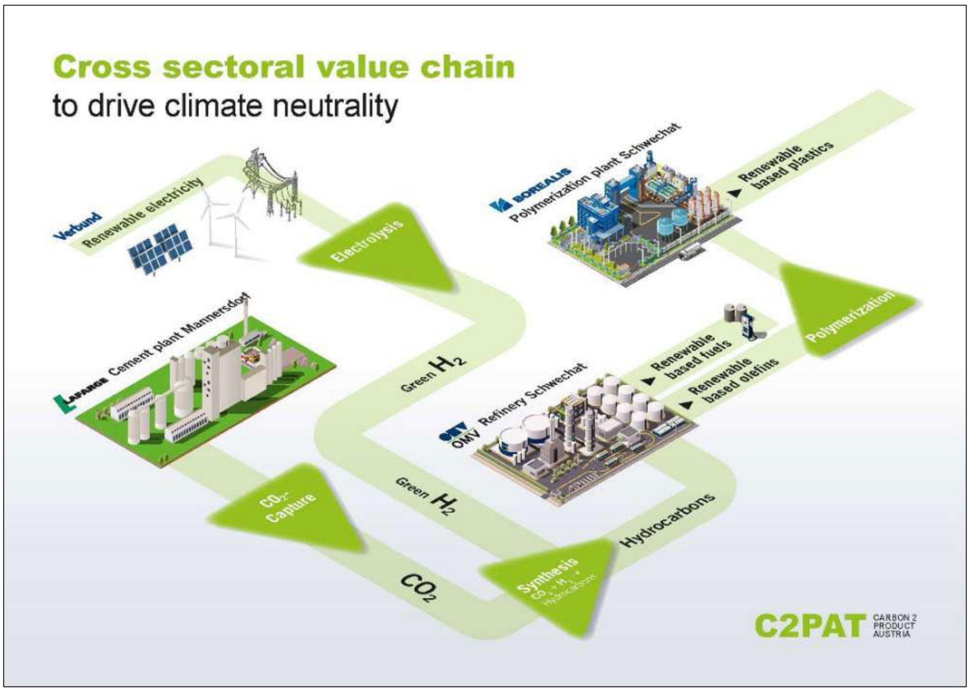

*Figure 2: Schematic outline of C2PAT value chain (Borealis, 2020)*

# 3. Cement production technology

## 3.1. Overview cement production

This section provides an overview of the cement production process to generate a general understanding of the production process and the inherent $CO_2$ emissions. Details of the production process as well as detailed chemical reactions will not be investigated in depth, unless relevant for the $CO_2$ assessment in the following sections. Cement manufacturing consists of three stages, comprising the preparation of raw materials, the clinker production and the grinding of clinker to produce cement (IEA, 2018, p. 12). These three stages will be described in the following sections.

### 3.1.1. Raw material preparation

Quarrying and crushing of raw materials

Raw materials for the cement production process are usually extracted from a quarry, where limestone as the primary raw material, as well as other materials and rocks in smaller quantities (e.g. clay, bauxite, iron ore, sandstone) are gained through opencast mining (Chatterjee, 2018, p. 1). The extraction usually takes place in the vicinity of the cement plant (IEA, 2018, p. 13). After the exploration, the quarried raw materials are getting crushed from 1-2 metres in size to usually less than 25 mm, as required as feed for the grinding mills, before being transported to the actual cement production plant. There are four general types of crushers that can be applied: jaw crushers and gyratory crushers that both operate based on a compression mechanism, hammer crushers operating on dynamic impact and double roll crushers that combine a shear, impact and compression mechanism. Energy intensity of the crushing process is relatively low, hammer crushers for example consume 0.5 to 1.0 kWh per metric ton of material. Abrasion of the metallic components of the crushers, with 1 to 10 grams per ton, can be rather high and needs to be considered. To avoid overgrinding, multiple stage crushing operations can be applied consisting of open and closed circuit crushing operations to ensure proper feed size (Chatterjee, 2018, pp. 54-55).

Pre-homogenisation of raw materials

To ensure that crushed limestone from different sector of the quarry has a homogeneous composition, a process called pre-homogenisation is applied, using pre-blending beds or stacking and reclaiming systems. This is usually performed by a mobile stacker conveyor, spreading the crushed limestone according to a certain pattern, the two most widely used stacking methods are Chevron and Window stacking. In both methods, the crushed limestone is reclaimed by belt conveyors at the end of the stack, using a rake to excavate the raw material across the whole lateral axis of the pile to ensure sufficient blending (Chatterjee, 2018, pp. 56-57), see the following figure for a schematic layout:

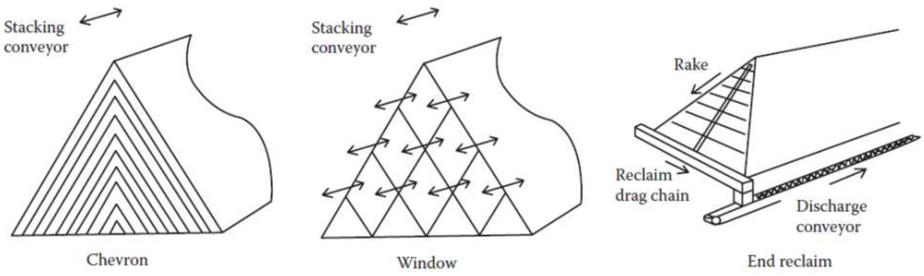

Figure 3: Schematic layout of pre-blending systems (Chatterjee, 2018, p. 57)

Ideally, the composition of the material in the pile is as close as possible to the chemical composition required by the further cement production process and needs to be constantly monitored and controlled (Chatterjee, 2018, p. 58).

Raw mill operation

The next process step is the raw milling, where the raw materials are grinded with four different types of raw milling system: ball mills, vertical roller mills and hydraulic roll presses as well as combinations. New installations often feature vertical roller mills, either for upgrading already existing ball mills or to reduce power consumption. All milling systems in this process step operate in closed circuit systems, and therefore operate also as classifiers (Chatterjee, 2018, pp. 58-60). The most relevant parameters for the raw milling feed are size, grinding behaviour of the feed material, drying capacity of the system, the required product fineness and the availability of power. All of the systems introduced have specific requirements and operating ranges that must be considered (Sorrentino, 2011, p. 618).

Homogenization process

The last process step in the preparation of the raw materials is the homogenization. This ensures the ideal composition of powdered raw materials which is now referred to as raw meal, before being fed into the kiln. Again, there are different methods to achieve the homogenization of the raw meal: mechanical, pneumatic and gravity systems, as well as slurry mixing, which is only used for wet process plants. Mechanical systems rely on multiple storage silos and the blending of the raw materials is achieved by mixing materials from various silos. Power consumption of these systems is rather low, but a high degree of material handling as well as space for the silos is required. Pneumatic homogenization systems however, have a higher power consumption (1.0 to 1.5 kWh/t of material) as they require compressed air, but are more efficient. With the help of compressed air introduced in the system, the raw meal behaves almost like a liquid and thus allows proper homogenization. Gravity blending on the other hand is again more energy efficient, as only 0.2 kWh/t of material or even less are needed. In these systems the raw meal is kept inside the silo in a constant sinking movement but with controlled sinking velocities over the cross-section of the silo. The raw meal is extracted from the silo through multiple outlets and thus mixed in a mixing vessel, that already is part of the kiln feeder (Chatterjee, 2018, pp. 60-63). The final raw meal mix has to be monitored regarding product fineness, chemical and mineral characteristics, existence of (un-)wanted minor constituents, homogeneity of the raw meal as well as reactivity and burnability, and additional materials have to be assessed if necessary to achieve the requirement parameters (Chatterjee, 2018, p. 70).

### 3.1.2. Clinker Production

The clinker production process is considered to be the “heart of the cement making process” (Chatterjee, 2018, p. 141) as the main chemical reactions as well as the largest share of the production costs accumulate in this process stage. The clinker formation process is a sequence of chemical reactions, occurring in the preheater-calciner, in the rotary kiln and the subsequent cooling of the clinker. The proper parameters of the raw meal ensure the reactivity and burnability during these process steps (IEA, 2018, p. 13; Chatterjee, 2018, pp. 141-143).

#### Preheater-calcination

The decarbonisation of limestone is conducted in the preheater-calcination stage. Limestone calcination is the process step of dissociating $CO_2$ from the limestone under the application of pressure and heat. This process step will be examined regarding the $CO_2$ emissions in further detail in the following sections. The basic principle of a preheater system is to pre-heat the cold raw meal, that is fed into the system at the top, with a counter flow of hot kiln gases. It then passes through a series of four to six vertical cyclones, that are placed above each other. To enhance the calcination process in the preheater, additional fuel can be fired in the riser duct of the preheater system, thus called preheater-calciner system. At the end of the preheater and the entry into the rotary kiln the raw meal reaches a temperature of over 900°C. During this processing, the material is evaporated and with increasing temperature, oxides of silicon, aluminium and iron are formed and the material loses one third of its original weight as it releases carbon molecules in form of $CO_2$. From an energy perspective, approximately 40% of the fuel required in the whole process is burned in this stage (Gao, et al., 2016, p. 555; IEA, 2018, p. 13). A typical preheater-calciner system, utilizing a Mitsubishi fluidized-bed calciner (MFC) is shown in the following figure:

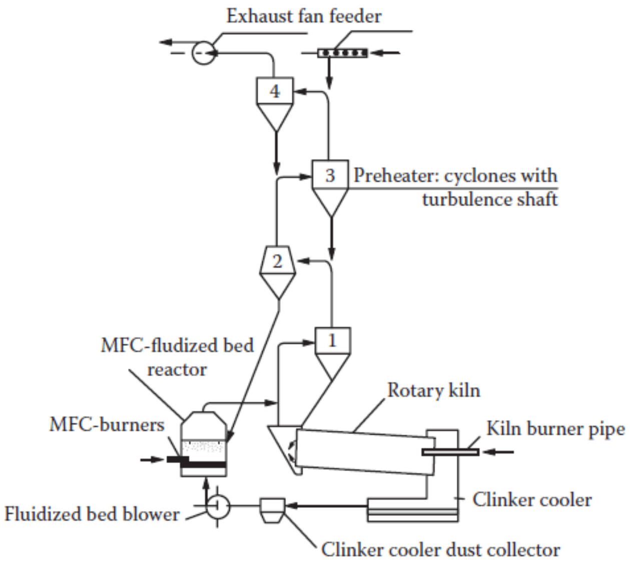

Figure 4: Schematic diagram of a preheater kiln with a MFC (Chatterjee, 2018, p. 148)

The degree of the calcination in the preheater-calciner depends on several variables, as the inside temperature, the residence time of the raw mix, the kinetic characteristics of the limestone etc. Modern preheater-calciner kiln systems are designed to achieve a calcination rate of above 85% (Chatterjee, 2018, pp. 146-149).

### Rotary kiln

After the preheating the precalcinated meal is fed into the rotary kiln, which is installed in a decreasing angle of 3 to 4 degrees. As the kiln rotates with about three to five rounds per minute the feed proceeds through the kiln. As the burner is situated at the end of the kiln, the material moves towards increasingly hotter zones, reaching temperatures of up to 1,450°C. The further 60% of the fuel demand is consumed by the rotary kiln burner, in order to maintain this very high temperature level to facilitate the desired chemical reactions, especially between calcium oxide and other elements in the material, as silica, aluminium oxide and ferric oxide to form a nodular compound. At the end of the rotary kiln, the prior raw meal is thus completely melted and fused together to form clinker and ready to enter the cooling process (IEA, 2018, p. 13; Gao, et al., 2016, pp. 555-556).

Clinker cooling

During the cooling phase, the hot clinker, leaving the rotary kiln still with a temperature of above 1,000°C, is rapidly cooled down to around 100°C. A variety of cooler systems is available but grate coolers are the preferred system in modern cement plants. The two main functions of this process step are, besides lowering the temperature of the clinker for further processing, the recovery of a substantial part of waste heat to recover its thermal energy. The hot air from the recuperation is used as secondary air for the combustion process and to fuel the pre-calciner as described before. The cooled clinker is then transferred to a storage, before the final blending and grinding occurs. (IEA, 2018, p. 13; Gao, et al., 2016, p. 556; Chatterjee, 2018, p. 157).

### 3.1.3. Clinker Grinding

In the final production steps, clinker is processed to the final cement. This is achieved by grinding the clinker and blending it together with gypsum to obtain Portland cement and other mineral components, for example as slag, power plant fly ash, limestone to produce blended cement. In general, all cement types contain 4 to 5% gypsum, in order to determine the setting time of the cement. The final composition of the cement is dependent on cement standards, market demands regarding specific types and characteristic, and last but not least, on the availability of additives for blending. In the last step, the mixture is ground to a fine grey powder, commonly using ball mills, in open-circuit or in combination with separators in closed-circuit systems. Lately more hybrid systems, with separators and hydraulic presses as well as vertical roller mills are also applied for the clinger grinding to enhance energy efficiency. The final product is stored in a cement silo before being dispatched to a packing station or a silo truck for commercialisation (Chatterjee, 2018, p. 175; IEA, 2018, p. 13; Gao, et al., 2016, pp. 556-557).

## 3.2. $CO_2$-assessement of cement production

In this section, the $CO_2$ emissions of the cement production process will be analysed for the further discussion. Generally, there are three categories of $CO_2$ emissions from the cement production process. The first is the decomposition of raw materials, i.e. the calcination process as described in the previous section, which is the largest share of total $CO_2$ emissions from cement production. Second is the combustion of fossil fuels, which depends on the specific type of kiln and preheater systems used. The third category is the indirect emissions from the power demand, in particular for the grinding process and the extraction and transport of the raw material (Zhou, et al., 2016, p. 465).The following figure provides an overview of $CO_2$ emissions along the cement production process:

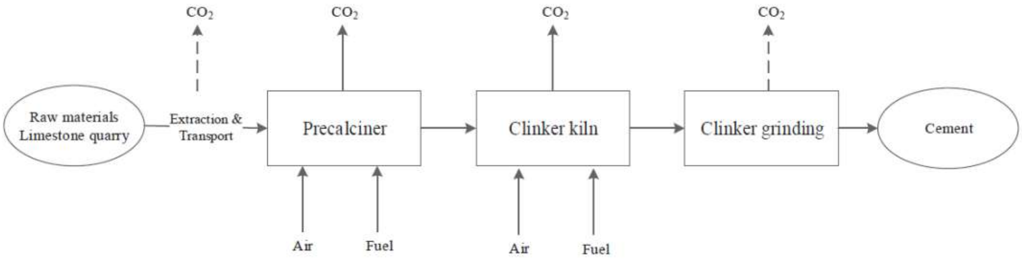

Figure 5: Overview of $CO_2$ emissions from cement production (Zhou, et al., 2016, p. 466)

As a general rule, approximately 2/3 of total $CO_2$ emissions originate from the conversion of limestone ($CaCO_3$) to lime (CaO) (Gardarsdottir, et al., 2019, p. 542). As emissions from the combustion process of fuel is highly dependent on the specific type of fuel and thus $CO_2$ emissions could be reduced by switching from fossil based fuels to bio based fuels, the focus of the following assessment will be put on the $CO_2$ emissions originating from the calcination process. The simplified stoichiometric relationship is provided in the following formula:

$$CaCO_3 + heat \rightarrow CaO + CO_2 \qquad \text{(see 1)}$$

Derived from this basic formula an emission factor (EF) for clinker production can be calculated as follows, based on the respective share of CaO in the raw material:

$$EF_{clinker} = fraction\ CaO\ \times (44.01\ \frac{g}{mole} CO_2 \div 56.08 \frac{g}{mole}\ \ CaO) \quad (10)$$

The resulting multiplication factor represents the molecular weight ratio of $CO_2$ to CaO in the raw material ($CaCO_3$), which result in 78.5%, based on the previous formula. Variations chemical compositions in different regions are not considered in this formula. Furthermore, the IPCC Guidelines provide a default value for the fraction of lime (64.6%) in the clinker, thus providing a formula for the emission factor as presented below:

$$EF_{clinker} = 0.646 \times 0.785 = 0.507 \quad \text{(see 2)}$$

For the further assessment, $CO_2$ emissions during the clinker production process will be determined based on the formula above (Gibbs, et al., 2001, pp. 175-177; IPCC, 1997).

## 3.3. $CO_2$ Emissions of the cement industry in Austria

The following section will provide an overview of the Austrian cement industry in therms of annual cement and clinker production as well as the corresponding annual $CO_2$ emissions.

### 3.3.1. Cement industry in Austria

The total Austrian cement production industry consists of 9 cement plants with a total installed clinker production capacity of 4,413,400 t/a. All of the production sites are based on a rotary kiln system, with only the plant in Peggau not featuring a 4 or 5 stage cyclone preheater system. Peggau and Vils are also the only 2 plants operate without a calciner system prior to the rotary kiln. The operators and locations of the sites can be seen in the following table, a more detailed overview is provided in Appendix 1:

*Table 1: Cement plant operators and site locations in Austria (Mauschitz, 2021, p. 4)*

| **Operator** | **Location (State)** |
|---|---|
| Zementwerk Leube GmbH | Gartenau (Salzburg) |
| Zementwerk Hatschek GmbH | Gmunden (Upper Austria) |
| Kirchdorfer Zementwerk Hofmann Ges.m.b.H. | Kirchdorf a.d. Krems (Upper Austria) |
| Lafarge Zementwerke GmbH | Mannersdorf (Lower Austria) |

| | |
|---|---|
| w&p Zement GmbH | Peggau (Styria) |
| Lafarge Zementwerke GmbH | Retznei (Styria) |
| Schretter & Cie GmbH & Co KG | Vils (Tyrol) |
| w&p Zement GmbH | Wietersdorf (Carinthia) |
| Baumit GmbH | Wopfing (Lower Austria) |

Cement production in 2020 totalled 5,4045367 t, which is a slight increase of 2.7 % compared to 2019. In the same period, the total clinker production also increased from 3.422.866 t in 2019 to 3,522, 299 t in 2020 (+2,9%). Based on the installed clinker production capacity, the capacity utilisation was 65% in 2020, respectively 64% in 2019. The clinker to cement ration was constant at around 70% during the last 5 years, the specific annual production numbers over this time period are shown in the following figure:

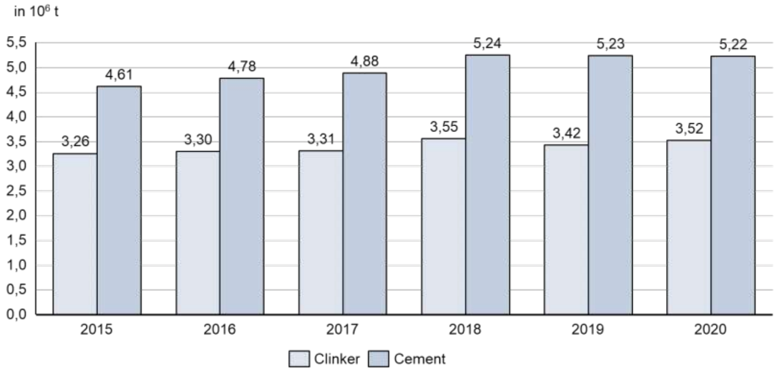

Figure 6: Clinker and cement production in Austria (own illustration based on Mauschitz, 2021, p. 7)

### 3.3.1. Energy consumption of cement industry in Austria

The total energy consumption for the total cement produced in 2020 was 15,863 TJ, which was an increase of 0.6% compared to 2019. A constant share of about 86% of the energy is consumed as thermal energy, while the remainder is electric energy consumption. (Mauschitz, 2021, p. 10)

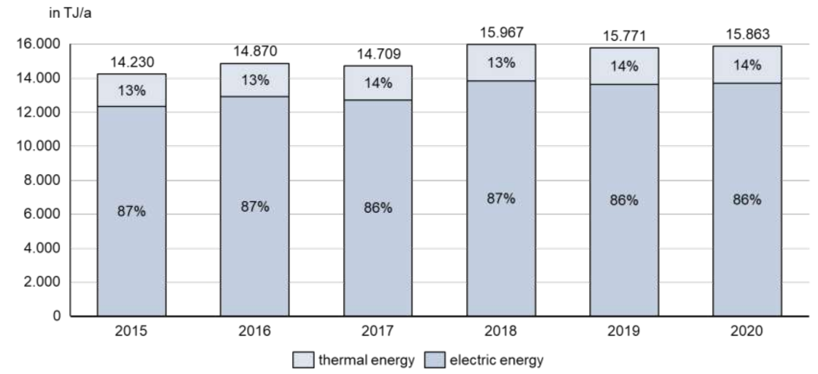

Figure 7: Total energy consumption of cement production in Austria (own illustration based on Mauschitz, 2021, p. 10)

For the total amount of clinker produced, the specific energy consumption was 3,891 MJ per tonne of clinker produced. This is a reduction of 2.3% compared to 2019, but an increase of 2.7% compared to 2015. (Mauschitz, 2021, p. 15).

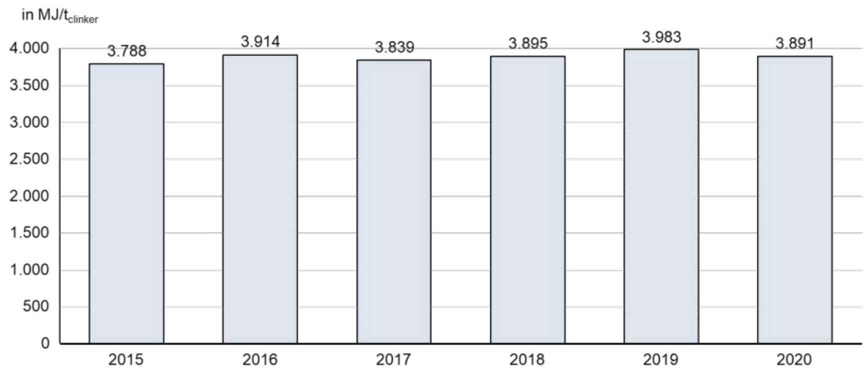

Figure 8: Specific consumption of thermal energy of cement production in Austria (own illustration based on Mauschitz, 2021, p. 15)

When looking at the fuel consumption, a differentiation has to be made between the consumption of conventional fuels and alternative fuels of biogenic or not-biogenic origin. In 2020 a total of 144,854 t of conventional fuels have been consumed by the Austrian cement industry, which is an increase of 30% compared to 2019, and even an increase of 35% compared to 2015. Coal contributed the major share 2020 with about 45%, followed by pulverized lignite with 34% and natural gas with 14% (Mauschitz, 2021, p. 9).

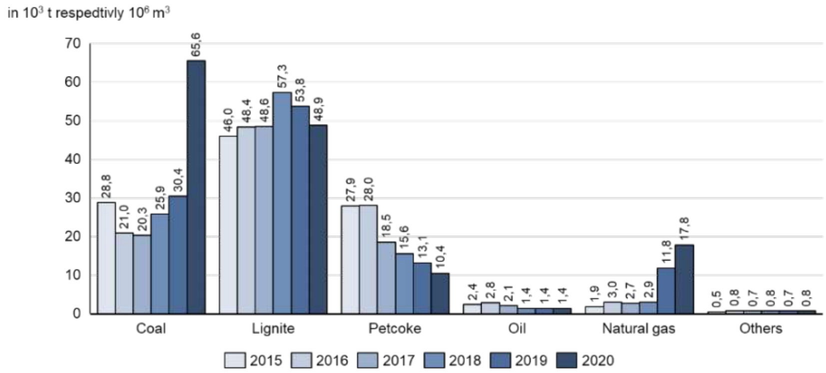

*Figure 9: Conventional fuel consumption of cement production in Austria (own illustration based on Mauschitz, 2021, p. 9)*

The consumption of alternative fuels was 491 t in 2020, which is a decrease of 8% compared to 2019 and the same value as in 2015. The majority of alternative fuel was plastic revenues, amounting to 61%, respectively 299,484 t in 2020.

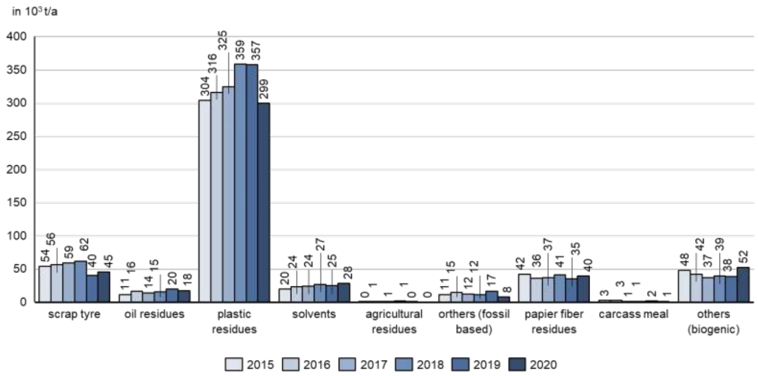

Figure 10: Alternative fuel consumption of cement production in Austria (own illustration based on Mauschitz, 2021, p. 10)

When comparing the energy content of conventional and alternative fuels, it is evident, that alternative fuels supply the majority of the thermal energy demand, with about 71% in 2020 and even higher values up to 81% in the previous years.

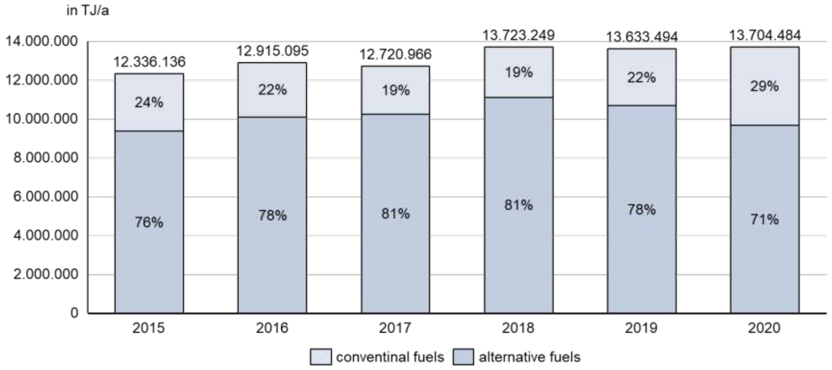

Figure 11: Share of fuels for thermal energy consumption of cement production in Austria (own illustration based on Mauschitz, 2021, p. 11)

### 3.3.1. $CO_2$ emissions of cement industry in Austria

When looking at $CO_2$ emissions of cement industry in Austria, a total of 2,947,166 tons was emitted in 2020, which is an increase of 1.8% compared to 2019 and even an increase of 6% compared to 2015. As assessed in the previous section, the majority of $CO_2$ emissions, about 62%, is generated because of the calcination process form limestone ($CaCO_3$) to lime ($CaO$). This share remains constant over the reference period of 5 years. The share of emissions originated form combustion of conventional fuels fluctuates between 8% and 12%, alternative fuels on the other hand combine to 27% in 2020, with a range up to 31%, as reported in 2018 (Mauschitz, 2021, p. 21).

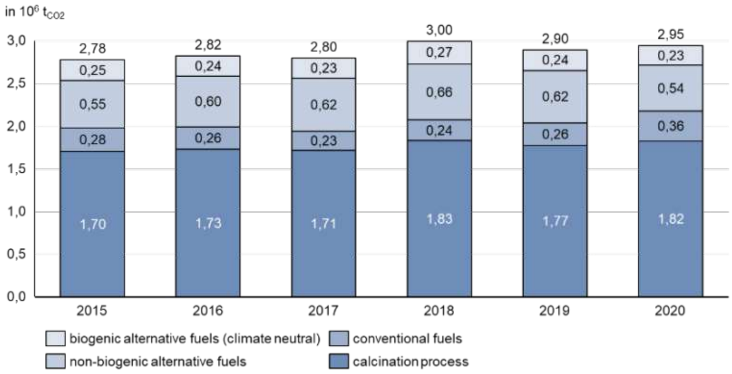

Figure 12: $CO_2$ Emissions from cement production in Austria (own illustration based on Mauschitz, 2021, p. 21)

When finally looking at the specific $CO_2$ emissions per ton of clinker produced, the clinker emission factor of 0.507 calculated by the IPCC as discussed in the previous section, provides a fairly accurate approximation of the $CO_2$ emissions originating in the calcination process. The specific $CO_2$ emissions of the Austrian cement industry per ton of clinker amount to 0,52 ton of $CO_2$ per ton of clinker in 2020, and also with minor deviations in the previous 5 years (Mauschitz, 2021, p. 22).

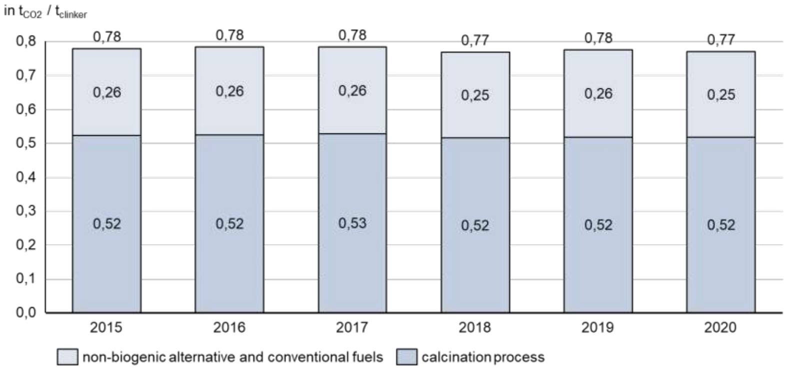

Figure 13: Specific $CO_2$ emissions (without biogenic $CO_2$ emissions) from cement production in Austria (own illustration based on Mauschitz, 2021, p. 22)

# 4. Carbon Capture and hydrogen production technology route of demonstration project

## 4.1. Carbon Capture technology

The fundamental approach of carbon capture is to maintain established production processes but complement them with a $CO_2$ sequestration unit. The available technological pathways can be divided into oxy-fuel combustion, pre-combustion and post-combustion capture processes (Ghiat & Al-Ansari, 2021). In oxy-fuel processes, nitrogen is separated from the ambient air, so the kiln is fed with pure oxygen. The advantage of oxy-fuel combustion is, that the $CO_2$ concentration in the flue gases is higher and therefore easier to capture. Oxy-fuel based carbon capture processes are primarily suitable for new cement plants, as for existing cement plants, the necessary modifications are highly challenging, as an air separation unit needs to be installed, the total plant needs to be sealed to avoid permeation of ambient air, equipment for flue gas recirculation has to be added and all effects of the different flue gas characteristics have to be considered (Schneider, et al., 2011, p. 646). In pre-combustion carbon capture technologies, the fuel for the combustion process, e.g. coal is gasified and reformed to CO and $H_2$ which can be further reformed to $CO_2$ and $H_2$ via a water-gas shift reactor. The $CO_2$ can be separated, while the $H_2$ can be used as a carbon-free feedstock (Ghiat & Al-Ansari, 2021; Mondal, et al., 2012, pp. 431-432).

As a large share of the $CO_2$ emissions of the cement production process results from the treatment of limestone, the imperative for carbon capture approaches is that not only the $CO_2$ emission of the combustion of the kiln fuel need to be considered, but also the $CO_2$ emissions from the calcination process. As this contributes also to a higher $CO_2$ concentration in flue gases, the application of post-combustion capture is even more suitable for cement plants, than it is for power plants (Bosoaga, et al., 2009, pp. 134-137). Adding to this, post-combustion capture technologies have the advantage that they do not require major changes in the existing plant design, therefore it is very suitable for the upgrading of existing cement production plants (Schneider, et al., 2011, p. 646).

Post-combustion carbon capture by amine scrubbing has been in use in the chemical industry and is thus a commercially mature technology and especially the process design of a cement plant allows an integration of amine based carbon capture technology without fundamental modifications (Bosoaga, et al., 2009, p. 137). For these reasons, technological maturity and ease of integration in the existing production site, amine scrubbing was selected for further feasibility studies in the C2PAT demonstration project.

The following section provides a general outline of amine scrubbing as the selected carbon capture technology. Subsequently, a comparative technological and economical assessment will be made with alternative carbon capture technologies, based on an comprehensive simulation as performed by Voldsund et al. (2019) and Gardarsdottir et al. (2019).

### 4.1.1. Amine scrubbing

The basic process of amine scrubbing was developed to absorb $CO_2$ from coal fired power plants and was patented in 1930 (Rochelle, 2009, p. 1652) This post-combustion capture technology is based on aqueous amines as solvents, as for example monoethanolamine (MEA), and is based on the fundamental difference of the chemical reactivity of $CO_2$ compared to others gases in the flue gas mix: $CO_2$ reacts first with water and thus forms carbonic acid, $H_2CO_3$ (formula (11). This carbonic acid reacts in further proton exchange reactions and primarily forms bicarbonate, $HCO_3^-$ (formula (12), which is non-volatile and can further be bound to the amine solution to from carbamic acid (formula (13) (Puxty & Maeder, 2016, pp. 13-21):

$$CO_2 + H_2O \rightarrow H_2CO_3 \qquad (11)$$

$$CO_2 + OH^- \rightarrow HCO_3^- \qquad (12)$$

$$CO_2 + RNH_2 \rightarrow RNHCO_2H \qquad (13)$$

While $SO_x$ and other acid gases can also react with water to acids, only $CO_2$ reacts with the amines in the solution. All others gases can escape during this chemical process. For the purpose of this paper, only these main chemical reactions will be introduced. For a more detailed assessment please refer to Puxty & Maeder (2016).

A big drawback of this process is that the energy required for the production of the chemicals emits more $CO_2$ than can be captured in one cycle. Therefore, the chemicals need to be used in a closed cycle, with a process step to release the $CO_2$

from the absorber before it can be used again in the first step (Puxty & Maeder, 2016, p. 13), as can be seen in the following illustration:

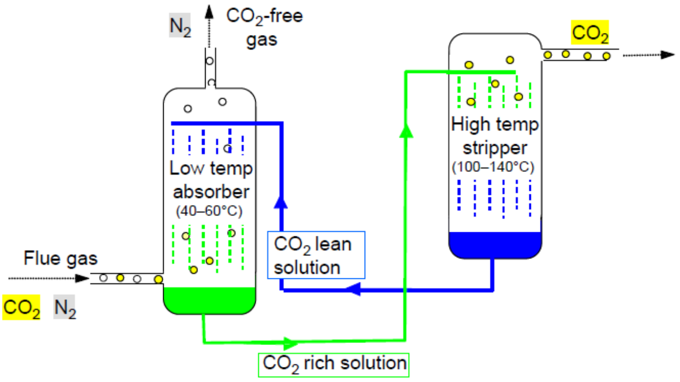

Figure 14: Schematic diagram of amine-based carbon capture system (Puxty & Maeder, 2016, p. 14)

This basic layout shows that the flue gas is fed into a so-called packed column. This is a hollow vessel, that is filled with a packing material, usually stainless steel rings. By arranging them densely, a large surface area is created for the flue gas, that entries the packed column at the bottom, and the amine solution, that is fed into to absorber from the top. When moving down, the amine solution absorbs more and more $CO_2$, until it reaches the bottom and will be transferred to the stripper. The stripper is again a packed column, where the $CO_2$ rich solution is fed into at the top. At the bottom, steam is produced, that flows upwards. The amine solution is thus heated up and released and flows upwards in order to leave the stripper column. The basic principle of this process is thus driven by the temperature swing, as the $CO_2$ is absorbed in an amine solution in the absorber column at a lower temperature to facilitate the reactivity, i.e. 40 to 60°C. The temperature of the solution is then heated up in the stripper to reduce the $CO_2$ solubility, thus the $CO_2$ is getting released again and the amine solution can be fed into the cycle again. Amines are selected for carbon capture because of this property to react with and release $CO_2$ depending on the temperature levels, even though also other chemicals can be used for these process step, for example potassium carbonate solutions (Puxty & Maeder, 2016, pp. 13-16).

### 4.1.2. Technological and economic assessment of alternative carbon capture technologies

The following assessments will be performed based on a process simulation of a reference cement plant based on a dry kiln process with a five-stage cyclone preheater-calciner system, and a rotary kiln with grate cooler system. Subsequently the process simulation is complemented by the various carbon capture technologies to be assessed. Further details can be obtained in Voldsund et al. (2019).

*Table 2: Main parameter of reference cement plant (Voldsund, et al., 2019, p. 3)*

| **Parameter** | **Value** |
|---|---|
| Clinker production | 1.0 Mt/a |
| Cement production | 1.4 Mt/a |
| Specific $CO_2$ emissions | 850 kg/$t_{clk}$ |
| Run time | >330 d/a |

## Oxy-fuel technology

In an oxy-fuel process, a pre-combustion carbon capture technology, pure oxygen, obtained from a separate air separation unit (ASU) and recycled $CO_2$ from the flue gas stream, are injected in the rotary kiln and the preheater. Thus, a very high content of $CO_2$ in the flue gas can be achieved, that is treated in a $CO_2$ purification unit, before further processing can be applied. A big disadvantage of the oxy-fuel technology next to the additional power demand mainly for the ASU is, that the kiln process must be modified. This changes the gas atmosphere in the whole system and must be considered accordingly, to control impacts on the clinker production process (Voldsund, et al., 2019, pp. 5-6).

## Chilled ammonia process (CAP)

CAP is a post-combustion carbon capture technology, where chilled ammonia is used as a solvent to remove the $CO_2$ form the flue gas. In a first process step, $SO_x$ is removed from the flue gas via ammonia scrubbing. In a second step, $CO_2$ is separated from the remaining flue gas stream in the absorber column at a temperature of around 12-13°C via an ammonia solvent. The ammonia then is recycled in a desorption column and fed back into the process. From the $CO_2$-desorber column, the $CO_2$ can be treated before further application in a CCS or CCU process (Voldsund, et al., 2019, pp. 6-7)

## Membrane-assisted $CO_2$ Liquefaction (MAL)

MAL is also a post-combustion carbon capture technology, where a in a first process step polymeric membranes are used for separation of large quantities of $CO_2$, but only at an unsatisfying degree of purity. Therefore, the $CO_2$ is further liquefied and thus gaining the needed purity of $CO_2$ for processing. The crux of the MAL process is the chemical stability of the polymeric membranes, which are still to be further developed for stable operation, that also provides high tolerance for impurities of the gas stream (Voldsund, et al., 2019, p. 7).

Calcium looping (CaL)

CaL is taking advantage of the reversible carbonation reaction (see formula (1), to separate $CO_2$ from the flue gas and can be applied in a tail-end or an integrated set-up. For both types of CaL-processes, an additional limestone supply and coal is needed. In the tail-end set-up, the $CO_2$ is separated from the remaining flue gas in a reaction with a CaO-based sorbent in the carbonator. The sorbent is regained in an additional coal-fired calciner under oxy-fuel conditions, where the oxygen is provided by a separate ASU. The waste heat can be used in a steam cycle to generate power, that can be used for prior clinker production process. Again, after a sufficiently high concentration of the $CO_2$ is achieved, it must be treated in a CPU, before the $CO_2$ can be further exploited. In contrast to the tail-end configuration, the integrated CaL process combines the calciner for the $CO_2$ capture process with the already existing calciner in the clinker production.

### 4.1.3. Technological efficiency assessment of carbon capture

As stated before, carbon capture based on chemical absorption with amines is the most mature technology, but there are also newer technologies, that have been the focus of research projects, but have not been put into practical operation in the same degree as amine scrubbing based on MEA. For an assessment of the technical efficiency of the above mentioned amine scrubbing as well as alternative technologies routes three different performance indicators are taken into consideration: the $CO_2$ capture ratio (CCR) and the equivalent $CO_2$ avoided ($AC_{eq}$) for assessment of the emission abatement, the specific primary energy consumption for $CO_2$ avoided (SPECCA) for the assessment of the energy performance, and finally a qualitative assessment of the retrofitability of the specific technology (Voldsund, et al., 2019).

The $CO_2$ capture ratio is defined as the amount of $CO_2$ captured from the flue gas ($\dot{m}_{CO2,capt}$), divided by the total $CO_2$ generated ($\dot{m}_{CO2,gen}$):

$$CCR = \frac{\dot{m}_{CO2,capt}}{\dot{m}_{CO2,gen}} \qquad \text{(see 3)}$$

As the CCR does not include the $CO_2$ generated by any additional equipment, for example natural gas boilers for additional steam supply. Thus, a further indicator needs to be considered: The equivalent $CO_2$ avoided is calculated based on the total equivalent $CO_2$ emissions avoided, including direct emissions from the flue gas as well as direct emissions from the generation of any additional steam etc. and the indirect emissions from electric power consumption. $e_{clk,eq,ref}$ is the specific equivalent emission in kg per ton of clinker produced from the reference plant without carbon capture, and $e_{clk,eq}$ the specific equivalent emission in kg per ton of clinker produced with carbon capture and including direct and indirect emissions as mentioned above (Voldsund, et al., 2019):

$$AC_{eq} = \frac{e_{clk,eq,ref} - e_{clk,eq}}{e_{clk,eq,ref}} \qquad (14)$$

The key indicator used for the assessment of energy performance, the specific primary energy consumption for $CO_2$ avoided (SPECCA) is defined as the delta between the equivalent primary energy consumption of the cement plant with ($q_{clk,eq}$) and without ($q_{clk,eq,ref}$) carbon capture, divided by the delta between the equivalent $CO_2$ emissions without and with capture in MJ per kg of $CO_2$ captured (Voldsund, et al., 2019):

$$SPECCA = \frac{q_{clk,eq} - q_{clk,eq,ref}}{e_{clk,eq,ref} - e_{clk,eq}} \qquad \text{(see 4)}$$

For the qualitative assessment of the retrofitability of different carbon capture technologies, the following factors have been taken into consideration: impact on the existing cement production process, additional equipment and necessary footprint, additional utilities and services, management of additional chemicals or subsystems and the availability of operational experiences with the specific system (Voldsund, et al., 2019).

The following results are based on a process simulation, where the authors examined the reference plant without carbon capture and with amine scrubbing via MEA as carbon capture reference technology. The other carbon capture technologies in the comparison are as described above an oxy-fuel process, a chilled ammonia process

(CAP), membrane-assisted $CO_2$ liquefaction (MAL) and a calcium-looping (CaL) process.

Assessment of emission abatement

When looking at the results for the MEA process, it can be seen, that a carbon capture rate of 90% can be achieved based on these simulations (Voldsund, et al., 2019). This can also be supported by the practical results from the demonstration project of Norcem in Brevik (Norway), where also a capture ratio of 90% could be achieved in operation (Knudsen, 2015, p. 9). For further comparison with other technologies, the carbon capture plants were assumed to be designed to all achieve a comparable carbon capture ratio (as defined in formula 3) of around 90%:

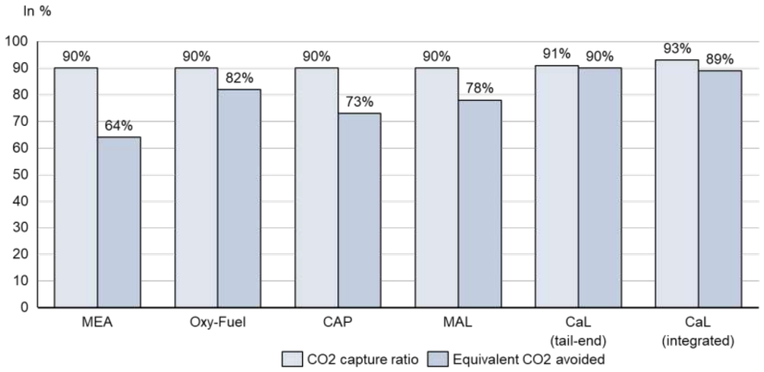

Figure 15: $CO_2$ capture ratio and $CO_2$ avoided (own illustration based on Voldsund, et al., 2019, p. 22)

When looking at the results for equivalent $CO_2$ avoided, it can be seen, that when taking direct emissions of steam generation with natural gas boilers and any additional electricity demand into consideration, ratio of equivalent $CO_2$ avoided for MEA drops from 90% to 64%, due to the high heat demand to release the $CO_2$ from the amine solution. All others technologies show a better performance in terms of emissions abatement, that has to be considered in further evaluations regarding the right technology choice for a specific project (Voldsund, et al., 2019). In absolute terms, the equivalent specific $CO_2$ avoided is shown in the following figure:

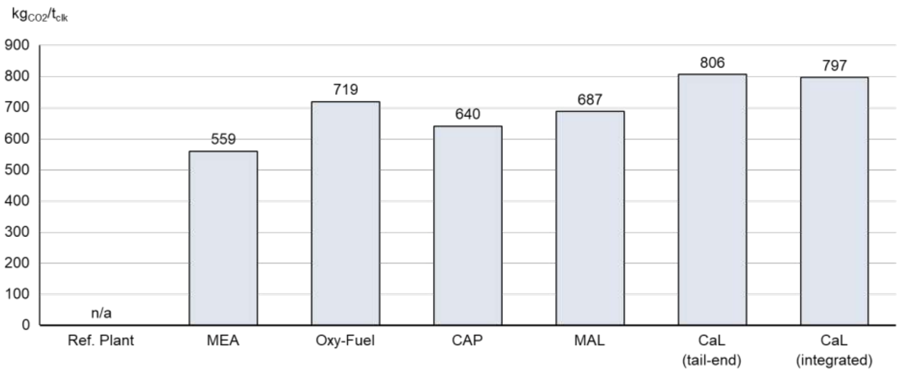

*Figure 16: Equivalent specific $CO_2$ avoided (own illustration based on Gardarsdottir, et al., 2019, p. 6)*

## Assessment of energy performance

When looking at the energy performance of the MEA process, it can be seen, that it has the highest SPECCA with 7.08 MJ per kg of $CO_2$ avoided. The biggest share of the additional energy demand is caused by the heat consumption (via stream from natural gas boilers), as well as additional electricity demand. All other technologies show a significantly lower specific primary energy consumption, ranging from 1.63 MJ/kg $CO_2$ (oxy-fuel) to 4.07 (tail-end calcium-looping) (Voldsund, et al., 2019).

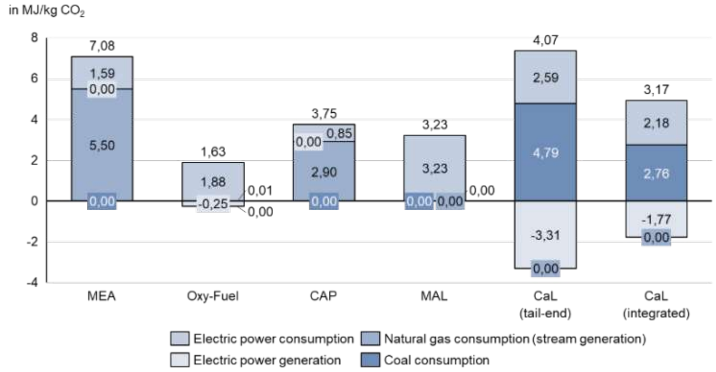

*Figure 17: Additional specific primary energy consumption for $CO_2$ avoided (own illustration based on Voldsund, et al., 2019, p. 23)*

## Assessment of retrofitability

For a comprehensive assessment, also the criteria regarding retrofitability, as mentioned above, need to be considered. The following table summarizes the results, with the colours-codes indicating the expected level of difficulty when retrofitting an existing cement plant. Green indicates a fairly straightforward retrofit, while the colours yellow (some attention necessary) and orange (special attention necessary) indicate an increasing degree of difficulty. The colour red states, that a retrofit with this technology would not be possible:

*Table 3: Assessment of retrofitability of carbon capture technologies (Voldsund, et al., 2019)*

| **Indicator** | **MEA** | **Oxyfuel** | **CAP** | **MAL** | **CaL Tail-End** | **CaL Int.** |
|---|---|---|---|---|---|---|
| Impact on the cement production process | 🟩 | 🟧 | 🟩 | 🟩 | 🟩 | 🟨 |
| Equipment and footprint | 🟨 | 🟧 | 🟨 | 🟨 | 🟨 | 🟧 |
| Utilities and services | 🟨 | 🟨 | 🟨 | 🟨 | 🟨 | 🟨 |
| Introduction of new chemicals/subsystems | 🟨 | 🟨 | 🟨 | 🟩 | 🟨 | 🟨 |
| Available operational experiences | 🟩 | 🟧 | 🟨 | 🟧 | 🟨 | 🟧 |

As a conclusion, it can be seen, that all post-combustion technologies are rated higher regarding the impact on the actual clinker production process, as they are generally less difficult to implement in an existing plant. For example, only a short shutdown of the production is necessary during the construction phase for the rerouting of the flue gas. While the indicators regarding equipment and footprint, utilities and services and introduction of new chemicals/subsystems show no unexpected discrepancy, the last indicator shows a strong advantage for MEA as the most mature carbon capture technology. Other technologies have been the centre of research and pilot projects, but have not been demonstrated along the whole process chain or in large scale operation respectively (Voldsund, et al., 2019) (Knudsen, 2015).

### 4.1.4. Economic assessment of $CO_2$ capture technologies

For the cost analysis, two important key performance indicators will be compared for the various $CO_2$ capture technologies discussed: cost of clinker (COC) in €/$t_{clk}$ and cost of $CO_2$ avoided (CCA) in €/$t_{CO2}$. Cost of clinker per ton is calculated as the sum of annualized CAPEX ($C_{cap}$), the cost of fuel ($C_{fuel}$), the cost of raw material ($C_{raw}$) and electricity cost ($C_{el}$):

$$COC = C_{cap} + C_{fuel} + C_{RM} + C_{el} \qquad \text{(see 5)}$$

The cost of avoided $CO_2$ is calculated based on the difference of COC between the reference plant without carbon capture ($COC_{ref}$)and the COC of the specific plant with one of the carbon capture technologies considered, divided by the difference of the specific emissions of the reference plant without $CO_2$ capture ($e_{clk,equ,ref}$)and the specific emissions of the plant with a $CO_2$ capture plant added ($e_{clk,qu}$):

$$CAC = \frac{COC + COC_{ref}}{e_{clk,eq,ref} - e_{clk,eq}} \qquad \text{(see 6)}$$

But before assessing these key parameters, total plant costs and total annual operational costs will be compared to gain a general understanding of the costs involved. The following figure summarizes the total plant costs of the reference cement plant and the total plant costs for the different cement plants with $CO_2$ capture plants. Total plant cost is defined as total direct costs plus total indirect cost. Total direct cost of the plant is calculated as the sum of equipment costs and installation costs of each component, multiplied with specific process contingency factors, depending on the maturity of the specific technology (Appendix 2). Total indirect cost is calculated as a factor of total direct costs, i.e. 14% of total direct cost for general indirect costs (such as yard improvement, costs for buildings and sundries, costs for service facilities, engineering and other consulting costs), 7% for owners costs and 15% for project contingency (Gardarsdottir, et al., 2019, p. 7). Based on all of these input parameters, MEA based $CO_2$ capture provides the lowest total plant costs, compared to the other capture technologies:

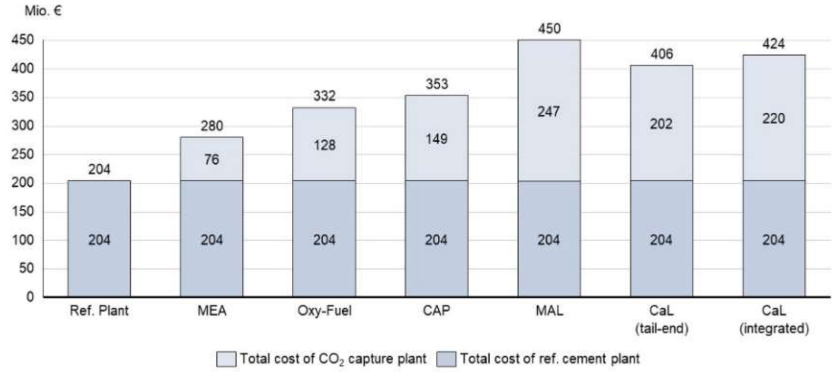

*Figure 18: Total cost of cement plant and $CO_2$ capture plant (own illustration based on Gardarsdottir, et al., 2019, p. 10)*

In the next figure compares annual operational expenditures (OPEX), which include fixed and variable operational expenses. Fixed OPEX comprise maintenance costs, including maintenance labour costs (2.5% of total pant costs), insurance costs and taxes (2% of total plant costs) and cost of operating personnel, i.e. 100 persons for the reference cement plant and additional 20 persons in the carbon capture plant (total annual cost of 60,000 € per person). Variable OPEX include fuel and raw material costs as well as utilities and other consumable material (see Appendix 3 for further detail). The results show a reversed picture, as MEA-based absorption is the most expansive technology on a yearly basis:

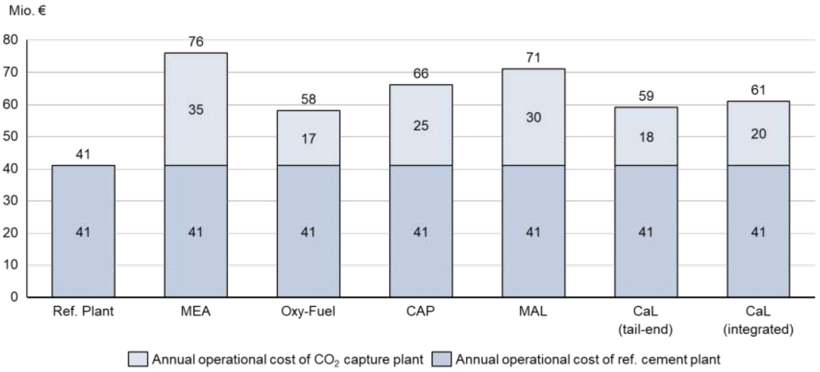

*Figure 19: Total annual operational cost of cement plant and $CO_2$ capture plant (own illustration based on Gardarsdottir, et al., 2019, p. 10)*

When now comparing the two key performance indicators as introduced above, the simulated costs for the reference plant total 62.5 €/$t_{clk}$ with considerable surcharges for the cost of clinker with installed $CO_2$ capture plants. While the oxy-fuel technology provides the lowest cost of clinker, it is notable that MEA-based absorption, chilled ammonia absorption and tail-end calcium looping have very similar cost of clinker produced:

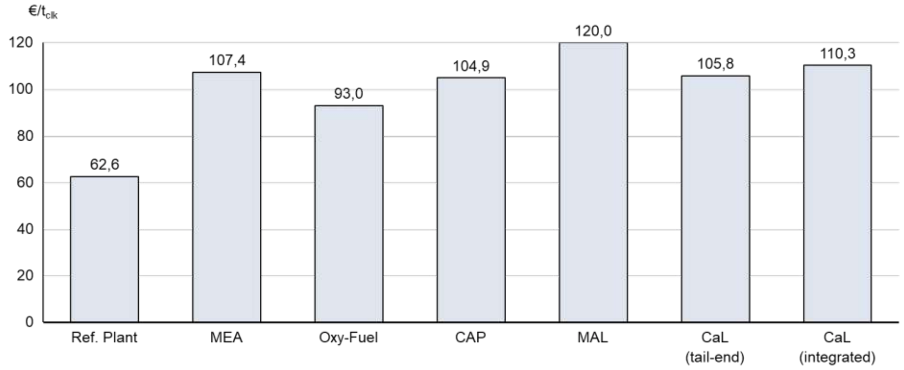

*Figure 20: Total cost of clinker of reference plant and with additional $CO_2$ capture plants (own illustration based on Gardarsdottir, et al., 2019, p. 10)*

However, as already shown in Figure 16, MEA-based absorption has the lowest equivalent specific $CO_2$ avoided, thus it is not surprising that the cost per ton of $CO_2$ avoided is almost the highest, second to MAL based $CO_2$ captureAppendix 4: Breakdown of COC for the reference cement plant and investigated $CO_2$ capture technologies. (Gardarsdottir, et al., 2019, p. 10):

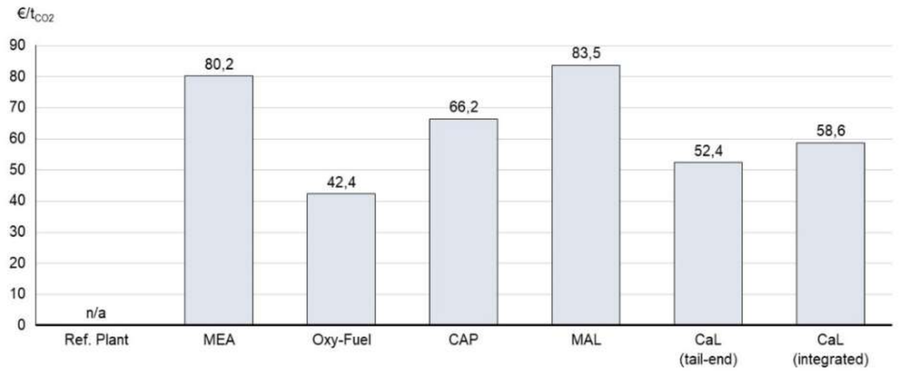

*Figure 21: Total cost of $CO_2$ avoided of reference plant and with additional $CO_2$ capture plants (own illustration based on Gardarsdottir, et al., 2019, p. 10)*

For a more detailed breakdown of the different cost parameters, see Appendix 4 and Appendix 5.

When comparing these results it has to be noted, that project contingencies are included in the simulation, i.e. the cost of less mature technologies are anticipated more conservatively because of uncertainties and lack of practical experiences, which is the case for oxy-fuel, MAL and integrated CaL. With further instalment of these technologies, the uncertainty in cost assumption can be further reduced, which would have an impact not only on CAPEX assumptions, but also on OPEX, as for example the fixed OPEX are also based on the total plant costs. When looking at the MEA-based absorption, the biggest contribution of CAC stems from the steam demand that is satisfied with a natural gas boiler. Depending on the properties of the actual cement production site, a higher share of waste heat could be available than assumed in the underlying simulations and thus reduce the operational expenditure for the stream production (Gardarsdottir, et al., 2019, pp. 10-13). A further improvement in the reduction of steam demand up to 33% can be achieved through the application of improved amines, compared to MEA, even though the results are hardly comparable to the simulations discussed above because of diverging assumptions and plant parameters (Jakobson, et al., 2017).

As the price for emitted $CO_2$ has not been discussed so far, Gardarsdottir et al. performed a sensitivity analysis of CAC to the price of a carbon tax. As can be seen in Figure 22, at a carbon taxation of approximately 75 €/t$_{CO2}$ the COC of the reference plant without carbon capture will be higher than the COC of the cement plant with MEA based capture technology. At an even lower carbon tax level of around 60€/t$_{CO2}$

other technologies reach cost parity, while oxy-fuel could become competitive already at around 40€/$t_{CO2}$ , based on these simulations (Gardarsdottir, et al., 2019, p. 14).

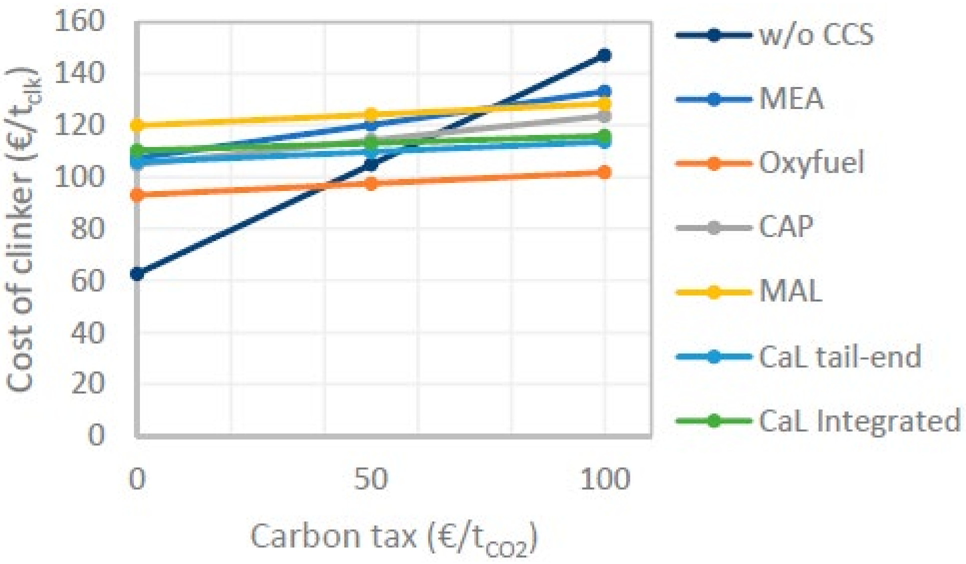

*Figure 22: Sensitivity of CAC to a carbon tax (Gardarsdottir, et al., 2019, p. 14)*

Other input parameters like fuel price, supply of waste heat etc. also influence the cost parity of carbon capture technologies, but must be evaluated carefully for the specific cement plant, where it should be supplied, also regarding the retrofitability aspects as discussed in the previous section. For the results of further sensitivity analysis from Gardarsdottir et al. (2019) on coal price (+/- 50%), cost of steam supply (+/- 50%), electricity price (+/- 50%) and total capex (+35/-15%) see appendix 6.

### 4.2. Hydrogen Production

The second major feedstock for the C2PAT project is hydrogen, as indicated by Figure 2. Today, the vast majority of the total global hydrogen demand of 70 mt/a is produced based on fossil fuels, especially hydrogen from the reforming of natural gas, which constitutes almost half (48%) of the global hydrogen supply, followed by oil reforming (30%) and coal gasification (18%):

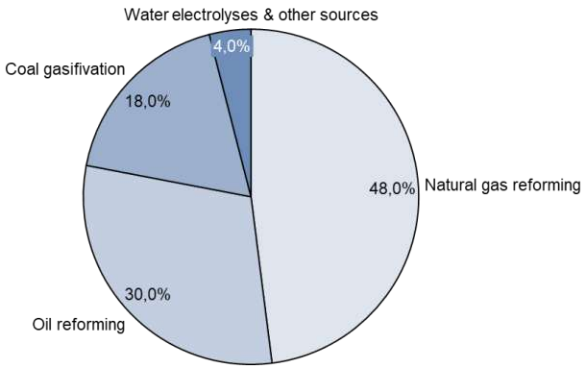

*Figure 23: Current global hydrogen production by source (own illustration based on Ji & Wang, 2021, p. 38615)*

The following figure provides an overview of different hydrogen production routes, based on the feedstock required and the primary energy source. For the purposes for the C2PAT as a CCU demonstrations project, fossil-based hydrogen production technologies are not suitable und thus will be not further examined.

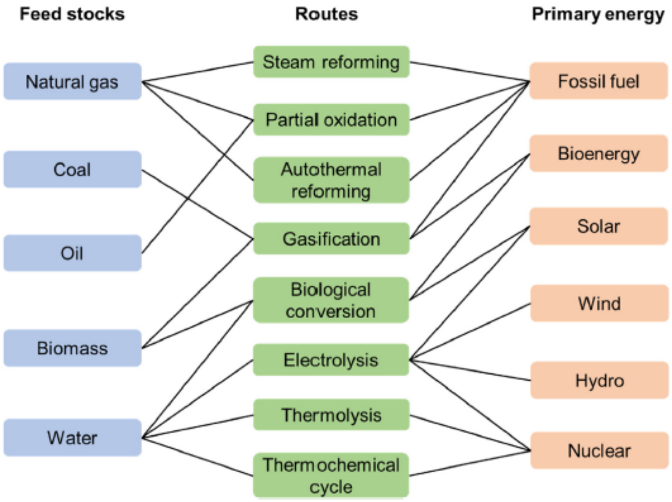

*Figure 24: Overview hydrogen production routes (Ji & Wang, 2021, p. 38615)*

Corresponding with the C2PAT project scope, further examination will focus on hydrogen production via electrolysis routes. Depending on the respective technology, these technologies provide a (pre-) commercial maturity and are suitable to provide hydrogen also in the quantities demanded for the envisioned scale of the demonstration project. The basic principle of water electrolysis to produce hydrogen is provided as follows (Ji & Wang, 2021, p. 38621):

$$H_2O \rightarrow H_2 + O_2 \qquad (15)$$

Hydrogen production via electrolysis need not be as integrated in the cement production process, as carbon capture plants, thus allowing a higher discretion for the technology section. As the final technology selection is to be decided yet, all three relevant technology options, i.e. alkaline electrolysis, polymer-exchange membrane electrolysis and high-temperature electrolysis will be introduced and assessed in the following section.

### 4.2.1. Alkaline electrolysis

Alkaline electrolysis (AEL) is the oldest and commercially most mature electrolysis technology, with a technology readiness level of TRL-9 (Varela, et al., 2021, p. 9304). The size of commercial AEL systems ranges up to 160 MW dimension, producing more than 33,000 Nm$^3$ of hydrogen per hour (Tremel, 2018, p. 22; Tenhumberg & Büker, 2020, p. 1588). At a temperature level of 60-90° C, AEL is also considered to be a low-temperature electrolysis. In an AEL, two electrodes are put in an alkaline solution, which is usually potassium hydroxide (KOH) or sodium hydroxide (NaOH). Both electrodes are separated by a porous diaphragm, separating the two gases, hydrogen and oxygen, but allowing negatively charged hydroxide ions ($OH^-$) to transfer from the negatively charged cathode to the positively charged anode. Electrodes for AEL system are typically made from non-noble nickel-based metals.

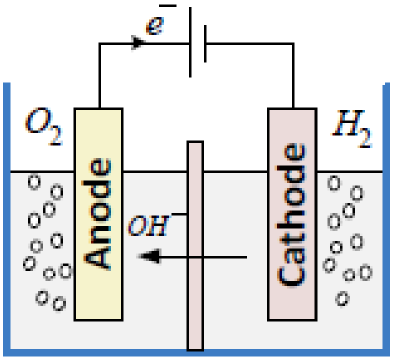

*Figure 25: Schematic overview AEL (El-Emam & Özcan, 2019, p. 597)*

The respective reactions taking place at the cathode and the anode are provided in the following formulas:

$$\text{Cathode:} \quad 4H_2O + e^- \rightarrow 2H_2 + 4OH^- \tag{16}$$

$$Anode: \quad 2H_2O \rightarrow O_2 + 4H^+ + 4e^- \tag{17}$$

The operational flexibility of AEL is limited as only a small pressure difference is allowed between the hydrogen and the oxygen side of the diaphragm, to prevent a diffusion of gases. This is especially critical under pressurized operation conditions, thus AEL are usually operated under atmospheric pressure (Tremel, 2018, p. 22).

### 4.2.2. PEM-electrolysis

Polymer-exchange membrane (PEM) electrolysis uses the same chemical principle shown in formula (16) and (21) for AEL, but uses a gas-tight polymer membrane as a solid electrolyte, where the charge carriers are not hydroxide-ions but positively charged hydrogen ions, i.e. protons (Ji & Wang, 2021, p. 38621; Tremel, 2018, p. 23).

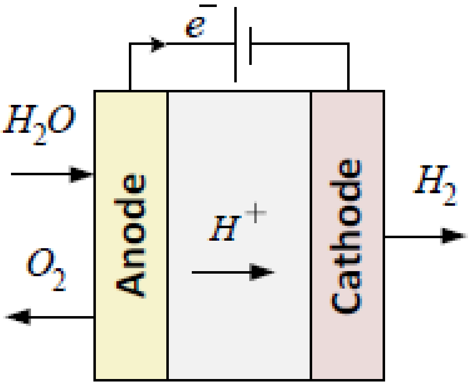

*Figure 26: Schematic overview PEM (El-Emam & Özcan, 2019, p. 597)*

PEM electrolysers are at a slightly lower TRL-8 (Varela, et al., 2021, p. 9304), with the largest systems installed currently at around 20 MW. PEM offer more compact design, a higher current density then AEL and also provide a higher operational flexibility, being able to ramp up from stand-by to full load operation within less than 10 seconds. Disadvantage of PEM electrolysers compared to AEL, despite the lower technological maturity are the demand for noble metals for the electrodes (such as iridium, ruthenium or platinum) and the degradation of the electrolyte, which leads to a shorter operational life cycle. Current research is thus focussing on improved polymer materials, catalysts and electrode materials to decrease costs and improve durability (Ji & Wang, 2021, pp. 38621-38622; Tremel, 2018, pp. 23-24).

### 4.2.3. High temperature electrolysis

The biggest difference of high temperature electrolysis, also called solid oxide electrolysis (SOEL) compared to AEL and PEM electrolysis is, that it is operated at a temperature between about 600 and 900° C and uses water steam as feedstock instead of liquid water (Tremel, 2018, p. 26). The advantage of high temperature electrolysis is, that part of the energy demand is consumed as heat, resulting in a significantly lower demand of electricity, while the total energy input only increases slightly. This is advantageous especially in environments where waste heat is available (El-Emam & Özcan, 2019, p. 598).

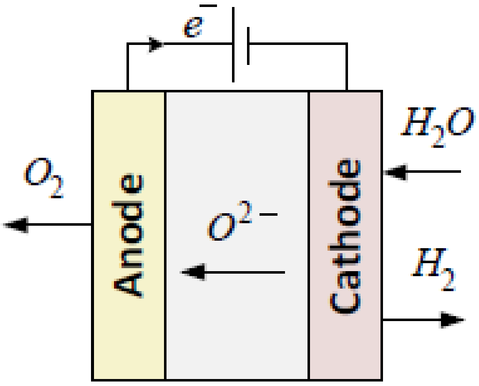

*Figure 27: Schematic overview SOEL (El-Emam & Özcan, 2019, p. 597)*

The electrolyte is made from ceramic, also solid oxide, i.e. usually zirconium dioxide ($ZrO_2$) with yttrium (III) oxide ($Y_2O_3$) for stabilization. The charge carrier in SOEC systems are oxide ions ($O^{2-}$), as can be seen in above Figure 26. This is also evident in the slightly different chemical reaction compared to low temperature electrolysis technologies (Ji & Wang, 2021, p. 38622):

$$\text{Cathode:}\quad 2H_2O + 4e^- \rightarrow 2H_2 + 2O^{2-} \qquad (18)$$

$$Anode{:}\quad 2O^{2-} \rightarrow O_2 + 4e^- \qquad (19)$$

A big disadvantage of SOEL is the currently still lower technology readiness level of TRL-5. SOEL is currently being explored in demonstration projects and thus not yet fully commercially applicable. A big challenge in the development of high-temperature electrolysers are the temperature and humidity levels, that cause degradation and general wear and tear on the equipment, also limiting the possibilities of flexible load changes during operation. However, because of the lower electricity demand, the expected future cost reduction potentials and the capability of reversing the process and using SOEL systems also in a fuel-cell mode, high temperature electrolysis is considered to be a very attractive alternative to the previously discussed low temperature electrolysis routes. (Varela, et al., 2021, p. 9304; Ji & Wang, 2021).

### 4.2.4. Process energy efficiency (electricity demand)

There are various definitions of energy efficiency, both in the scholar and industrial approaches. For the purpose of this evaluation a pragmatic approach used by the industry will be applied. On the input side, the total energy requirements of the whole system, including the balance of plant must be considered. In the output side, the energetic content of the hydrogen produced is specified in terms of the lower heating value (LHV):

$$\eta_{system,LHV} = \frac{energy\ content\ of\ product\ flow}{total\ electrtical\ power\ input} \qquad \text{(see 7)}$$

This definition is very specific to the relevant technology or application, as available heat sources and gas pressures required for the final products can vary significantly (Millet, 2015).

When comparing data from expert interviews conducted in a study by Smolinka et al. (2018) on behalf of the German Federal Ministry of Transport and Digital Infrastructure, the following data can be including forecasts to 2030 and 2050 respectively:

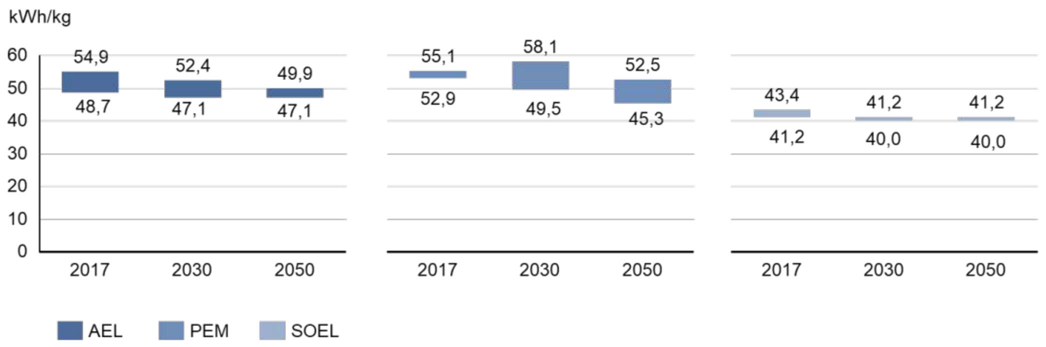

*Figure 28: Electric energy demand of electrolysis technologies (own illustration based on Smolinka, et al., 2018, p. 176)*

Currently AEL show a lower electric energy demand as PEM, even though the expected values for PEM should decrease to a lower level than AEL. The interim increase of energy demand is the result of the currently ongoing efforts to decrease investment costs, but as soon the respective targets are obtained, electricity consumption should catch up quickly. For SOEL it is important to note, that only electric energy demand is considered in the above figure (Smolinka, et al., 2018, p. 10).

Efficiency values, as defined in formula (24), for the different technologies are calculated based on the data given above and for the LHV of hydrogen, i.e. 33.33 kWh/kg (Linde Gas GmbH, 2013):

*Table 4: System efficiencies estimate for different electrolysis technologies (own calculation based on Smolinka, et al., 2018, p. 176 and Linde Gas GmbH, 2013)*

| **Electrolysis technology** | **2017** | **2030** | **2050** |
|---|---|---|---|
| AEL | 61-68% | 64-71% | 67-71% |
| PEM | 60-63% | 57-67% | 63-74% |
| SOEL | 77-81% | 81-83% | 81-83% |

An extensive market survey conducted by Buttler & Spliethoff (2018) came to a very similar result, but it has to be noted, that according to this market survey, the above estimates for current electrolysis systems are overstating the system efficiencies:

*Table 5: System efficiencies for current electrolysis systems (Buttler & Spliethoff, 2021, p. 2451)*

| **Electrolysis technology** | **Nominal system efficiency (LHV)** |
|---|---|
| AEL | 51-61% |
| PEM | 46-60% |
| SOEL | 76-81% |

### 4.2.5. Economic analysis of hydrogen production via electrolysis

To assess production costs of hydrogen, generally the concept of 'Levelized Cost Of Hydrogen' (LCOH) is applied, as provided by the following formula:

$$LCOH = \frac{LHV}{\eta_{system,LHV}} \left( \left( \frac{i * (1 + \frac{i}{100})^{n}}{(1 + \frac{i}{100})^{n} - 1} + O\&M \right) \frac{CAPEX}{FLH} + P_{el} \right) \qquad \text{(see 8)}$$

*Table 6: Input variables for LCOH calculations*

| **Variable** | **definition** |
|---|---|
| i | Interest rate |
| n | system lifetime |
| O&M | operation and maintenance cost (% of Capex) |
| CAPEX | Specific investment cost (€/kW) |
| FLH | full load hours (h) |
| $P_{el}$ | Cost of electricity (€/kWh) |

The biggest share of the cost of hydrogen productions via electrolysis is related to the energy input, i.e. electric power for AEL and PEM and electric power and heat, if a SOEL is applied. The cost of electricity can contribute up to 85% of the total production cost per kg of hydrogen, depending on the actual plant size, which determines the economies of scale, especially in respect to balance of plant (El-Emam & Özcan, 2019, p. 600).

There are many hydrogen production cost assessments available in literature, see for example Ji & Wang (2021), but there are very few evaluations, that allow a direct comparison of different electrolysis technologies previously introduced, especially in a layout that allows for inferences for the C2PAT project. However, an economic assessment was performed as part of the Carbon2Chem project, which is a CCU project, where $CO_2$ from steel mill gases is used together with renewable hydrogen to produce alcohols, plastics and fertilizers (Tenhumberg & Büker, 2020, p. 1586). The calculations were performed based on the data from the study by Smolinka et al. (2018), as introduced in the previous section. Even though this CCU project differs in several aspects from the C2PAT project, the results provide a relevant basis for an economic assessment.

For the economic analysis the cost of hydrogen production was calculated for a potential large scale electrolysis system with a production capacity of 130,000 $Nm_3$/h on the basis of the forecast value for 2030, as provided by Smolinka et al. (2018). As can be seen in formula .the main cost components are investment costs (CAPEX), the specific power consumption ($\eta_{system,LHV}$) as well as the load factor, i.e. FLH divided by the amount of total hours per year, and the cost of electric energy ($P_{el}$). The two latter components are independent from the electrolyser technology, but highly dependent on the available source of electricity, where a directly connected renewable energy source, e.g. PV parks or wind farms, can provide low electricity prices, but allows only a limited load factor. A grid connection, on the contrary, allows a high number of full load hours, but is subject to electricity market prices, which are generally higher. To reflect these uncertainties, hydrogen production costs are calculated as a function of the two variables, electricity price (in €/MWh) and load factor (in % of maximum full load hours):

*Table 7: Hydrogen production cost of AEL 160,000 Nm$^3$ system in 2030 in €/kg (own illustraton based on Tenhumberg & Büker, 2020, p. 1593)*

| | 25% | 40% | 50% | 60% | 75% | 90% |
|---|---|---|---|---|---|---|
| 25 €/MWh | 3,6 | 2,8 | 2,5 | 2,3 | 2,1 | 2,0 |
| 40 €/MWh | 4,4 | 3,5 | 3,2 | 3,0 | 2,8 | 2,7 |
| 50 €/MWh | 4,8 | 4,0 | 3,7 | 3,5 | 3,3 | 3,2 |
| 60 €/MWh | 5,3 | 4,5 | 4,2 | 4,0 | 3,8 | 3,7 |
| 75 €/MWh | 6,1 | 5,2 | 5,0 | 4,8 | 4,6 | 4,4 |
| 100 €/MWh | 7,3 | 6,5 | 6,2 | 6,0 | 5,8 | 5,7 |
| 125 €/MWh | 8,6 | 7,7 | 7,4 | 7,2 | 7,0 | 6,9 |

*Table 8: Hydrogen production cost of PEM 160,000 Nm$^3$ system in 2030 in €/kg (own illustration Tenhumberg & Büker, 2020, p. 1593)*

| | 25% | 40% | 50% | 60% | 75% | 90% |
|---|---|---|---|---|---|---|
| 25 €/MWh | 5,3 | 4,0 | 3,5 | 3,2 | 2,9 | 2,7 |
| 40 €/MWh | 6,2 | 4,8 | 4,4 | 4,1 | 3,8 | 3,6 |
| 50 €/MWh | 6,7 | 5,4 | 4,9 | 4,6 | 4,3 | 4,1 |
| 60 €/MWh | 7,3 | 5,9 | 5,5 | 5,2 | 4,9 | 4,6 |
| 75 €/MWh | 8,1 | 6,7 | 6,3 | 6,0 | 5,7 | 5,5 |
| 100 €/MWh | 9,4 | 8,1 | 7,6 | 7,3 | 7,0 | 6,8 |
| 125 €/MWh | 10,8 | 9,5 | 9,0 | 8,7 | 8,4 | 8,2 |

*Table 9: Hydrogen production cost of AEL 160,000 Nm$^3$ system in 2030 in €/kg (own illustration based onTenhumberg & Büker, 2020, p. 1593)*

| | 25% | 40% | 50% | 60% | 75% | 90% |
|---|---|---|---|---|---|---|
| 25 €/MWh | 4,0 | 3,0 | 2,6 | 2,4 | 2,2 | 2,0 |
| 40 €/MWh | 4,6 | 3,6 | 3,2 | 3,0 | 2,8 | 2,6 |
| 50 €/MWh | 5,0 | 4,0 | 3,6 | 3,4 | 3,2 | 3,0 |
| 60 €/MWh | 5,4 | 4,4 | 4,0 | 3,8 | 3,6 | 3,4 |
| 75 €/MWh | 6,0 | 5,0 | 4,6 | 4,4 | 4,2 | 4,0 |
| 100 €/MWh | 7,0 | 6,0 | 5,6 | 5,4 | 5,2 | 5,0 |
| 125 €/MWh | 8,0 | 7,0 | 6,6 | 6,4 | 6,2 | 6,0 |

The data provided above shows that the highest hydrogen production costs result from the PEM electrolysis because of the highest investment costs and highest electric energy demand. AEL and SOEL both allow lower production cost, as AEL allows for the lowest investment costs and SOEL the lowest electricity demand. For the final technology selection, further criteria must be considered, as for example the specific location and available electricity sources, as well as the requirements of the planned production schedule (Tenhumberg & Büker, 2020, pp. 1592-1594). For more details on the underlying assumptions see Appendix 7 and Appendix 8.

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## 4.3. $CO_2$ utilisation

The term $CO_2$ utilisation describes the further processing of the previously described $CO_2$ and $H_2$ to a new chemical feedstock, that can be further applied. For these processes, there are rather mature value chains available: Fischer-Tropsch synthesis, methanol production and methanol production (Quarton & Samsatli, 2020, p. 5). For the purpose of the C2PAT demonstration project, the Fischer-Tropsch (FT) route was selected. Thus, this technology route will be examined in the following section to arrive at an estimation for the demand of $H_2$ required, for the amount of $CO_2$ as shown in section 3.3.1. Before the actual FT synthesis, focus will be put on the reverse water gas shift reaction, as the FT process requires not $CO_2$, but synthesis gas as feedstock.

### 4.3.1. Syngas production via reverse water gas shift reaction

The reverse water gas shift (rWGS) reaction provides a technological solution to convert $CO_2$ to synthesis gas (also called syngas), i.e. carbon monoxide (CO) and hydrogen ($H_2$), thus providing a valuable feedstock for different sorts of industrial applications. The rWGS is an endothermic catalytic reaction, where $CO_2$ reacts with hydrogen to CO and $H_2O$ in equal moles as can be seen in the following formula (Brown, et al., 2021):

$$CO_2 + H_2 \rightarrow CO + H_2O \qquad \text{(see 9)}$$

At the same time, the rWGS shift reaction is competing with the $CO_2$ methanation reaction, as shown in formula (20). $CO_2$ methanation is an exothermic reaction and thus occurring at lower temperatures than the rWGS shift reaction, i.e. 200-500 °C (Brown, et al., 2021):

$$CO_2 + 4H_2 \rightarrow CH_4 + 2H_2O \qquad (20)$$

Only temperatures above 500 °C provide suitable conditions for the rWGS reaction, and only at 700 °C carbon monoxide (CO) becomes the dominant product of these variety of reactions. Regarding temperature, atmospheric pressure is favourable for rWGS, while $CO_2$ methanation increases with pressure. The following figures demonstrate these effects on the selectivity of $CH_4$ in the methanation reaction and the CO selectivity in the rWGS reaction at a $H_2$ to $CO_2$ Ratio of 4:1, at a constant temperature of 700 °C when varying pressure (left diagram) and at a constant pressure of 1.01 bar when varying temperature (right diagram):

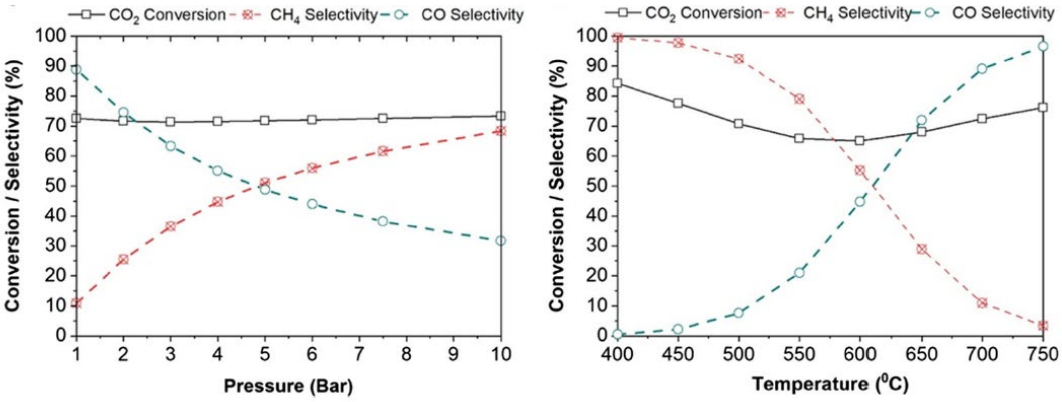

*Figure 29: Effect of pressure and temperature on CO selectivity in rWGS (Brown, et al., 2021)*

The application of different catalysts is still being explored, especially in regard to increasing the share of desired rWGS reactions as opposed to the undesired $CO_2$ methanation process (Schmidt, et al., 2016, p. 27; Brown, et al., 2021). For a general overview, a typical process setup with a rWGS reaction to convert $CO_2$ and $H_2$ to syngas, followed by a FT synthesis is depicted in the following figure:

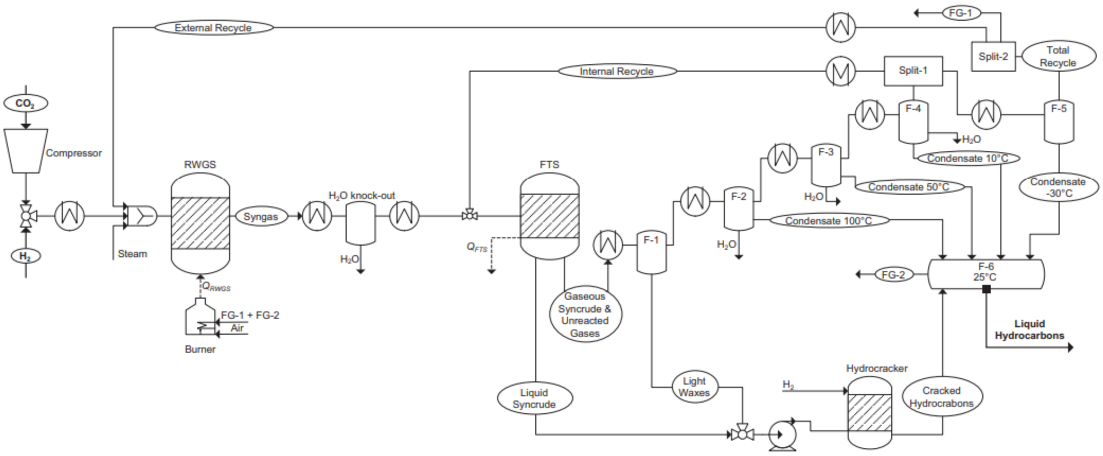

*Figure 30: Flowsheet of a rWGS reaction and FT-synthesis (König, et al., 2015, p. 291)*

Especially from the high heat requirements, it is evident, that a low carbon energy or heat source need to be provided locally in order to implement this process step efficiently (González-Castaño, et al., 2021, p. 956).

As already shown in formula (9), in the rWGS reaction basically occurs, if the input factors $CO_2$ and $H_2$ are provided in a ratio of 1 to 1. As mentioned before, by adapting this input ratio, the resulting output can be varied, depending on the further usage of the syngas. Three different options are summarized in the following table:

*Table 10: Optimal output ratio for rWGS depending on target application (Brown, et al., 2021)*

| **Target application** | **Target output** | **Optimum temperature** | **Optimum $H_2$:$CO_2$ ratio** |
|---|---|---|---|
| Methanol synthesis | $CO_2$: ~10%<br>CO: ~ 14-18%<br>$H_2$: ~ 70%<br>$CH_4$: < 4% | 665 °C | 4:1 |
| FT synthesis | $H_2$:CO ratio: ~2:2<br>Min. $CO_2$ and $CH_3$ | 750 °C | 2:1 |
| CO production | Max. CO | 750 °C | 1:1 |

### 4.3.2. Hydrocarbon production via Fischer-Tropsch synthesis

The origins of the Fischer-Tropsch (FT) synthesis date back to the early 1920s, when Franz Fischer and Hans Tropsch developed the production of a synthetic oil. Fundamentally speaking the FT synthesis produces syncrude, which is a mixture of liquid hydrocarbons with different chain lengths with up to 60 carbon atoms. In a stepwise conversion with a metallic catalyst, first $CH_2$ intermediates are formed (21), before longer hydrocarbon chains are formed, i.e. paraffins (22) and olefins (23) as well as oxygen-containing components (Gruber, et al., 2021, p. 2282):

$$2H_2 + CO \rightarrow\ (CH_2) + H_2O \tag{21}$$

$$(2n + 1)H_2 + nCO \rightarrow\ C_nH_{2n+2} + nH_2O \tag{22}$$

$$(2n)H_2 + nCO \rightarrow\ C_nH_{2n} + nH_2O \tag{23}$$

The distribution of the different products of the FT synthesis in the syncrude can be described by the Anderson-Schulz-Flory (ASF) distribution, where the mass fraction $W_n$ of hydrocarbons with n carbon atoms can be obtained experimentally by a gas chromatography and the chain growth parameter α from the straight-line proportion of the ASF plot according to the following formula:

$$\log\left(\frac{W_n}{n}\right) = n * \log(\alpha) + log\,\frac{(1-\alpha)^2}{\alpha} \tag{24}$$

The FT process requires carbon monoxide (CO) from the reverse water gas shift process. For the application of FT synthesis, the ratio between $CO_2$ and $H_2$ of 2:1 is very critical and needs to be controlled carefully (Zhou, et al., 2016, pp. 175-176), see also Table 10.

For the selection of the most suitable FT technology for a specific application many design options have to be considered. For the purpose of this paper, these decisions will not be discussed in detail, as they are very specific to the requirement of the desired application. The most critical aspects though are the composition of the syngas in terms of $H_2$ to CO ratio, the syngas purity, catalyst deactivation characteristics and replacement strategy, turndown ration and robustness, steam quality and syn-crude composition and quality (de Klerk, et al., 2013, pp. 71-74).

Regarding the output of Fischer Tropsch synthesis, a variety of different products, as shown in the following table, is produced and can be used for further processing:

*Table 11: Composition of Fischer Tropsch syncrude (Maitlis & de Klerk, 2013, p. 83)*

| **Product fraction** | **Carbon range** | **Compound class** | **Syncrude composition (mass %)** | | |
|---|---|---|---|---|---|
| | | | *Fe-HTFT* | *Fe-LTFT* | *Co-LTFT* |
| Tail gas | $C_1$ | Alkane | 12.7 | 4.3 | 5.6 |
| | $C_2$ | Alkene | 5.6 | 1.0 | 0.1 |
| | | Alkane | 4.5 | 1.0 | 1.0 |
| LPG | $C_3$-$C_4$ | Alkene | 21.2. | 6.0 | 3.4 |
| | | Alkane | 3.0 | 1.8 | 1.8 |
| Naptha | $C_5$-$C_{10}$ | Alkene | 25.8 | 7.7 | 7.8 |
| | | Alkane | 4.3 | 3.3 | 12.0 |
| | | Aromatic | 1.7 | 0 | 0 |
| | | Oxygenate | 1.6 | 1.3 | 0.2 |
| Distillate | $C_{11}$-$C_{22}$ | Alkene | 4.8 | 5.7 | 1.1 |
| | | Alkane | 0.9 | 13.5 | 20.8 |
| | | Aromatic | 0.7 | 0 | 0 |
| | | Oxygenate | 0.2 | 0 | 0 |
| Residue/was | $>C_{22}$ | Alkene | 1.6 | 0.7 | 0 |
| | | Alkane | 0.4 | 49.2 | 44.6 |
| | | Aromatic | 0.7 | 0 | 0 |
| | | Oxygenate | 0.2 | 0 | 0 |
| Aqueous product | $C_1$-$C_5$ | Alcohol | 4.5 | 3.9 | 1.4 |
| | | Carbonyl | 3.9 | 0 | 0 |
| | | Carboxylic acid | 1.3 | 0.3 | 0.2 |

As for the purpose of this thesis, only the respective feedstock for FT synthesis is of relevance, subsequent processing and eventual applications of this FT products will not be further evaluated, but further information is provided by Maitlis & de Klerk (2013) for further reference.

# 5. Results

## 5.1. Results of technological assessment

For the technological assessment the findings of Volsund et al. (2019) provide a solid basis for the comparison of alternative carbon capture technologies with the selected MEA-based absorption technology. While all technologies can be engineered to a similar carbon capture ratio, they show relevant differences for equivalent $CO_2$ avoided, which is more critical, as additional $CO_2$ emissions caused by additional energy demand of the carbon capture unit are also considered. This is especially critical, as the MEA technology also has the highest the specific primary energy consumption for $CO_2$ avoided (SPECCA) with 7.1 $MJ/kg_{CO2}$, compared to 4.1 $MJ/kg_{CO2}$ or lower for the alternative technologies.

These relative disadvantages of the MEA-based absorption technology compared to the other technologies need to be contrasted with the specific requirements regarding retrofitability. Even though post combustion technologies are generally preferable because of the relative ease of retroactive installation in existing plants, MEA is the only technology, that has already been applied in industrial scale applications, thus providing the most profound operation experiences.

## 5.2. Results of economic assessment

The economic assessment performed by Gardarsdottir et al. (2019) shows that MEA is the technology with the lowest CAPEX demand. The additional costs for the carbon capture plant increases the total investment cost of the reference cement plant by only about 37%, while the most CAPEX-intensive technology, which is MAL, requires additionally up to 125% of the original investment costs of the reference plant. This picture is reversed when looking at annual OPEX of the different technologies: MEA has the highest additional OPEX figures, adding about 85% to the OPEX demand of the reference cement plant, followed by MAL with about 73%. Tail-end CaL is the most cost-efficient post-combustion technology based on a mere comparison of annual OPEX. For a more comprehensive view, the previously introduced indicator of total cost of $CO_2$ avoided needs to be considered. Even though higher process contingencies have been in included in the alternative technologies to reflect their respective lower maturity, all but MAL show lower costs per tonne of $CO_2$ avoided compared to MEA. Again, tail-end CaL as the most cost-efficient technology allows approx. 35% lower $CO_2$ avoidance costs than MEA and thus should be able to provide a suitable alternative, once the technological maturity is further advanced.

When looking at the economics of hydrogen production as shown in the work of Tenhumberg & Büker (2019), it becomes evident, that the most decisive factor for the production cost of hydrogen via water electrolysis is not the choice of technology between the three concepts introduced, but to select to best available electricity source in accordance with the requirements of the respective production strategy. From a mere LCOH perspective, AEL seems to be preferable compared to SOEL up to an electricity price level of about 50 €/MWh, where SOEL starts to become the most cost-efficient technology with increasing full load hours – only provided that a source of waste heat is available on-site. The advantages of PEM electrolysers regarding flexibility are to be considered separately, as these additional revenues are very difficult to assess accurately.

## 5.3. Results of Energy demand assessment for hydrogen production

Based on the previous analysis, the electricity demand for the hydrogen production necessary for the decarbonisation of the cement industry in Austria will be calculated to allow for conclusions. As illustrated in Figure 12, the total $CO_2$ emissions of cement production in Austria amount to 2,95 million tonnes of $CO_2$. At a given $CO_2$ capture ratio 90%, 2,66 million tonnes of $CO_2$ could be used as a feedstock for the further carbon utilization route as described previously. The required amount of hydrogen for the optimal transformation of $CO_2$ and $H_2$ to syngas for further processing via FT synthesis, can be calculated based on formula (9), the desired $H_2$:$CO_2$ ratio of 2:1 (as shown in Table 10) and the molar masses of the molecules involved (see Appendix 9): The total hydrogen demand to utilize the $CO_2$ emissions of the Austrian cement production amounts to approx. 0,243 million tonnes of hydrogen (for calculation see Appendix 10). The electricity demand for the production of 0,243 million tonnes of hydrogen can be calculated based on the estimated process efficiencies for the year 2030 based on Figure 28:

*Table 12: Electricity demand for required hydrogen production (own calculation, see Appendix 11)*

| **Electrolysis technology** | **TWh (min.)** | **TWh (max.)** | **TWh (Ø)** |
|---|---|---|---|
| AEL | 11,5 | 12,7 | 12,1 |
| PEM | 12,0 | 14,1 | 13,1 |
| SOEL | 9,7 | 10,0 | 9,9 |

The above electricity demand must be compared to the renewable energy target for 2030 of the Austrian renewable energy strategy (see section 0). Assuming the hydrogen production for an eventual implementation of CCU projects in the cement industry will be based on a mix of the different electrolysis technologies, an average electricity demand of 11,7 TWh will be necessary. The additional 27 TWh envisioned in the renewable energy strategy do not incorporate additional electricity demand for such carbon capture and utilization applications in an industrial scale.

## 6. Conclusion

For the reasons stated in 5.1, the MEA technology proves to be the most suitable choice, especially as it is the most mature technology, and even more so when considering the total scope and complexity of the C2PAT project. But once the alternative post-combustion carbon capture technologies have been further tested and demonstrated, they should be thoroughly considered, as they provided higher efficiencies regarding $CO_2$ abatement, whilst still feasible to retrofit.

A similar picture can be drawn for the technological evaluation of $H_2$ production technologies. While SOEL show the highest efficiency rates, as shown in Figure 28: Electric energy demand of electrolysis technologies Figure 28, and thus provide lower levels of electricity demand, they also have to cope with lowest technological maturity. The TRL still needs to increase significantly and SOEL are ultimately dependent on the availability of an on-site (waste) heat source, which needs to be not just cost-effective, but also low in $CO_2$-emissions, not to jeopardize the fundamental objective of CCU projects.

Concluding the economic assessment based on the arguments made 5.2, the technology selection made in the C2PAT project reflects the current state of art, as

MEA-based absorption is the only suitable mature carbon capture technology, also from an economic perspective. Even though alternative technologies, in particular technologies previously described provide potentials for higher process efficiencies and lower costs, they currently still impose a relatively high operational and thus economic risks, as they are not sufficiently demonstrated in industrial scale applications before.

Furthermore, it became evident, that a holistic evaluation of CCU technologies for the cement industry was not available. Such a holistic evaluation will be elaborated for the first time in the scope of the C2PAT, also incorporating the further utilisation of the FT-products. However, in order to enhance and foster the application of CCU projects, not just for the cement industry but also other hard to abate industrial sectors, further research and comparable studies will be of great interest.

As a consequence of the assessment of the projected energy demand as outlined in 5.3, the decarbonisation of the cement industry in Austria is highly dependent on a sufficient supply of renewable energy and thus requires a further development of renewable energy sources to produce the hydrogen required locally. Alternatively, it will depend on a future hydrogen import infrastructure, which will allow the production of renewable hydrogen in countries with sufficient renewable energy potential. Both, the increase of the renewable energy targets and the eventual import of renewable hydrogen are highly complex endeavours, from technological economic and also ecologic perspectives, and thus require further research to assess and discuss the respective technical, economic and ecologic feasibilities.

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## List of abbreviations

| | |
|---|---|
| a | annum |
| $AC_{eq}$ | equivalent $CO_2$ avoided |
| ASU | air separation unit |
| $e_{clk,eq}$ | specific equivalent $CO_2$ emissions |
| $e_{clk,eq,ref}$ | specific equivalent $CO_2$ emissions in reference plant |
| C | Celsius |
| CaL | Calcium Looping |
| CAP | chilled ammonia process |
| CAPEX | capital expenditures |
| C2PAT | Carbon to product Austria |
| CCR | $CO_2$ capture ratio |
| CCS | carbon capture and storage |
| CCU | carbon capture and usage |
| CCUS | carbon capture, utilisation and storage |
| clk | clinker |
| Co | Compagnie |
| d | day |
| EF | emission factor |
| eq | equivalent |
| IEA | International Energy Agency |
| IPCC | International Panel on Climate Change |
| FT | Fischer-Tropsch |
| GHG | greenhouse gas |
| GmbH | Gesellschaft mit beschränkter Haftung |
| KG | Kommanditgesellschaft |
| kg | kilogram |
| kWh | kilo Watt hour |
| LHV | lower heating value |
| m | meter |
| MAL | membrane-assisted $CO_2$ liquefaction |
| MJ | mega-joule |
| mm | milimeter |
| MEA | monoethanolamine |
| $\dot{m}_{CO2,capt}$ | mass flowrate of $CO_2$ captured |
| $\dot{m}_{CO2,gen}$ | mass flowrate of $CO_2$ generated |
| MFC | Mitsubishi Fluidized-bed Calciner |
| Mt | Mega ton |
| n/a | not applicable |
| NECP | National Climate and Energy Plan |
| OPEX | operational expenditures |

| | |
|---|---|
| PV | photovoltaic |
| ref | reference |
| rWGS | revers water gas shift |
| SPECCA | Specific primary energy consumption for $CO_2$ avoided |
| t/a | tonnes per year |
| t | ton |
| TJ | tera-joule |
| WAM | with additional measures |

## List of Tables

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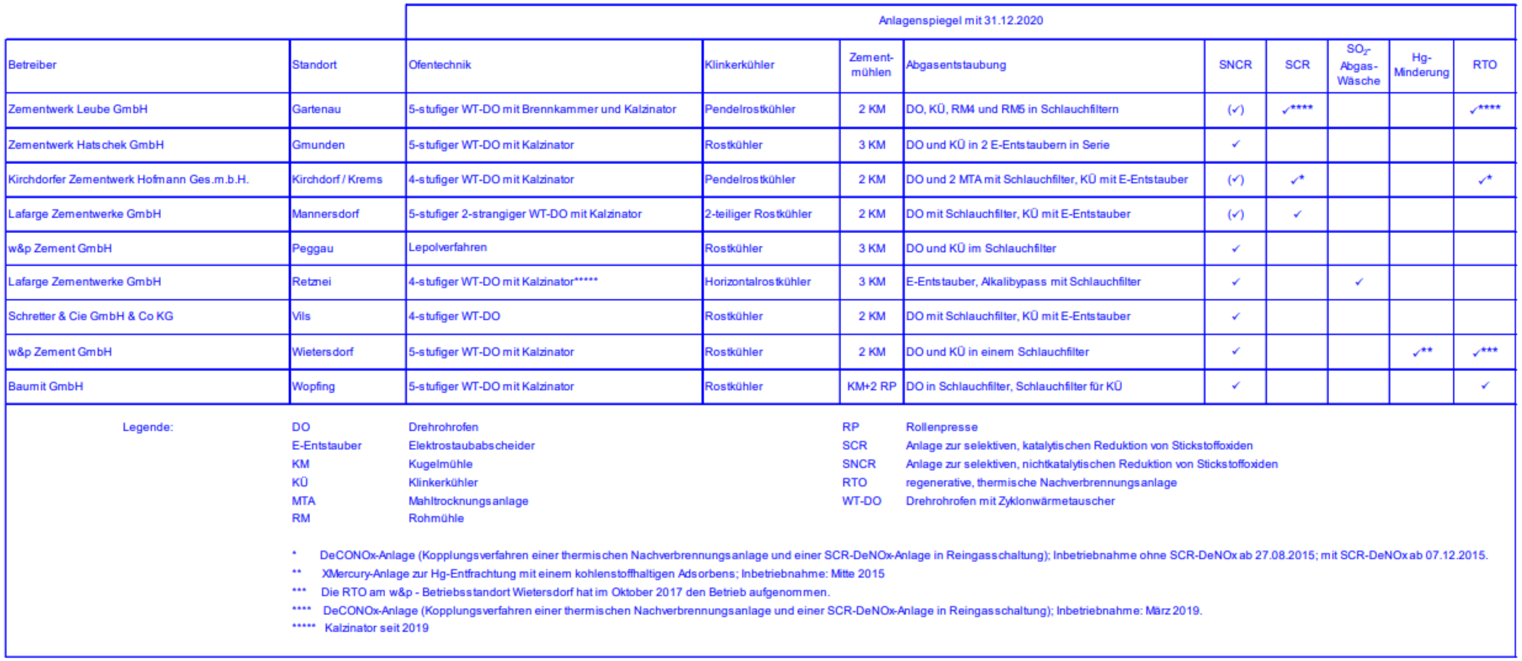
# Appendix

Appendix 1: Detailed overview of cement production plants in Austria (Mauschitz, 2021, p. 5)



| Betreiber | Standort | Ofentechnik | Klinkerkühler | Zement-mühlen | Abgasentstaubung | SNCR | SCR | $SO_2$-Abgas-Wäsche | Hg-Minderung | RTO |
|---|---|---|---|---|---|---|---|---|---|---|
|  |  | Anlagenspiegel mit 31.12.2020 |  |  |  |  |  |  |  |  |
| Zementwerk Leube GmbH | Gartenau | 5-stufiger WT-DO mit Brennkammer und Kalzinator | Pendelrostkühler | 2 KM | DO, KÜ, RM4 und RM5 in Schlauchfiltern | (✓) | ✓**** |  |  | ✓**** |
| Zementwerk Hatschek GmbH | Gmunden | 5-stufiger WT-DO mit Kalzinator | Rostkühler | 3 KM | DO und KÜ in 2 E-Entstaubern in Serie | ✓ |  |  |  |  |
| Kirchdorfer Zementwerk Hofmann Ges.m.b.H. | Kirchdorf / Krems | 4-stufiger WT-DO mit Kalzinator | Pendelrostkühler | 2 KM | DO und 2 MTA mit Schlauchfilter, KÜ mit E-Entstauber | (✓) | ✓* |  |  | ✓* |
| Lafarge Zementwerke GmbH | Mannersdorf | 5-stufiger 2-strangiger WT-DO mit Kalzinator | 2-teiliger Rostkühler | 2 KM | DO mit Schlauchfilter, KÜ mit E-Entstauber | (✓) | ✓ |  |  |  |
| w&p Zement GmbH | Peggau | Lepolverfahren | Rostkühler | 3 KM | DO und KÜ im Schlauchfilter | ✓ |  |  |  |  |
| Lafarge Zementwerke GmbH | Retznei | 4-stufiger WT-DO mit Kalzinator***** | Horizontalrostkühler | 3 KM | E-Entstauber, Alkalibypass mit Schlauchfilter | ✓ |  | ✓ |  |  |
| Schretter & Cie GmbH & Co KG | Vils | 4-stufiger WT-DO | Rostkühler | 2 KM | DO mit Schlauchfilter, KÜ mit E-Entstauber | ✓ |  |  |  |  |
| w&p Zement GmbH | Wietersdorf | 5-stufiger WT-DO mit Kalzinator | Rostkühler | 2 KM | DO und KÜ in einem Schlauchfilter | ✓ |  |  | ✓** | ✓*** |
| Baumit GmbH | Wopfing | 5-stufiger WT-DO mit Kalzinator | Rostkühler | KM+2 RP | DO in Schlauchfilter, Schlauchfilter für KÜ | ✓ |  |  |  | ✓ |

Legende:

| | | | |
|---|---|---|---|
| DO | Drehrohrofen | RP | Rollenpresse |
| E-Entstauber | Elektrostaubabscheider | SCR | Anlage zur selektiven, katalytischen Reduktion von Stickstoffoxiden |
| KM | Kugelmühle | SNCR | Anlage zur selektiven, nichtkatalytischen Reduktion von Stickstoffoxiden |
| KÜ | Klinkerkühler | RTO | regenerative, thermische Nachverbrennungsanlage |
| MTA | Mahltrocknungsanlage | WT-DO | Drehrohrofen mit Zyklonwärmetauscher |
| RM | Rohmühle | | |

* DeCONOx-Anlage (Kopplungsverfahren einer thermischen Nachverbrennungsanlage und einer SCR-DeNOx-Anlage in Reingasschaltung); Inbetriebnahme ohne SCR-DeNOx ab 27.08.2015; mit SCR-DeNOx ab 07.12.2015.

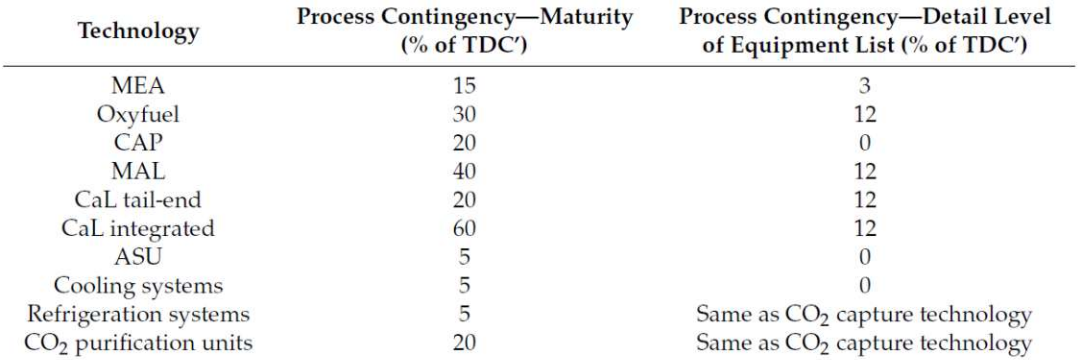
** XMercury-Anlage zur Hg-Entfrachtung mit einem kohlenstoffhaltigen Adsorbens; Inbetriebnahme: Mitte 2015

*** Die RTO am w&p - Betriebsstandort Wietersdorf hat im Oktober 2017 den Betrieb aufgenommen.

**** DeCONOx-Anlage (Kopplungsverfahren einer thermischen Nachverbrennungsanlage und einer SCR-DeNOx-Anlage in Reingasschaltung); Inbetriebnahme: März 2019.

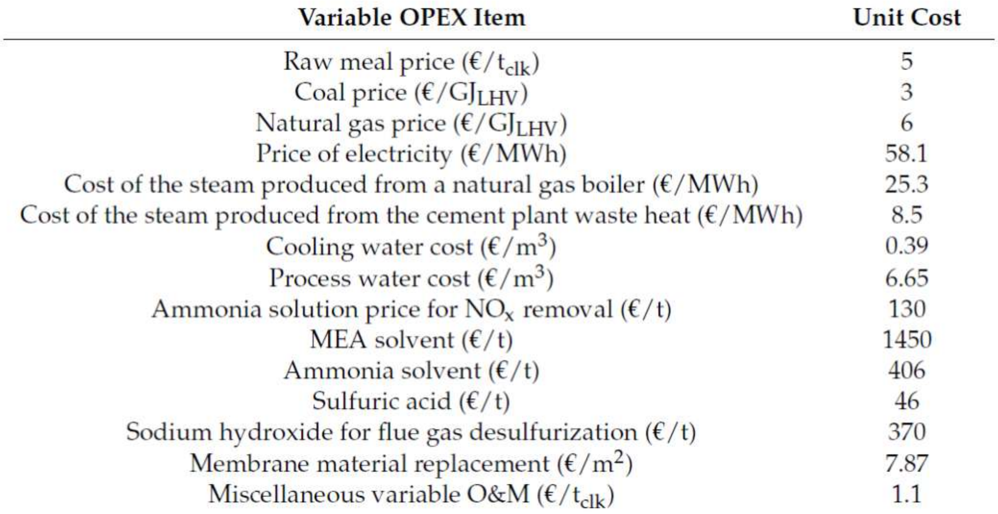
***** Kalzinator seit 2019

*Appendix 2: Process contingency factors for $CO_2$ capture technologies and subsystems (Gardarsdottir, et al., 2019, p. 7)*



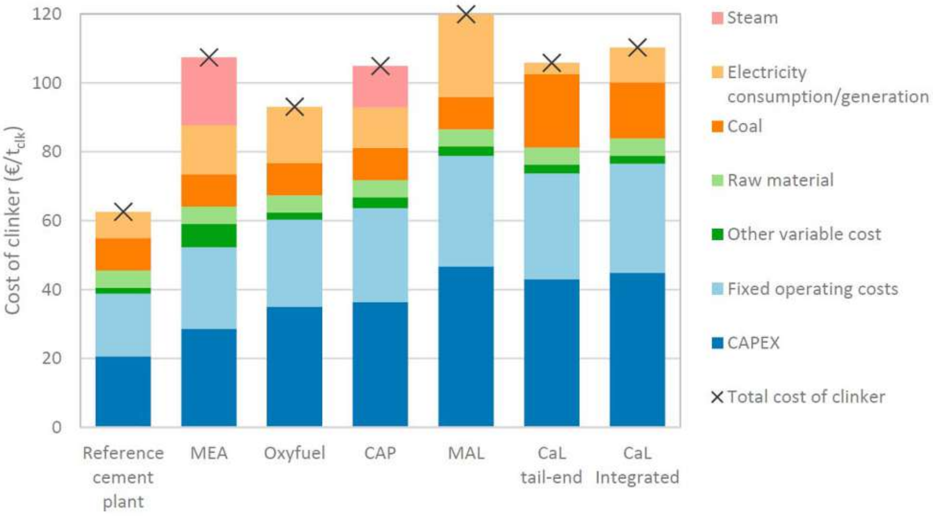
| Technology | Process Contingency—Maturity (% of TDC′) | Process Contingency—Detail Level of Equipment List (% of TDC′) |
|---|---|---|
| MEA | 15 | 3 |
| Oxyfuel | 30 | 12 |
| CAP | 20 | 0 |
| MAL | 40 | 12 |
| CaL tail-end | 20 | 12 |
| CaL integrated | 60 | 12 |
| ASU | 5 | 0 |
| Cooling systems | 5 | 0 |
| Refrigeration systems | 5 | Same as $CO_2$ capture technology |
| $CO_2$ purification units | 20 | Same as $CO_2$ capture technology |

*Appendix 3: Unit cost of variable OPEX (Gardarsdottir, et al., 2019, p. 8)*

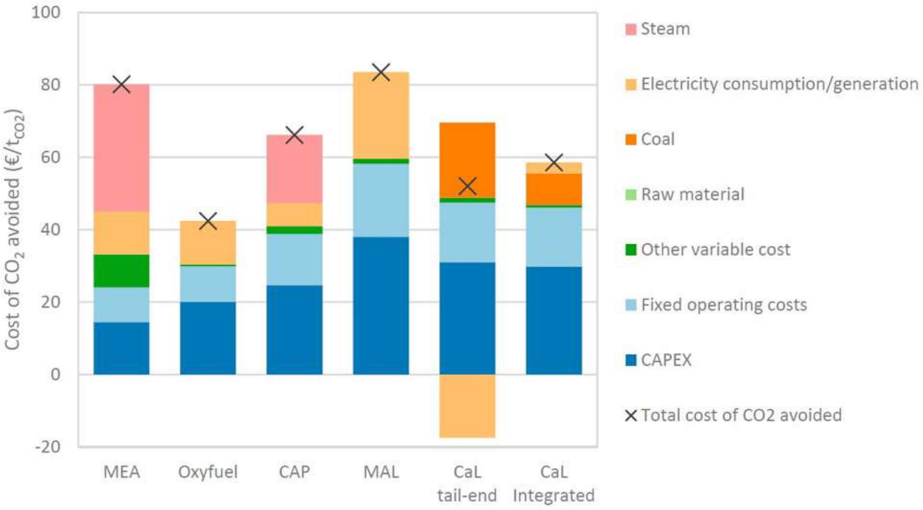


| Variable OPEX Item | Unit Cost |
|---|---|
| Raw meal price (€/$t_{clk}$) | 5 |
| Coal price (€/$GJ_{LHV}$) | 3 |
| Natural gas price (€/$GJ_{LHV}$) | 6 |
| Price of electricity (€/MWh) | 58.1 |
| Cost of the steam produced from a natural gas boiler (€/MWh) | 25.3 |
| Cost of the steam produced from the cement plant waste heat (€/MWh) | 8.5 |
| Cooling water cost (€/$m^3$) | 0.39 |
| Process water cost (€/$m^3$) | 6.65 |
| Ammonia solution price for $NO_x$ removal (€/t) | 130 |
| MEA solvent (€/t) | 1450 |
| Ammonia solvent (€/t) | 406 |
| Sulfuric acid (€/t) | 46 |
| Sodium hydroxide for flue gas desulfurization (€/t) | 370 |
| Membrane material replacement (€/$m^2$) | 7.87 |
| Miscellaneous variable O&M (€/$t_{clk}$) | 1.1 |

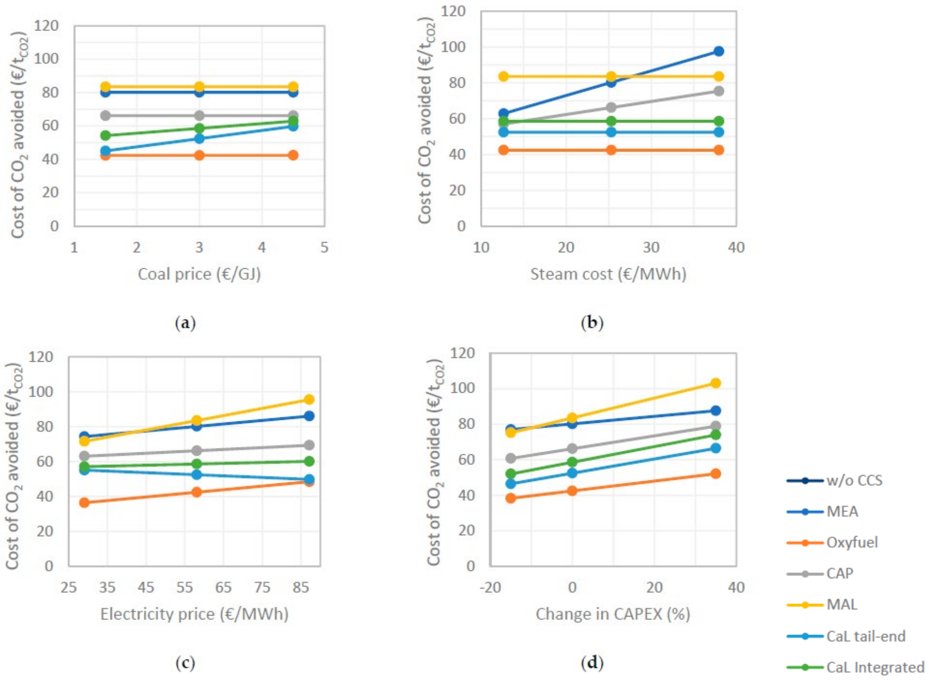
*Appendix 4: Breakdown of COC for the reference cement plant and investigated $CO_2$ capture technologies. (Gardarsdottir, et al., 2019, p. 10)*



*Appendix 5: Breakdown of CAC for the reference cement plant and all the investigated $CO_2$ capture technologies. (Gardarsdottir, et al., 2019, p. 11)*



*Appendix 6: Sensitivity of CAC to (a) coal price, (b) stream cost, (c) electricity cost and (d) CAPEX (Gardarsdottir, et al., 2019, p. 13)*



*Appendix 7: Total energy demand of electrolysis technologies for 2030 (Tenhumberg & Büker, 2020)*

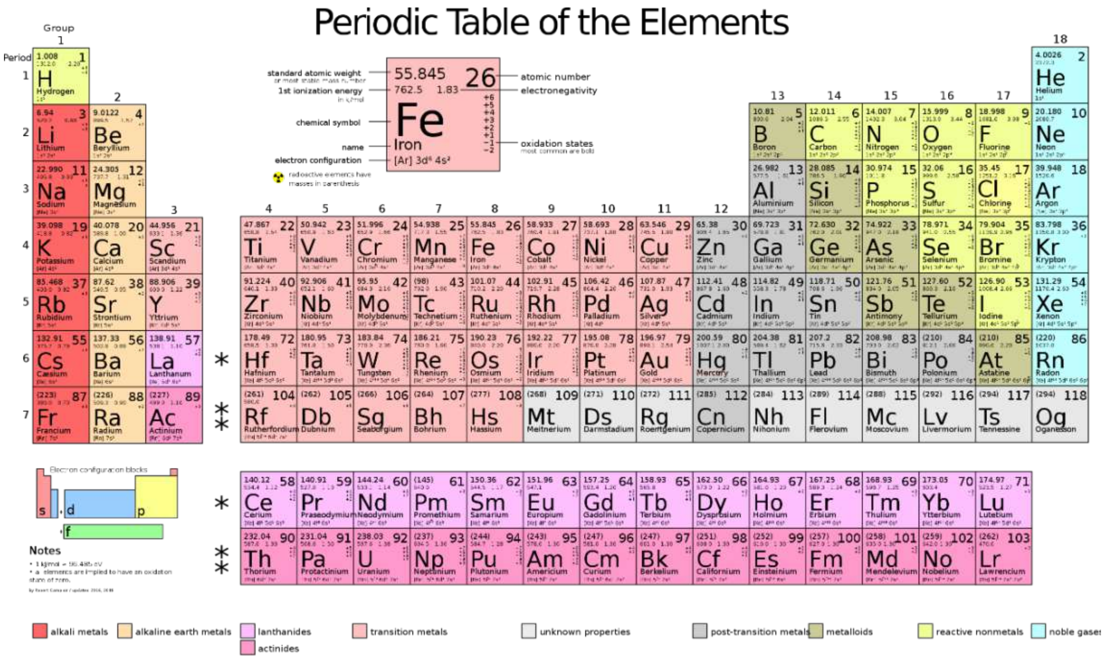
| | | AEL | PEM | SOEC | Integrated C2C-SOEC |
|---|---|---|---|---|---|
| Operating pressure | [bar] | 20[a] | 30[a] | 10[c] | 10[c] |
| Energy demand | | | | | |
| Electrical energy | $[kWh/Nm^3\ H_2]$ | 4.42[a] | 4.81[a] | 3.55[a] | 3.55[a] |
| Thermal energy demand | $[kWh/Nm^3\ H_2]$ | | | 0.70[c] | 0.26 $kWh_{el}$[e] |
| $H_2$ compression to 30 bar | $[kWh/Nm^3\ H_2]$ | 0.02[b] | | 0.06[d] | 0.06[d] |
| Total energy demand | $[kWh/Nm^3\ H_2]$ | 4.44 | 4.91 | 4.31 | 3.87 |

a) Smolinka et al. Studie IndWEDe, 2018 [1]; b) Calculated energy demand for compression of hydrogen from 20 to 30 bar; c) Calculated thermal energy demand for steam generation (150°C, 3 bar); d) Calculated energy demand for compression of hydrogen from 10 to 30 bar; e) Additional electricity demand as compensation for steam generation in power plant.

*Appendix 8: Assumptions made in calculating hydrogen production costs (Tenhumberg & Büker, 2020)*

| | | AEL | PEM | SOEC |
|---|---|---|---|---|
| Capital expenditure (CAPEX) [3] | [€/kW] | 450 | 810 | 800 |
| Share of costs relating to stacks [3] | [% CAPEX] | 40 | 51 | 30 |
| $H_2$ compressor (130,000 $Nm^3/h$) | [Mio €] | 10 | -- | 20 |
| Service and maintenance (system) [3] | [€/kW] | 21.9 | 11.3 | 11.3 |
| Stack lifetime [3] | [h] | 82,500 | 60,000 | 50,000 |
| Natural gas | [€/mmBtu] | - | - | 2.45 |
| Water | [€/t] | 1.0 | | |
| General plant overhead | [% CAPEX] | 1.5 | | |
| Taxes and insurance | [% CAPEX] | 1.5 | | |
| Depreciation period | [Years] | 10 | | |
| Debt interest rate p.a. | [%] | 5 | | |

*Appendix 9: Periodic table of chemical elements (Wikipedia, 2022)*



*Appendix 10: Calculation of total hydrogen demand based on molar mass*

**MOLAR MASS**

| | | |
|---|---|---|
| H | 1,00794 | g/mol |
| C | 12,0107 | g/mol |
| O | 15,9994 | g/mol |
| H2 | 2,01588 | g/mol |
| 2H2 | 4,03176 | g/mol |
| CO2 | 44,0095 | g/mol |

| | |
|---|---|
| Mass factor H2 to CO2 | 0,0458 |
| Mass factor H2 to CO2 ratio 2:1 | 0,0916 |

| | | |
|---|---|---|
| Total CO2 emission from cement production in Austria | 2,95 | million t |
| carbon capture rate | 90% | |
| usable CO2 emissions | 2,66 | million t |
| H2 demand based on mass factor (2:1) | 0,243 | million t |

*Appendix 11: Calculation of electricity demad for required hydrogen production*

| | MWh/t (min.) | MWh/t (max.) | TWh max.) | TWh (min.) |
|---|---|---|---|---|
| **AEL** | 47,1 | 52,4 | 11,5 | 12,7 |
| **PEM** | 49,5 | 58,1 | 12,0 | 14,1 |
| **SOEL** | 40 | 41,2 | 9,7 | 10,0 |