Introduction of an Innovative Energy Concept for low Emission Glass Melting based on Carbon Capture and Usage

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Abstract: Due to the very high fossil energy demand, the glass industry is looking for innovative approaches for the reduction of CO_2 emissions and the integration of renewable energy sources. In this paper, we present a novel power-to-gas concept, which has no impact on established melting processes and discuss it for this purpose. A special focus is set on the required CO_2 capture from typical flue gases in the glass industry, as this process has not been investigated in detail yet. We used a process simulation approach to investigate post-combustion CO_2 capture by absorption processes, followed by a techno-economic evaluation. Our investigations found the designed CO_2 capture plant to be approx. 400 times smaller than absorption based CO_2 separation processes for conventional power plants. Due to the many options for waste heat utilization, the waste heat required for CO_2 desorption can be generated in a particularly efficient and cost-effective way. The resulting CO_2 avoidance costs range between 41-42 €/t CO_2, depending on waste heat utilization for desorption, and thus offer a cost effective way of CO_2 removal from glass industry melting processes. These costs are well below the values of 50-65 €/t CO_2 described so far for comparable industrial applications. In addition, we describe optimization options, like solvent and process improvements, to enable further cost reductions. These results motivate further research and development on the overall process presented in this work.

Keywords: Power-to-Gas, methanation, oxyfuel, glass industry, CO_2-separation, economic evaluation

1 Introduction

Commercial glass production is a very energy-intensive industrial process. Converting the raw materials, such as silica sand, sodium carbonate, lime, dolomite and others into molten glass requires high process temperatures of up to 1600 °C. This melting process is the central phase of glass production and accounts for 50-80 % of total energy consumption in overall glass production. The dominating source for achieving the required process temperatures has been the combustion of fossil fuels such as natural gas (NG) and crude oil for a long time. [1]

Due to this dominance of fossil fuels, glass production is currently still associated with high carbon dioxide (CO_2) emissions. In 2007, about 27.5 million tonnes CO_2-equivalent (eq.) were emitted within the European Union, the world's largest glass producer (approx. 30 % of world output). Approx. 17 mio. t CO_2-eq. of these emissions, i.e. 81 %, are exclusively due to emissions from combustion for the melting process [2]. Although there has been a slight reduction in CO_2 emissions since then, it fails to meet climate protection targets. The causes of this decline also include economic fluctuations and consumer demand. For example, the level of these emissions in Germany has stagnated at a constant level of about 4 mio. t CO_2-eq. per year since 2007 [3].

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Of course, there are efforts towards increased sustainability and climate protection in the glass industry. Accordingly, there are many research approaches and developments for decarbonization and the integration of renewable energies into the glass melting process. The most important concepts are all-electric melting and the switch to hydrogen (H_2) as combustion fuel instead of NG [4]. However, all of these concepts, despite their promising CO_2 emissions reduction potential, have certain restrictions:

All-electric melting is well established at small scale glass melting systems, but large scale applications are still controversial [5]. Recent all-electric melting projects have improved and could reach melting capacities of up to 250 t/d. However, complex melting tank design and extensive heat control strategies of such large scale melting tanks are challenging and result in high investment and operating costs [6]. In addition, low glass production rate (so called pull rate) flexibility, short melting tank lifetime, high electricity costs and low operating experience are disadvantages. Besides that, not all glass types are feasible for all electric melting, for example non-ionic glasses [4].

Recent projects have investigated hydrogen as a fuel substitution option, like HyGlass [7], HyNet [8] and Koper-nikus P2X [9]. Since the combustion of H_2 does not cause any CO_2 emissions, this would significantly reduce the CO_2 footprint of glass melting. However, this reduction highly depends on the way of hydrogen production (green, blue, yellow,... hydrogen). So far, only green H_2 can guarantee a high reduction of CO_2 emissions if electricity comes exclusively from renewable energies, as for example blue H_2 also comes with 9-12 % less CO_2 emissions than conventional (= grey) hydrogen [10]. However, green H_2 has been cost-intensive and therefore not considered to be economically competitive with established fossil fuels. By contrast, recent price dynamics on the large-scale markets may indicate a turnaround in terms of cost competitiveness of green H_2. Nevertheless, H_2 shows much different combustion behavior, like higher flame temperatures, different flame velcoity, faster ignition behavior and changed heat radiation properties. Some of these effects are compensated by oxyfuel combustion, for example changes in adiabatic flame temperature, as shown in [6]. Nevertheless, there are still some questions unanswered, such as effects of higher water content in the flue gases in the firing chamber on glass quality, and melting tank life time.

Due to these limitations of currently discussed decarbonisation options, there is an urgent need for innovative energy concepts which allow the integration of renewable energies, a reduction of CO_2 emissions, and which have ideally no impact on established melting processes. This work is the first to demonstrate the integration of a power-to-methane (PtM) system into the glass melting process that meets all of these requirements. Since the PtM concept has been known for a long time as a sector coupling option in energy systems [11, 12], there are a lot of established technical options for most of the process steps. The novelty of these established options is briefly described, before the remaining open process of CO_2 capture from exhaust gases is discussed in more detail. In addition, a techno-economic analysis is provided, highlighting the economic aspects of CO_2 separation for this specific application.

2 Integration of Power-to-Methane into Oxyfuel Glass Melting

Figure 1 shows a simplified process flow sheet for the integration of PtM into oxyfuel glass melting processes. In this section, each process unit in the flow sheet is described, along with established technical options for its technical implementation. We paricularly focus on CO_2 separation from the flue gases of oxyfuel glass melting tanks, since this process has not been investigated yet.

2.1 Electrolysis

In the first process unit, water (H_2O) is separated into (H_2) and oxygen (O_2) by electrolysis. Both H_2 and O_2 are subsequently stored to overcome the intermittent supply of renewable electricity from wind or solar power (sections 2.2). Besides H_2, also O_2 is an important product for the downstream PtM processes, as it can be used in oxyfuel combustion (section 2.4).

Alkaline (AEL), solid oxide (SOEC) and polymer electrolyte membrane electrolysis (PEMEL) are currently considered as the technically most established processes for this purpose [11, 13]. AEL has the highest

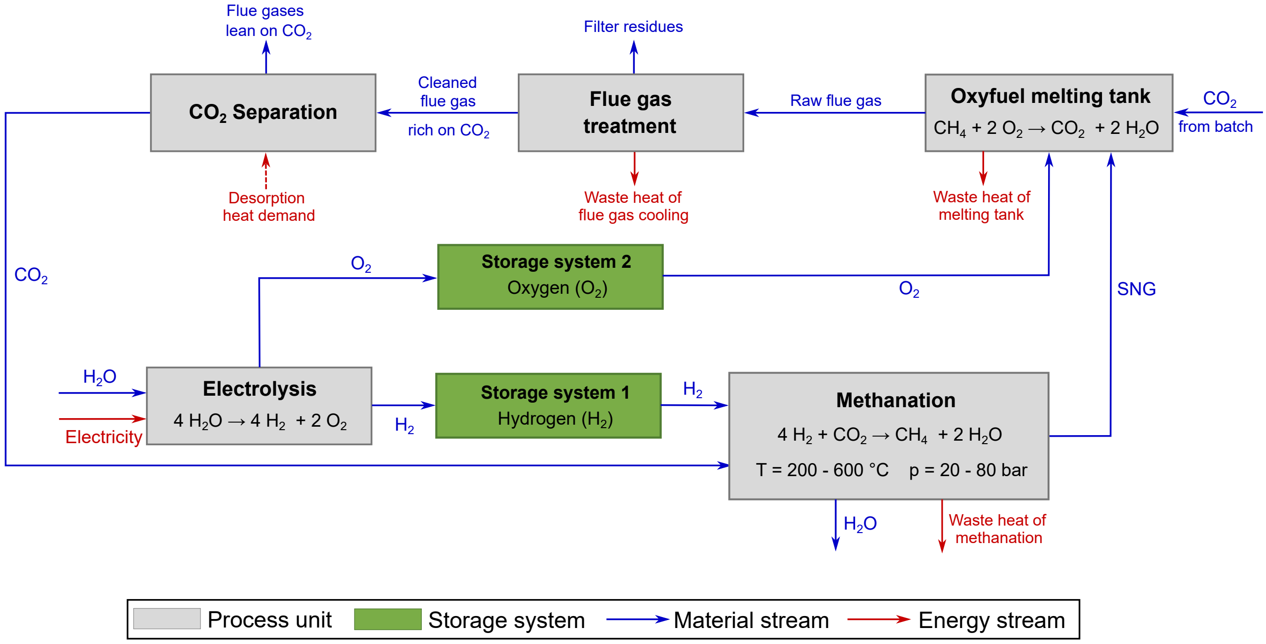


Figure 1: Simplified flow sheet of the integration of Power-to-Methane (PtM) into oxyfuel glass melting processes.

technology readiness level (TRL) 9, but offers only limited part-load capability (30-100 %) due to thermal restrictions [13]. SOEC would offer the highest efficiency of all options (up to 95 % [14]), but requires a operating temperature level of 600-1000 ℃ [13]. These temperatures can be provided in an energy-efficient way by integrating waste heat from catalytic methanation. However, heating and cooling creates a thermal inertia of SOEC, which impedes rapid adaptation to fluctuations of renewable energy sources. In addition, the TRL of 5-6 is the lowest of all technical options described here. The most promising technology for the application proposed in this work is PEMEL, as it has the widest part-load capability (0-100 %) and can thus adapt to the fluctuations of renewable energy sources. With a slightly lower TRL of 7, PEM is behind AEL in terms of development and availability [13]. Nevertheless, PEM electrolysers are already available in power categories of multiple MW (e.g. Siemens Silyzer 300 [15]). Development potential is primarily focused on the industrial series production of this electrolysis technology, which is expected to result in a significant reduction of costs [11].

2.2 Gas Storage

The intermediate storage of H_2 (storage system 1 in Figure 1) is necessary because there are currently technological restrictions on the flexibility of the down-stream catalytic methanation process (see section 2.3). In general, there are various options for H_2 storage:

- Physical storage technologies like compressed gas, liquid, or cryo-compressed H_2 storage [16];
- adsorption technologies, relying on carbon based materials like multi-walled carbon nanotubes (MWCNTs), metal organic frameworks (MOFs) or zeolites [16];
- absorption technologies, based on metal hydrates, like iron-oxide pellets [17].
- chemical H_2 storage like liquid organic hydrogen carriers (LOHCs) [18],

For the process described in this work, H_2 storage in pressurized gas tanks is currently the most suitable option. It is described as technologically mature and therefore a comparatively cost-effective option, especially in combination with battery storage systems [16]. Nevertheless, the application of innovative H_2 storage technologies within the described PtM process, should be focused in further research. The high waste heat potential in glass melting processes offers an attractive option for an energy-efficient use of LOHC or iron-oxide based H_2 storage technologies.

Storage system 2 in Figure 1 is necessary to ensure a constant flow of O_2 for a stable combustion process in the melting tank. Various options for O_2 storage exist at industrial scale since a long time and can therefore be considered robust [19]. So far, the O_2 required for oxyfuel combustion in the glass industry has been produced primarily by air separation using vacuum pressure swing adsorption (VPSA) processes [19]. This is done either directly at the industrial site, or by delivery and storage in compressed gas tanks (liquid O_2). In both cases, oxygen storage facilities are usually available on site.

Methanation, melting furnace, flue gas conditioning and CO_2 capture can be considered as stationary processes with only minor transient behavior. However, a constant educt gas flow is required for a stable thermo-cathalytic methanation process. Nevertheless, a constant educt gas flow is required for stable methanation. In order to compensate for the slight fluctuations in the product stream of the CO_2 capture plant, a CO_2 storage system may also be required. The storage of CO_2 is a long-proven and available technology and can therefore be regarded as mature [20]. Due to the comparatively low storage capacity required, no further details of this storage system are discussed in this work.

2.3 Methanation

Thermocatalytic and biological methanation are the main tech currently used in power to gas systems [13, 21]. Due to the higher TRL and technical performance, catalytic methanation is mainly used in large-scale PtM plants [11].

However, this methanation process requires constant operating conditions to achieve a high CH_4 content in the synthetic natural gas (SNG) [13, 21]. For this purpose, the educt gases CO_2 and H_2 must be supplied in the ideal stoichiometric ratio of 1:4. In addition, pressure and temperature conditions must be constant to avoid damages on the catalysts through hot-spot formation. From a thermodynamic point of view, low temperatures and high pressure would be ideal for a high conversion rate. However, the exothermic nature of the dominant methanation reactions challenges the operation at thermodynamic optimum. For many commercially available fixed-bed reactors, operating temperatures of 300 ℃ and pressures of 20 bar have proven to be suitable operating conditions. The operating temperatures of established reactor concepts vary between 200-600 ℃ and a pressures range of 20-80 bar [13, 21]. In any case effective cooling of the reactor is required. In the PtM system described above, this waste heat could be used for SOEC or CO_2-desorption (see section 2.7).

In addition to the technical properties mentioned above, the purity of CO_2 is crucial for a stable methanation process. In order to investigate the effects of CO_2 impurities on methanation, experiments investigating direct methanation of flue gases for different types of power plants have been performed. The flue gases investigated were obtained both from lignite-fired power plants with conventional combustion and from pilot plants with oxyfuel combustion. In both cases, direct methanation of the flue gases led to heavy damage on the catalyst, so that the experiments had to be stopped. However, oxyfuel flue gases could ensure stable methanation after purification and treatment. Within these investigations, mainly SO_2 and halogen compounds (F, Cl, Br, At, TS) have been described as strong acting catalyst toxicants [22, 23].

2.4 Oxyfuel Glass Melting Tank

In oxyfuel combustion processes, a fuel like natural gas is burned in a pure oxygen atmosphere instead of ambient air. Oxyfuel combustion is more energy-efficient because nitrogen content in the furnace atmosphere is significantly reduced, thus lowering the thermal capacity of the exhaust gases. This also enables improved mass transport in the combustion chamber. In addition, a higher adiabatic flame temperature is achieved [24]. The higher overall efficiency of this combustion technology also enables a reduction in CO_2 emissions compared to the regenerative processes that are primarily used at present. A major limitation of this technology has been the energy-intensive production of oxygen and the associated higher investment costs. Oxyfuel

Table 1: Critical parameters of the melting system investigated in [25].

Parameter	Value
Melting technology	oxyfuel
Operating temperature	1600 °C
Thermal power	6.08 MW
Fuel	Natural Gas H
Glass type	borosilicate
Nominal pull rate	40 t/d
Operating hours	8760 h/year

furnaces are established state-of-the-art despite these limitations, especially in the special glass production sector. Due to the much more energy-intensive glasses that are molten for this purpose, the higher efficiency of oxyfuel melting offers an economic advantage [6].

2.5 Flue Gas Purification Systems

Currently, mainly fabric filters and electrostatic precipitators are used for flue gas purification in the glass industry. These cleaning systems are primarily aimed for compliance with air pollutant limits set by country-specific regulations. The air pollutants relevant for the glass industry are mostly dust resulting from turbulence in the furnace, carbon monoxide (CO), sulfur and nitrogen oxides (SO_x, NO_x), as well as hydrochloric and fluoric acid (HCl, HF). While NO_x and CO emissions are mainly caused by combustion, SO_x-, HCl- and HF-emissions occur due to glass batch impurities. To remove these air pollutants from the flue gas mixture, sorbents such as lime (calcium hydroxide, $Ca(OH)_2$) are used, which then deposit as so-called filter dust. In order to prevent the filter systems from being damaged by excessively high gas temperatures, a heat exchanger is usually installed upstream to cool the uncleaned flue gases. Especially fabric filters are much more sensitive to high gas temperatures than electrostatic filters. In addition, cooling by ambient air leaks in the flue gas system is common.

2.6 Flue Gas Properties and Composition

Detailed data on the flue gas composition and properties must be available to enable an evaluation of the CO_2 separation process. However, the composition of flue gases in the glass industry is highly variable. The main influencing factors are: batch composition, cullet fraction, glass type, melting technology and fuel, as well as flue gas treatment and cooling. Therefore, we conducted a literature screening for data from flue gas analyses of melting systems, suitable for the application of the PtM process presented in this paper. We identified the exhaust gas study by Roger et al. [25] was identified as the most promising source.

In this study, flue gas analyses were performed on several special glass melting tanks. The aim was to investigate the potential for removal of boron compounds (namely boric acid (H_3BO_3) and meta boric acid (HBO_2)) from the flue gases generated during the melting of the typical borosilicate glasses. Although these investigations do not fully meet the scope of this work, several oxyfuel melting systems were investigated that would be well suited for the use of a PtM system. The most promising system is the one with the data given in Table 1. A conventional soda-lime glass was assumed to be molten in this furnace in these studies, rather than borosilicate glass. As a result, boron compounds will be absent in the flue gas. The effects of H_3BO_3- and HBO_2-contaminated flue gases on currently established absorbents such as MEA have not been investigated so far and are therefore unpredictable.

Based on these assumptions, the flue gas properties and composition shown in Table 2 are used for the examination of the CO_2 absorption plant in this work. Additional thermodynamic properties for the flue gases were calculated using the open source software Cantera with GriMech 3.0 reaction mechanism [26, 27]. The properties of the clean gas lean on CO_2 (③) are resulting from the ASPEN Plus model, described in section 3.1. Trace substances were neglected in this modeling approach.

Table 2: Flue gas composition and properties for examination of the CO_2 absorption plant. Based on experimental results of [25]. Additional properties are calculated using [26, 27], and the developed ASPEN Plus model. STP = standard temperature and pressure conditions (0 ℃; 1.0135 bar). Numbers of gas compositions refer to Figure 2.

Properties	Unit	① Raw gas	② Clean gas rich on CO_2	③ Clean gas lean on CO_2
Temperature	℃	470	226	26
Volume flow rate				
- dry	m^3/h	4780	6910	4290
- wet	m^3/h	5910	8020	4437
Density at STP	kg/m^3	1.266	1.268	1.161
Spec. heat capacity (c_p)	J/(kg·K)	1219.2	1138.4	1022.5
Composition				
H_2O	Vol%	19.1	13.8	3.3
O_2	Vol%	19.8	20.5	24.8
CO_2	Vol%	11.3	7.0	0.8
N_2	Vol%	49.8	58.7	71.1
Trace Substances				
Dust	mg/m^3	1482	0.3	-
HCl	mg/m^3	2.6	0.2	-
HF	mg/m^3	72.7	0.5	-
SO_2	mg/m^3	19.0	8.1	-

The volume fractions of CO_2 in the flue gas are very low, while N_2 and O_2 are very high for an oxyfuel combustion system. As a result in this particular case, high amounts of ambient air are entering the flue gas system. The rise of O_2 fractions from raw gas to clean gas may also be explained due to ambient air leaks in the system.

2.7 CO_2 Separation

Technologies for CO_2 capture from combustion processes can be categorized as pre-combustion, post-combustion processes and processes that are directly connected to combustion (in this work further referred to as ”in-combustion” processes).

Pre-combustion processes primarily involve the separation of CO_2 from fuels such as lignite, hard coal or natural gas, in order to prevent CO_2 emissions prior to combustion. The coal is first converted into a CO_2 + H_2 gas mixture. In the case of natural gas, the main component methane is converted to CO_2 + H_2 by steam reforming. The CO_2 is subsequently separated by absorption processes such as rectisol, selexol or purisol. Such processes are widely established in the petrochemical industry, for example, to avoid acidification of natural gas and to be able to offer a high-purity product [28]. The remaining gas mixture, consisting mainly of H_2, can then be further used as fuel.

In-combustion processes such as chemical looping or so-called oxyfuel processes are directly involved in the combustion process. In the chemical looping process, metal oxides are added to the combustion chamber in fluidized bed reactors in order to effect a redox reaction, binding the CO_2 [29]. Oxyfuel processes are closely related to the combustion process described above. When natural gas is burned in pure oxygen, the exhaust gas theoretically consists only of CO_2 and water vapor. The H_2O can then be condensed by cooling and a highly pure CO_2 stream would remain [28]. Both processes are unsuitable for the glass industry. Metal oxides in the combustion chamber would react with the glass batch as well, thus negatively affecting the glass quality. The separation of CO_2 by cooling is unsuitable, since the exhaust gases in the glass industry also contain evaporation from the molten glass (Section 2.6). Accordingly, the CO_2 product stream would be contaminated after the condensation of H_2O.

Post-combustion processes concern the capture of CO_2 from combustion flue gases. Membrane processes as well as adsorption and absorption processes are discussed for this purpose. Membrane processes provide for the separation of CO_2 from the flue gas stream by means of molecular lattices. However, these membranes are currently not yet sufficiently temperature-stable and scalable for rapid implementation. Adsorption processes are currently mainly used for CO_2 capture from ambient air, known as direct air capture [30]. However, de- and adsorption cycles are still too slow for application in exhaust gas streams and are not available in sufficient capacity [28]. For CO_2 capture from exhaust gases, absorption processes are currently being discussed as the main option [28]. In particular, the use of monoethanolamine (MEA) as an absorbent is widely investigated and can be considered a standard. MEA suffers from degradation, caused by carbamate polymerization, as well as oxidative and thermal degradation. Therefore, a switch to more stable and efficient solvents is widely discussed [31, 32].

Nevertheless, CO_2 absorption processes can be considered the most promising technology for the glass industry, since they have been investigated for a long time and have already been tested in several worldwide projects for CO_2 separation from flue gases. They were also proposed for applications in other industry sectors, such as the cement industry [33].

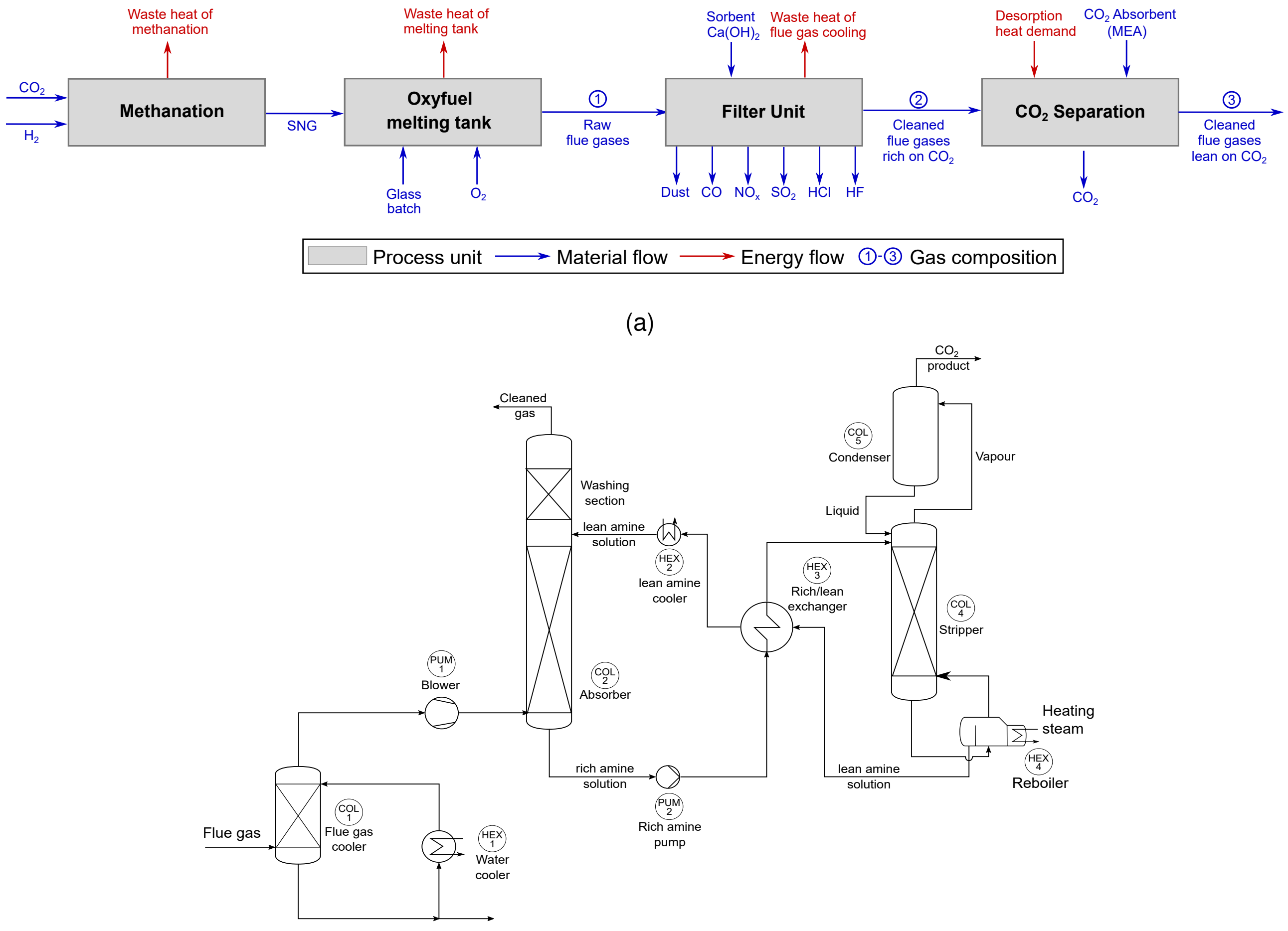
A integration of such a post-combustion CO_2 absorption plant is preferably realized after the already existing flue gas purification systems. At this point, the exhaust gases are already cleaned of harmful residues such as dust and a large proportion of the SO_2 emissions. Most of the remaining SO_2 is removed in the flue gas cooler in order to prevent excessive solvent degeneration. A flow sheet for this integration is shown in Figure 2.a).

2.7.1 Process Description of Amine based CO_2 Absorption

CO_2 separation from flue gases using absorption process can be divided into two main process steps. First, CO_2 reacts with the lean aqueous amine solution (30 wt.-% MEA in this case), in the absorber column. The rich solvent, containing chemically bounded CO_2 enters the stripper column by passing a lean/rich heat exchanger. This preheats the rich solution close to the stripper operating temperature of about 120 °C and consequently cools the lean solution. In the stripper column, the MEA solution is regenerated by an endothermic process. Low-pressure steam is supplied to the reboiler to maintain stable regeneration conditions as a result. To ensure an efficient desorption process, saturated steam at a temperature level of at least 140 °C, and a pressure of 3.6 bar is required. This thermal energy demand for desorption of CO_2 is considered to be the main cost factor during long-term operation of amine based CO_2 separation plants. In power plant technology, usually steam from intermediate stages of turbines is used for this purpose. This reduces electrical power output of the turbines and thus has a significant impact on increased electricity production cost of carbon capture power plants. However, the PtM concept introduced in this work, offers several options for waste heat utilization to meet the desorption heat demand in an energy efficient way.

2.7.2 Heat Supply for CO_2 Desorption

The ability to provide the necessary heat for desorption process is crucial for the economic viability of a post-combustion CO_2 absorption plant. The use of waste heat from these processes for steam generation is evident, especially for heat-intensive processes at common temperature levels of the glass industry. Figure 2. a) shows the most promising waste heat sources of the PtM process for the glass industry, namely i) methanation, ii) melting process and iii) flue gas filter unit. These options which will be described in more detail below.



(a)

(b)

Figure 2: a) Principal flow sheet of the integration of a CO_2 absorption plant in the glass industry.
b) Basic flow sheet of a CO_2 absorption plant with an amine-based solvent.

It should be mentioned that waste heat is also generated during PEM electrolysis. Depending on the performance and design of PEM electrolysis systems, either water or air cooling is used. The temperature level of this waste heat is at approx. 80 ℃ [14]. The utilization of this waste heat potential for steam generation is limited as a result. Further processing, for example by means of high-temperature heat pumps would be required to generate steam on the required temperature and pressure levels of CO_2 desorption. Due to these limitations, the waste heat option of PEM electrolysis is not considered in detail in this work.

Waste Heat of Thermochemical Methanation For thermochemical methanation processes, continuous cooling of reactors is required to prevent degeneration of established catalyst materials. The exothermic natue of the dominant reaction processes would otherwise lead to catalyst particle sintering. Commercially established tubular bulb reactors for methanation processes are usually operated at a temperature level of 300 ℃ and a pressure of 20 bar. Usually water cooling is a common way to maintain constant operating conditions in these reactors, generating steam at a temperature of approx. 260 ℃ and a pressure of 45 bar [34]. A Sankey diagram of tubular bulb reactors for methanation and their cooling process is shown in Figure 3.

Waste Heat of Melting Tank Due to the melting temperatures of up to 1600 ℃, and an efficiency of about 42 % of oxyfuel furnaces [6], a high waste heat potential occurs in the direct furnace environment. However, the use of this heat reservoir is limited by a number of restrictions. For example, the installation of heat

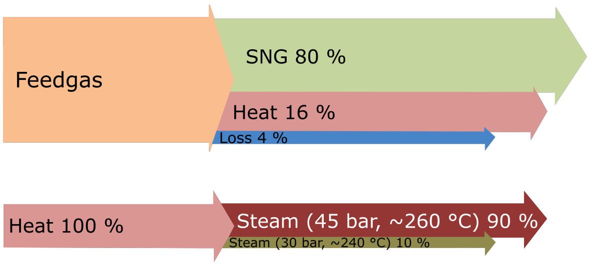


Figure 3: Sankey diagram of a tubular bundle methanation reactor [34].

exchangers close to the furnace wall is restricted, since permanent access for industrial maintenance and servicing is necessary. The remaining option is to use the heated ambient air close to the tank environment. However, the low temperature level of the ambient air (below 100 ℃) is an obstacle for the generation of process steam at the temperature and pressure level required for CO_2 desorption [35].

Waste Heat of Flue Gas Treatment In many cases, the waste gases from the furnaces are still at too high a temperature level to be directly passed on to a filter system. In particular, the textiles in cloth filter systems are sensitive to excessively high temperatures. Heat exchangers are therefore installed in many flue gas cleaning systems to cool flue gases to a suitable level for filter systems. The heat recovered in this way is used, for example, to supply heat to office and administration buildings at glass industry sites [36]. In some cases reheating of the waste gases is also necessary to ensure sufficient flow conditions for the removal of the waste gases through pipes and stacks [35, 37].

3 Materials and Methods

3.1 Simulation Approach

The technical and economic analysis of CO_2 capture from flue gases of the glass industry requires design and sizing of the plant components. For this work, the process simulation tool Aspen Plus was used for component design. Numerous other studies found this approach to be useful and validated [38, 39]. The most important equipment of the CO_2 capture plant, namely the absorber and stripper column (see Figure 2) were designed in detail, using the simulation results of the Aspen Plus model (v12).

The flowsheet of the Aspen Plus model is depicted in Figure 4. The columns are modeled in detail using a rate-based approach. The other components as the compressor, the pump or the heat exchanger are modeled in a simplified way. The ELECNRTL property method in Aspen is used to calculate media data using the eNRTL model for the liquid phase and the Redlich-Kwong equation of state for the vapour phase.

The dissociation reactions are considered by defining equilibria reactions and whereas the kinetics are considered for all reactions including CO_2 as a reactant using built-in power laws in Aspen Plus. Because of the fast reaction of the CO_2, the liquid film is discretized in five parts. As structured packings, the Melapak 250Y from Sulzer are used in all columns. The mass transfer coefficients, interfacial areas and column hold-up are calculated using the correlation of Bravo et al. [40, 41]. The heat transfer coefficients are determined according to the theory of Chilton and Coburn [42].

In order to calculate the column diameters the fractional approach to maximum capacity is used in Aspen Plus [43]. A factor of 0.65 is chosen. A constant capture rate of 90 % is set using the lean solvent mass flow. The heat flow into the reboiler in the stripper column is varied in such a way that the CO_2 mass balance is met. The lean solvent loading is varied to ensure an optimal operation point of the plant and minimum heat demand in the reboiler. The packed column height was set according to [38], and the relevant parameters of the plant are listed in Table 3.

In order to calculate the equipment costs, a comparison with an identical system is required (see section 3.2). A simulation model was developed for the CO_2 separation plant with the exhaust conditions ② shown in

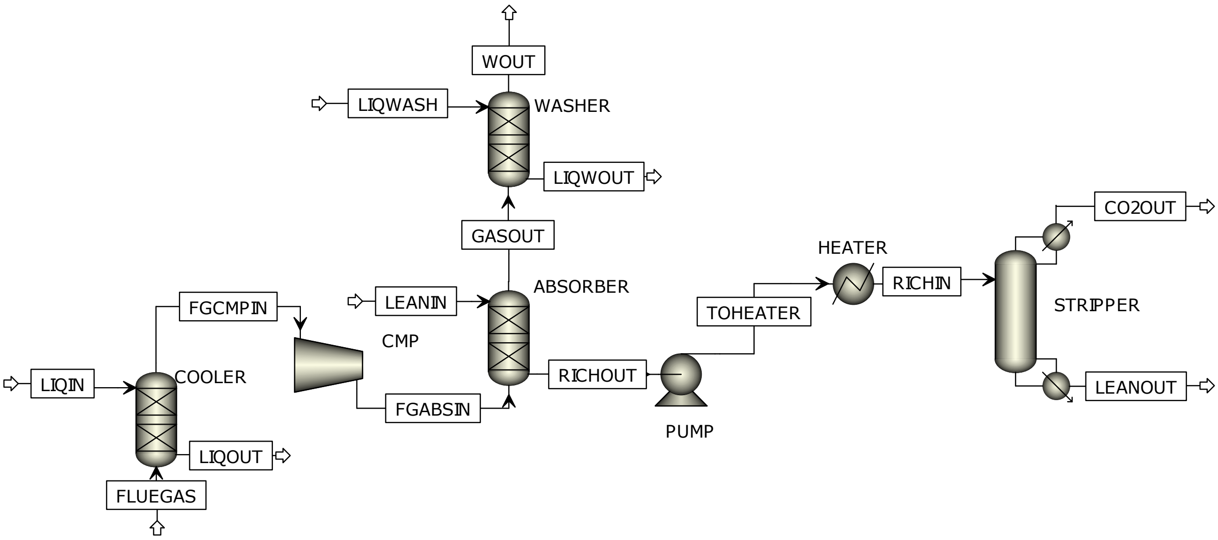


Figure 4: Main flow sheet overview of Aspen Plus Model used for the simulation

Table 3: Critical input design parameters of amine scrubbing plant

parameter	value
Solvent temperature at absorber inlet	35 ℃
MEA-concentration	30 w.-%
Desorber pressure	2 bar
Absorber flue gas outlet pressure	1 bar
CO_2 product temperature	25 ℃
Washing fluid temperature	25 ℃
Desorber solvent inlet temperature	105 ℃
carbon capture rate (set point)	90 %

Table 2, as well as a model for the reference CO_2 absorption plant described in [39, 44].

3.2 Economic Analysis

The costs of the plant components were calculated using the "estimating equipment cost by scaling" method, described in [45]. For this purpose, the costs of closely related reference plant equipment k_r is compared to equipment cost of the proposed plant k_e by an exponential factor m:

$$k_e = k_r \left(\frac{G}{G_r} \right)^m \tag{1}$$

The basis of characteristic component parameters of the reference equipment G_r such are for example column volume, pump mass flow rate or heat transfer area. These parameters are in relation to the one of the proposed plant G. Various degression exponents $m < 1$ for the respective components allow the cost comparison for each equipment type.

Other components of the investment costs, such as installation, instrumentation and control, piping or electricity supply, can subsequently be calculated as a proportion of total equipment costs, as proposed by [45] and already used for similar approaches for cost calculations of CO_2 absorption plants [44].

The well established equivalent annual cost (EAC) method was used to calculate costs of owning and operating assets over their entire life time.

The “ProcessNet - Chemical Plant Price Index for Germany” was used to take inflation-related price development since 2007 as well as increased material, distribution and logistics costs into account. According to this index, prices have risen by 21.5 % since 2008. Due to the impact of the global COVID-19 pandemic, the effects of production cutbacks and limited logistics capacities are particularly evident from 2020 onward. By 2019, the increase would have been only 16 % (see Figure 5) [46].

4 Results and Discussion

4.1 Simulation Results

For CO_2 absorption plants, the absorber and stripper column, as well as the gas blower and the cooling water pump for the pre-scrubber are the main cost factors with a share of more than 80 % [44]. The relevant comparative parameters for equation 1 were considered during simulations according to Table 6. Columns 4 and 6 in Table 6 show the simulation results for the CO_2 absorption plant considered in this work, as well as the re-evaluation of the reference plant, considered in [39, 44]. The most important results are shown in Table 4:

Table 4: Major simulation results for the design of the amine scrubbing plant

parameter	value
Absorber diameter	0.95 m
Desorber diameter	0.5 m
Scrubber diameter	0.95 m
Solvent flow rate	2.28 kg/s
Heat demand in stripper	556.5 kW
Specific heat demand in stripper	3.354 MJ/kg CO_2
CO_2-purity	99.3 %
CO_2 product mass flow	0.167 kg/s
Washing fluid temperature	25 °C
Desorber solvent inlet temperature	105 °C
carbon capture rate (Sollwert)	90 %

4.2 Cost Calculation Results

Cost calculations for the CO_2 absorption plant were determined considering the following steps: i) calculation of equipment cost (section 4.2.1), ii) calculation of capital expenditures (CAPEX, section 4.2.2)), iii) calculation of operational expenditures (OPEX, section 4.2.2) and iv) the calculation of resulting CO_2 avoidance cost (section 4.4). Basic assumptions for operating parameters of the CO_2 absorption plants are shown in Table 5.

4.2.1 Equipment Cost

Table 6 gives an overview of the equipment cost for both of the modeled CO_2 absorption plants. Details concerning the reference plant are given in [44]. The plant for this work was designed for the same CO_2 capture rate of 90 % and stripper operating conditions of 2 bar and 130 °C.

The most expensive equipment is the absorber column, which accounts for approx. 65 % of total equipment costs. The second most important contribution to equipment costs is related to the stripper column, responsible for another 20 %. As a result, 85 % of the total costs can be attributed to the most important columns in the process. The costs for cold water pump, blower and DC water cooler were also calculated in detail. These equipment parts contribute another 10 % of total costs. The remaining 10 % can be attributed to the lean/rich

Table 5: Operation parameter and economic assumptions for cost calculations of CO_2 absorption plant. All parameters are based on [44], except others are named. * Results of own simulations.

Parameter	Value
Specific desorption heat demand	928 kWh/t CO_2
Reboiler heat power	915 kW
Life time	20 years
Interest rate	5.0 % p.a.
Operating hours	8688 h/year
Maintenance cost	4.0 % of FCI
MEA price [44]	1000 €/t
MEA degradation rate	1.5 kg/t CO_2
Cooling water make up	1.0 m^3/ GJ thermal
Cooling water cost	0.20 €/m^3
Water cooler duty	640 kW
CO_2 product temperature	25 ℃
CO_2 product pressure	20 bar
Electricity demand for CO_2 compression [47]	72 kWh/t CO_2
CO_2 emissions before *	9601 t CO_2/year
CO_2 separation rate *	90 %
CO_2 avoided *	8641 t CO_2/year
CO_2 emissions after *	960 t CO_2/year

heat exchanger, amine pumps, reboiler, and other minor important equipment parts. The distribution of these expenses is in line with equipment cost calculations of [44].

The costs for CO_2 compression after separation is calculated following the approach given by [47] for CO_2 compression for carbon capture and storage (CCS) processes. Lower pressure levels are required for methanation than for underground storage of CO_2. [47] suggest a four stage compression process for a CO_2 product pressure of 120 bar. We assumed a one stage compression process to 20 bar is assumed. This pressure level is sufficient for established large scale methanation reactors like [49]. Consequently, equipment costs for CO_2 compression in the PtM concept are much lower than for CCS processes [44]

4.2.2 Capital Expenditures

Further direct and indirect costs for calculation total CAPEX of the CO_2 separation plant were estimated as a factor of the overall equipment cost, as suggested in [45]. Table 7 shows the parameters taken into account and their influence on the total CAPEX. Due to the comparably small size of the CO_2 capture plant, the lower limits of the ranges proposed in [45] were selected.

The OSBL costs for buildings, yard improvements, and service facilities are assumed to be lower than recommended by [45] due to low column volumes and compactness of the plant. As a result, a container solution for plant housing is a viable alternative. Considering this option, OSBL costs of over 100,000 € building infrastructure is justified.

The price index correction, according to Process Net [46], since 2007 has a significant impact of 21 % on total CAPEX. The extent to which prices develop according to current influences such as the COVID-19 pandemic and recent conflicts should be further examined. These effects have not yet been taken into account in the current values of [46].

CAPEX of plants for CO_2 capture from flue gases of power plants are far above the values calculated in this study. [44] and [50] propose € 147 million and € 179 million, respectively. By considering column

Table 6: Equipment cost calculation. Indicatiors (Ind.) refer to Figure 2.b for easier identification of equipment. *Remaining heat exchanger and pumps with less significant influence on total component costs.

Ind.	Equipment	Type	Exponent	Reference plant		Own plant	
			m	G_r	k_r in M€	G	k_e in €
COL 2	Absorber	Column	0.60 [48]	12560 m^3	10.94	39.25 m^3	340,000
COL 4	Stripper	Column	0.60 [48]	3391 m^3	3.43	8.75 m^3	110,000
–	Cold water pump	Pump, reciprocating	0.34 [45]	1.21 m^3/s	2.04	0.0015 m^3/s	21,000
PUM 1	Blower	Blower, centrifugal	0.59 [45]	485.44 m^3/s	3.10	2.236 m^3/s	13,000
COL 1	Flue gas cooler	Column	0.60 [45]	1570 m^3	0.54	4.75 m^3	17,000
–	Others*	mixed	-	-	3.89	-	20,000
				Total equipment cost:	23,94		521,000
	CO_2 compressors	Compressor, rotary, single stage	0.79 [45]	31.49 m^3/s	31.73	0.07761 m^3/s	69,000

geometry and main equipment dimensions, the plant presented here is about 400 times smaller. This factor is also outside the limit proposed by [45] for the applicability of the cost comparison method. The determined cost frameworks can be considered valid, by comparing specific CAPEX to other literature sources. [33] investigated CO_2 capture for carbon-intensive industrial processes and reported specific CAPEX of 160 €/t CO_2 and year for amine based post-combustion CO_2 capture in cement industry processes. The resulting specific CAPEX of this work are 188 €/t CO_2 and year. Considering the price increases since 2012 (approx. 16 % according to [46], see also Figure 5), the deviation of 15 %, calculated in this work seems justified. Different boundary conditions and assumptions of this work can influence the costs of the implementation of a CO_2 capture plant.

4.2.3 Operational Expenditures

The total operational expenditures (OPEX) include fixed charges for insurance and taxes, direct production costs, general expenses and plant overhead costs. The total annual OPEX of the novel CO_2 separation plant are shown in Table 8.

The yearly OPEX were calculated to be approx. 146,500 €. The main OPEX factors are operating labor costs, contributing about 27 % to total OPEX. We assumed a 0.1 job per shift for operation of the CO_2 separation plant. In the glass industry, a three-shift operation of 8 hours each for 365 days per year is common to maintain labor supply. The assumption that 10 % of the working time of a maintenance employee per shift is spent on maintenance and servicing of the CO_2 separation plant thus corresponds to a total time expenditure of approx. 2.5 h/day or 912.5 h/year. Others studies such as [44] assumed a two job per shift staff expenditure for plant operation. However, as the separation plant discussed in this work is much smaller, such a reduction in operating labor expenses seems justified. In addition, operating labor costs influence supervision and support labor costs, laboratory charges and plant overhead costs, which result in operating labor costs contributing to approximately 60 % of total OPEX.

MEA makeup and maintenance costs contribute about 18 % of total OPEX. Other works report this factors to have an influence of aprox. 32 % [44, 50], but consider a smaller plant size. Due to the comparatively small amounts of CO_2 involved, correspondingly less MEA makeup and maintenance is required.

We found OPEX to be 9 % of total CAPEX. In literature, a OPEX range of 7-12 % of total CAPEX for a post combustion MEA separation plant in the cement industry was considered [33]. The calculated OPEX is consistent with other literature sources, indicating potential for further optimization.

Table 7: Total capital investment (CAPEX) for MEA CO_2 separation plant.

	Range [45] %	Used %	Cost €
Direct cost			
Inside battery limit (ISBL) cost			
Total equipment cost (EC)	100		521,000
Installation	25-55	25	130,250
Instrumentation and control	8-50	8	41,680
Pipes	20-80	20	104,200
Electrical equipment	15-30	15	78,150
		Total ISBL:	875,250
Outside battery limit (OSBL) cost			
Building and building services	10-80	5	26,050
Yard improvements	10-20	5	26,050
Service facilities	30-80	10	52,100
		Total OSBL:	104,200
		Total direct cost:	875,250
Indirect cost			
Engineering	10	10	52,100
Construction expenses	10	10	52,100
Contractor's fee	0,5	0,5	2,605
Contingency	17	17	88,570
		Total indirect cost:	106,805
CO_2 compressor equipment			69,000
		Fixed capital investment (FCI):	1,155,285
Working investment	12-28	12	136,634
Start-up cost and MEA cost	8-10	8	92,423
Price index correction (2007-2022); % of FCI [46]		21,5	248.386
		Total capital investment (CAPEX):	1,634,278

4.3 Cost for Desorption Heat Generation

The supply of the necessary heat for desorption is crucial for the economic viability of a post-combustion CO_2 absorption plant (section 2.7.2). As shown in Table 5, the heat demand for the designed CO_2 separation process is 928 kW, at a steam temperature and pressure of 150℃ and 2 bar.

In particular, the methanation process as well as the flue gas filtration systems are the most promising options regarding the use of waste heat for steam generation.

4.3.1 Methanation Waste Heat Utilization

Considering the thermal power demand of the melting tank described in Table 1, a 7.60 MW methanation reactor is required to ensure a constant heat supply. This reactor can provide high pressure and temperature steam (HP-steam) and low pressure and temperature steam (LP-steam, see Figure 3). Table 9 shows the power distribution of a tubular bulb reactor, based on vendor information [34]. It is evident that the HP steam of the methanation reactor can already cover approx. 120 % of the heat demand for CO_2 desorption. According

Table 8: Total operating expenditures (OPEX) for MEA CO_2 separation plant.

	Range [45] %	Used %	Cost €/year
Fixed Charge			
Local taxes	1.0-4.0 % of FCI	1.0 %	11,533
Insurance	0,5-1,0 % of FCI	0.5 %	5,776
			17,329
Direct production cost			
Cooling water			4,003
MEA Makeup		1.5 kg/t CO_2	14,401
Maintenance (M)	1.0-10 % of FCI	1.0 %	11,553
Operating labor (OL)	0.1 job/shift	45 €/h	39,096
Supervision and support labor (S)	30 % of total labor cost	30 %	11,729
Operating supplies	15 % of maintenance	15 %	1,733
Laboratory charges	10-20 % of operating labor	10 %	3,910
			86,425
Plant overhead cost	50-70 % of M + OL + S	50 %	31,189
General expenses			
Administrative cost	15-20 % of OL	15%	5,864
Distribution and marketing	2-20 % of OPEX	2 %	2,816
Research and Development cost	2-20 % of OPEX	2 %	2,816
	Total operating expenditures (OPEX):		146,440

to vendor information, a specific CAPEX of 40 €/kW thermal can be expected for the cooling equipment of the methanation reactor. Based on the total steam power of 1.22 MW, a total CAPEX of approx. 49.000 € can be considered for cooling equipment of the methanation reactor in the considered PtM system of this work. Thus, assuming a lifetime of 20 years and an interest rate of 5 %, specific costs of 0.0004 €/kWh thermal are achieved.

Table 9: Waste heat potentials of methanation reactor equipment. HP = high pressure and temperature, LP = low pressure and temperature.

	Reference point	Power MW
Thermal power	heating value of SNG	6.08
HP-steam	45 bar, 260 ℃	1.09
LP-steam	30 bar, 240 ℃	0.12
Power loss	heat losses	0.30
Methanation reactor	combined	7.60

4.3.2 Flue Gas Treatment Waste Heat Utilization

In addition to methanation, waste heat recovery from flue gas cleaning for desorption heat demand is a promising option. However, considering the properties shown in Table 2, the enthalpy content in the investigated flue gas is about 850 kW for a ΔT of 247 ℃. This is not sufficient to provide the required thermal power of the reboiler of 915 kW.

The flue gas needs to be cooled to approx. 130 ℃ to ensure sufficient heat output for adequate steam generation. At this ΔT of 340 ℃ an enthalpy of 1171 kW is contained in the flue gases. This heat content is sufficient to cover the reboiler's heat demand as well as to compensate for heat losses through a flue gas heat exchanger.

Based on vendor information and calculations, a 5 bundle plain tube heat exchanger is sufficient to generate steam at the required desorption conditions. Depending on the exhaust gas properties, conventional steel or, in the case of high exhaust gas corrosivity, stainless steel would be suitable as a construction material for this heat exchanger. A cost estimate for these material options is around 40,000 € for conventional steel, and 90,000 € for stainless steel respectively [36]. Assuming a lifetime of 20 years and an interest rate of 5 %, specific costs of 0.0007 €/kWh thermal for common steel and 0,001 €/kWh thermal for stainless steel are achieved.

The lower outlet temperature of the flue gases from the filter system would also affect the CO_2 separation system. As a result, the capacity of the flue gas cooler could be reduced, which in turn would result in lower investment costs. This would also influence flow characteristics in the flue gas channel. Consequently, existing flue gas systems may no longer be able to achieve the required flow capacities and may require re-planning and design.

4.4 Cost of CO_2 Avoidance

With a lifetime of 20 years and an interest rate of 5 %, the annual depreciation or EAC costs of the CO_2 separation plant designed in this work are 131,175 €/year. As shown in Table 8, there are additional OPEX of 146,440 €/year. According to [47], a specific electrical power of 73 kWh/ t CO_2 is required to compress the captured CO_2 to a pressure of 20 bar. Specific electricity costs are taken from [6] where 0.12 €/kWh were assumed for glass industry companies. The three cost factors EAC of CAPEX, OPEX and electricity demand for CO_2 compression add up to an annual cost of 353,348 € for the CO_2 separation plant designed in this work.

The heat demand for desorption in year-round operation is 8017 MWh/year, or 928 kWh/t CO_2. The described CO_2 capture plant can avoid 8641 t CO_2/year (Table 5). The three options for waste heat utilization, described in section 4.3, were used to investigate their influence on CO_2 avoidance costs. The results are shown in Table 10. The CO_2 avoidance costs are 41,31 €/t CO_2 for waste heat utilization of the methanation process. For the flue gas heat exchanger option, slightly higher costs of 41,61 €/t CO_2, or 41,92 €/t CO_2 are achieved. These heat exchangers generate a lower heat output, and the use of stainless steel results in increased material costs. Despite the marginal increased cost, they illustrate the potential for cost-effective waste heat utilization for absorption based CO_2 capture in the glass industry.

Table 10: Calculation of CO_2 avoidance costs for different waste heat utilization options. EAC = equivalent annual costs of CAPEX.

	Specifc cost		Total cost
			€/year
EAC of CAPEX		189 €/(t CO_2 ·year)	131,175
OPEX			146,440
Electricity for CO_2 compression	0.12 €/kWh	8.60 €/t CO_2	82,521
		Fixed costs:	353,348
Heat supply cost options			
1. Methanation reactor	0.0004 €/kWh	0.37 €/t CO_2	3,264
		Total annual costs:	356,912
		CO_2 avoidance cost:	41,31 €/t CO_2
2. Flue gas heat exchanger conventional steel	0.0007 €/kWh	0.65 €/t CO_2	6,235
		Total annual costs:	359,584
		CO_2 avoidance cost:	41,61 €/t CO_2
3. Flue gas heat exchanger stainless steel	0.0010 €/kWh	0.93 €/t CO_2	8,908
		Total annual costs:	362,256
		CO_2 avoidance cost:	41,92 €/t CO_2

Other studies reported CO_2 separation costs from industrial waste gases to range from 25 to 135 €/t CO_2 [33, 51]. But these reported values depend very much on the used CO_2 capture technology. Comparable process designs in the cement industry with post-combustion CO2 capture using MEA assume short to mid-term CO_2 avoidance costs to be about 65 €/t CO_2 at low cost for external steam import [33]. In studies investigating CO_2 capture from industrial point sources for methanation, 50 €/t CO_2 were found [51].

4.5 Optimization Options

The CO_2 avoidance costs of 41-42 €/t CO_2 indicate the promising potential for low-cost and energy-efficient integration options for the post combustion CO_2 separation within the introduced PtM process for the glass industry. In addition, there are further options for optimizing the CO_2 capture process that have not yet been investigated:

In this work, a 30 wt.-% aqueous solution of MEA was assumed. Other studies have shown that increasing the MEA content in the aqueous solution up to 40 wt.-% can further reduce the CO_2 avoidance costs. This can be achieved by reducing the energy requirement for desorption and lowering investment cost for the plant equipment due to reduced liquid flow rates [44].

MEA is considered to be the standard solvent for post combustion CO_2 absorption. In addition, other absorbents such as secondary or tertiary amines, as well as various mixtures, are described to have improved performance compared to MEA. A mixture of the tertiary amine methyl-diethanolamine (MDEA) and piperazine (PZ) is described to have increased resistance to thermal and oxidative degradation, higher CO_2 loading capacity and a reduced heat demand of 2.6 GJ/t CO_2 vs. the 3.4 GJ/t CO_2 of MEA [52, 53].

In addition to solvent replacement and changing solution properties, an advanced process design could positively influence the costs of CO_2 separation. Improved designs for the rich/lean heat exchanger [54], vapour recompression and split-stream processes [55] have been investigated and showed promising potential for enhances CO_2 removal capacity or reduced energy demand.

Combinations of these improved process designs and careful solvent selection could further optimize costs and performance of absorption based CO_2 separation from flue gases of glass melting processes.

5 Conclusion

An innovative energy concept towards carbon free glass melting processes by effectively integrating a PtM process into an oxyfuel glas melting plant is introduced and discussed. The energy concept utilizes of the well-established infrastructure and know-how related to natural gas based oxyfuel glass melting plants. The integration of the PtM process seems to be a very promising option for the decarbonization of glass melting processes. The novelty of the main process components is described and recommendations for suitable technology options are provided in these studies.

The required capture of CO_2 from glass industry flue gases has been identified as the process with the highest research expenditure. Post-combustion CO_2 absorption capture processes, which have also been applied in power plant technology, were identified as the most promising option to meet this challenge. A process simulation of this technology allowed a detailed design of the most important plant components and their cost evaluation. The following conclusions can be drawn for CO_2 absorption from flue gases of the glass industry:

- Amine based CO_2 capture processes allow a reduction of 90 % of CO_2 emissions of combustion based glass melting processes. The designed CO_2 absorption plant for the glass industry is approx. 400 times smaller than comparable concepts for power plants, due to the much lower volumes of flue gas.
- This enables cost reductions in the required building infrastructure, as well as in staff costs for maintenance and monitoring. At the same time, cost-reducing scaling effects for large plants have a negative impact on the small plant described here.
- Various options for waste heat utilization are available within the introduced PtM process for oxyfuel glass melting, which offer cost-effective options for desorption heat supply. Especially the methanation process can be seen as the most suitable option, as it provides steam at sufficient temperature, pressure and volume, with no impact on existing glass industry infrastructure.
- The low operating and heat supply costs result in low CO_2 avoidance costs of 41 €/t CO_2. These could be further reduced by described optimization approaches, like changes in solvent loading, the use of advanced solvents and/or advanced CO_2 absorption processes.

These promising results for CO_2 capture also motivate further research on the entire PtM process presented. Initially, further simulative and techno-economic analyses should be carried out in order to assess the technical and economic parameters. Subsequently, both lab-scale plants and step-by-step integration into real glass melting processes can be rapidly implemented.

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Appendix

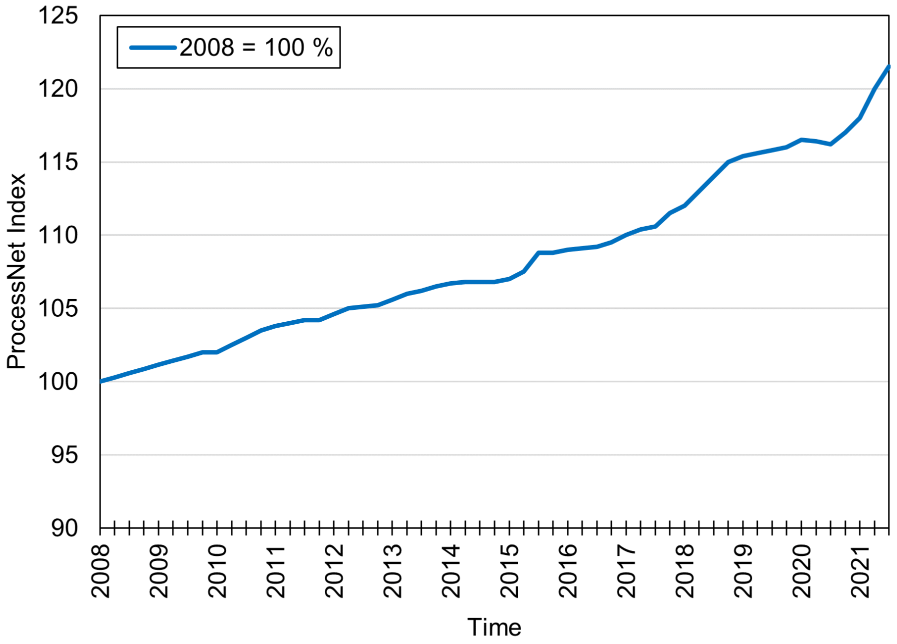

Figure 5: Quarterly development of the ProcessNet - Chemical Plant Price Index Germany since 2008