

Article

Techno-Economic Analysis of Carbon Dioxide Separation for an Innovative Energy Concept Towards Low-Emission Glass Melting

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Abstract: The currently still high fossil energy demand is forcing the glass industry to search for innovative approaches for the reduction in CO₂ emissions and the integration of renewable energy sources. In this paper, a novel power-to-methane concept is presented and discussed for this purpose. A special focus is on methods for the required CO₂ capture from typical flue gases in the glass industry, which have hardly been explored to date. To close this research gap, process simulation models are developed to investigate post-combustion CO₂ capture by absorption processes, followed by a techno-economic evaluation. Due to reduced flue gas volume, the designed CO₂ capture plant is found to be much smaller (40 m³ absorber column volume) than absorption-based CO₂ separation processes for power plants (12,560 m³ absorber column volume). As there are many options for waste heat utilization in the glass industry, the waste heat required for CO₂ desorption can be generated in a particularly efficient and cost-effective way. The resulting CO₂ separation costs range between 41 and 42 EUR/t CO₂, depending on waste heat utilization for desorption. These costs are below the values of 50–65 EUR/t CO₂ for comparable industrial applications. Despite these promising economic results, there are still some technical restrictions in terms of solvent degradation due to the high oxygen content in flue gas compositions. The results of this study point towards parametric studies for approaching these issues, such as the use of secondary and tertiary amines as solvents, or the optimization of operating conditions such as stripper pressure for further cost reductions potential.

Keywords: power-to-gas; methanation; oxyfuel; glass industry; CO₂-separation; economic evaluation



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1. Introduction

Commercial glass production is a very energy-intensive industrial process. Converting raw materials, such as silica sand, sodium carbonate, lime, dolomite, etc., 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 demand 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 the dominance of fossil fuels, glass production is currently still associated with high carbon dioxide (CO₂) emissions. The container and flat glass industry can be considered the dominating glass-producing sector and currently emit over 60 million tons of CO₂-emissions a year. This is more than the annual emissions of Portugal. The glass

industry in the EU, which is the world's largest producer [2], emits more than 20 million tons of CO₂-emissions a year, with an energy demand of more than 350 PJ. Approximately 75% of these emissions can be located in combustion processes, while the remaining 25% are contributed by the dissociation of carbonate raw materials [3]. The level of glass manufacturing-related CO₂ emissions in Germany has stagnated at a constant level of approximately 4 mio. t CO₂-eq. per year since 2007 [4]. The CO₂ emission budget from 2020 to 2050 of the entire German glass industry is 17.7 Mt CO₂-eq. for a strict 1.5 °C climate target and 112.0 Mt CO₂-eq. for a 2 °C climate target [5]. Due to the current high CO₂ emissions of the global glass production, as well as these severely limited remaining emission budgets, fast and effective measures to reduce emissions in the glass industry are of vital importance.

Of course, there are efforts towards greater sustainability and climate protection in the glass industry. Accordingly, there are many research approaches and developments for the 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 (H₂) as combustion fuel instead of NG [6]. However, all of these concepts, despite their promising CO₂ emissions reduction potential, have certain restrictions:

All-electric melting is well established for small-scale glass melting systems, but large-scale applications are still controversial [7]. Recent all-electric melting projects have made some progress and could reach melting capacities of up to 250 t/d. However, the 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 [8]. In addition, a 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 [6].

Recent projects, such as HyGlass [9], HyNet [10], and Kopernikus P2X [11], have investigated hydrogen as a fuel substitution option. Since the combustion of H₂ does not cause any CO₂ emissions, this would significantly reduce the CO₂ footprint of glass melting. However, the total CO₂ emissions of the glass industry are not only caused by fossil fuel combustion but also by carbonate reactions during batch to melt conversion. As a switch to H₂-combustion will not influence the batch composition of glass manufacturing, further CO₂ emissions will remain. The CO₂-emission reduction of H₂-combustion-based melting systems will significantly depend on the means of hydrogen production (green, blue, grey, ... H₂). To date, only H₂ production based on renewable energies can guarantee a large reduction in CO₂ emissions (green H₂). Current scientific debates mainly focus on the climate impact of blue hydrogen. Ref. [12] states that blue hydrogen comes with only 9–12% less CO₂ emissions compared to conventional (i.e., grey) hydrogen. However, according to [13], the climate impact of blue hydrogen significantly depends on the key parameters such as methane emissions during the natural gas supply chain, CO₂ capture rate during production, and the applied global warming metrics. Thus, the authors concluded that blue hydrogen can be competitive to green hydrogen production in terms of climate impacts, if state-of-the-art process technologies and metrics are applied [13].

However, both green and blue H₂ are still very cost-intensive and are therefore not fully economically competitive with established fossil fuels. In addition, H₂ shows a very different combustion behavior, including higher flame temperatures, different flame velocity, faster ignition behavior, and changed heat radiation properties [8]. Some of these effects are compensated by oxyfuel combustion, for example, changes in adiabatic flame temperature, as shown in [8]. Nevertheless, there are still some uncertainties, such as the effects of higher water content in the flue gases in the firing chamber on glass quality, and the melting tank life time.

Scope of This Work

Due the limitations of currently discussed decarbonization options, there is an urgent need for innovative energy concepts which allow the integration of renewable energies, a

reduction in CO₂ emissions, and which ideally have no impact on the 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 [14,15], there are a lot of established technical options for most of the process steps. The state of the art of these established options is briefly described, before the remaining open process of CO₂ capture from exhaust gases is discussed in detail. Since CO₂ capture from exhaust gases in the glass industry has not yet been considered in this particular application, this process shows the most extensive research and development demand. Therefore, a technology concept for the integration of amine-based CO₂ capture processes into the already existing flue gas treatment systems of the glass industry is presented, and various possibilities of waste heat utilization are investigated. The main focus of this work was the techno-economic analysis of the CO₂ separation concept, in order to provide a basis for the economic analyses of the entire PtM system in future work.

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.

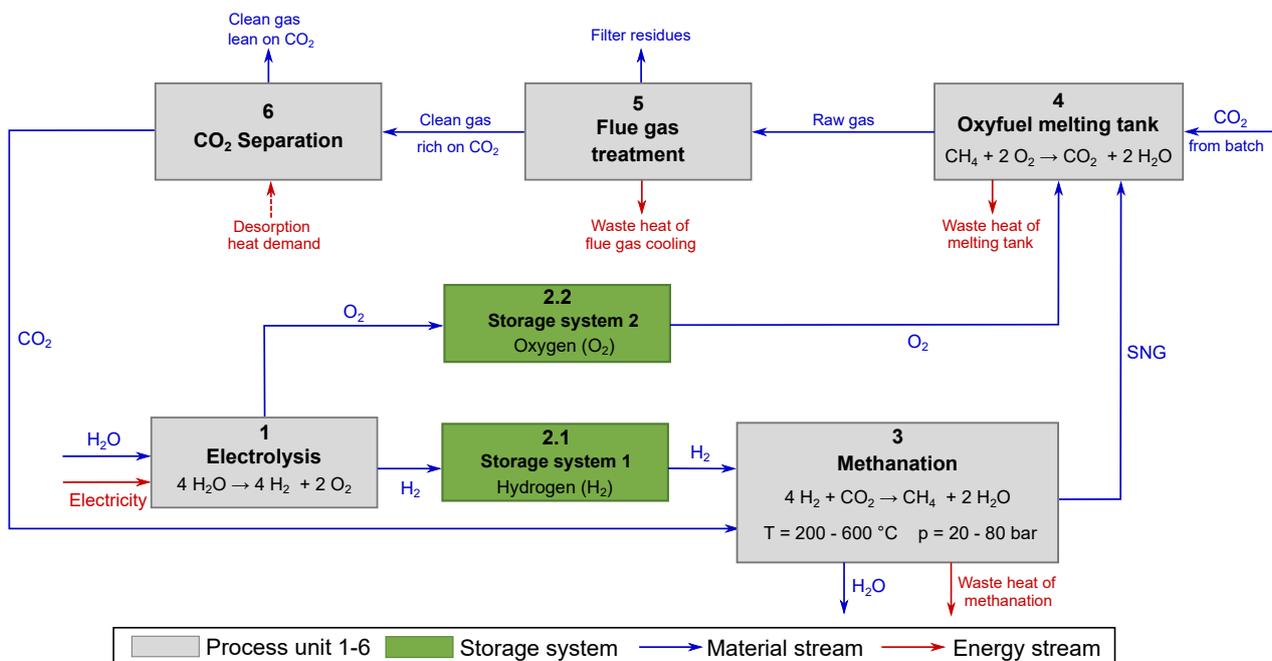


Figure 1. Simplified flow sheet of the integration of power-to-methane (PtM) into oxyfuel glass melting processes.

In the first process unit (Figure 1, 1), water (H₂O) is separated into (H₂) and oxygen (O₂) by electrolysis. Both H₂ and O₂ are subsequently stored in compressed gas storage systems (Figure 1, 2.1 and 2.2, details in Section 2.1.2). Besides H₂, O₂ is also an important product for downstream PtM processes, as it can be used in oxyfuel combustion (see Section 2.1.4).

The intermediate storage of H₂ is necessary due to the technological restrictions on the flexibility of the downstream catalytic methanation process (Figure 1, 3, details on which can be found in Section 2.1.3). In methanation, H₂ reacts with CO₂ in a pressure range of 20–80 bar and a temperature range of 200–600 °C in a catalytic process to form CH₄ and steam. The exact thermodynamic characteristics of methanation reactors may vary for each specific reactor design. As CH₄ is the main component of fossil natural gas compositions, the methanation product stream is also referred to as synthetic natural gas (SNG).

SNG is subsequently used in the oxyfuel glass melting furnace (Figure 1, 4) with O₂ from electrolysis. Thus, the technique of O₂ production in glass manufacturing processes via energy intensive air separation units established to date can be avoided, by utilizing combined effects in the overall process. In oxyfuel glass melting, CO₂ is mainly emitted from combustion (approximately 90%), but also from carbonate reactions during the batch-to-melt conversion (approximately 10%, depending on batch composition) [16]. CO₂ is subsequently emitted from the melting tank by the exhaust gases.

In the flue gas treatment process (Figure 1, 5), pollutants such as NO_x and SO_x are removed due to regulatory air pollution control laws. Such flue gas cleaning processes were introduced in the 1990s and aim to improve air quality, protect the health of residents, and reduce environmental pollution [6]. The used technologies are scrubbers, electrostatic, and cloth filters, as well as absorption-based processes [17]. Since the separation of pollutants such as NO_x and SO_x is important for air pollution control, but not for the described integration concept for PtM processes, the separation technology and its costs will not be discussed in detail.

However, these processes do not focus on CO₂-separation from flue gases. Therefore, an additional separation process is included in the concept to remove CO₂ from cleaned flue gases Figure 1, 6. The separated and purified CO₂ is subsequently used as a product stream for the methanation process. Holistically, this concept for integrating PtM into glass melting processes creates an almost closed carbon cycle (depending on CO₂ capture rate), which enables a significant reduction in greenhouse gas emissions. Additionally, the established oxyfuel glass melting processes, associated with highly stable glass quality and output, can still be used. Moreover, a switch to a renewable electricity supply from fluctuating sources such as wind and solar power can be achieved, without losing proven know-how and associated process stability.

However, the multiple process steps such as electrolysis, storage, and methanation lead to losses in the overall efficiency of the system. Thus, access to low-cost energy from renewable sources, as well as high costs for natural gas or CO₂ emission certificates, are crucial for the economic viability of the overall concept. In addition, only a renewable energy supply for electrolysis will lead to a reduction in total CO₂ emissions (see [8]).

There are adequate technical options with a significant degree of technological readiness for the most important PtM process steps electrolysis, H₂ and O₂ storage, and methanation (see Section 2.1). However, CO₂ capture from the flue gases of the glass industry has not been investigated in detail yet. To close this research gap, this work discusses the technical background and options for CO₂ separation processes in the glass industry in detail (see Section 2.2, provides a simulation-based design approach for such plants, (Section 4), and investigates the techno-economic evaluation of CO₂ separation from the flue gases of the glass industry (Section 5.2). This techno-economic analysis focuses explicitly on CO₂ capture, not on the overall concept. The economic evaluation of the overall system that includes electrolysis, storage concepts, and methanation will be investigated in subsequent work.

2.1. State-of-the-Art of Process Components

2.1.1. Electrolysis

Alkaline (AEL), solid oxide (SOEC), and polymer electrolyte membrane electrolysis (PEMEL) can currently be considered the most technically established processes for this purpose [14,18]. AEL has the highest technology readiness level (TRL) 9, but only offers limited part-load capability (30–100%) due to thermal restrictions [18]. SOEC would offer the highest efficiency of all options (up to 95% [19]), but requires an operating temperature level of 600–1000 °C [18]. This temperatures can be provided in an energy-efficient way by integrating the waste heat from catalytic methanation. However, the 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 such as wind power and photovoltaic. While the TRL of AEL is considered to be mature, the TRL of PEM is slightly behind [20]. Nevertheless, PEM electrolyzers are already available in the power categories of multiple MW (e.g., Siemens Silyzer 300 [21]) and the technology can therefore be considered to be of commercial technology status [20]. Development potential is primarily focused on the industrial series production of this electrolysis technology, which is expected to result in a significant reduction in costs [14].

2.1.2. Gas Storage

In general, there are various options for H₂ storage:

- Physical storage technologies such as compressed gas, liquid, or cryo-compressed H₂ storage [22].
- Adsorption technologies, relying on carbon-based materials such as multi-walled carbon nanotubes (MWCNTs), metal–organic frameworks (MOFs), or zeolites [22].
- Adsorption technologies, based on metal hydrates, like iron-oxide pellets [23].
- Chemical H₂ storage including liquid organic hydrogen carriers (LOHCs) [24],

For the process described in this work, H₂ 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 [22]. Nevertheless, the application of innovative H₂ storage technologies within the described PtM process should be focused on in further research. The especially high waste heat potential in glass melting processes offers attractive options for an energy-efficient use of LOHC or iron-oxide based H₂ storage technologies.

Storage system 2 in Figure 1 is necessary to ensure a constant flow of O₂ for a stable combustion process in the melting tank. Various options for O₂ storage have existed at the industrial scale for a long time and can therefore be considered robust [25]. To date, the O₂ required for oxyfuel combustion in the glass industry has primarily been produced by air separation using vacuum pressure swing adsorption (VPSA) processes [25]. This is performed either directly at the industrial site, or by delivery and storage in compressed gas tanks (liquid O₂). In both cases, oxygen storage facilities are usually available on site.

2.1.3. Methanation

Currently, thermocatalytic and biological methanation are the main technologies being discussed for use in power-to-gas systems [18,26]. Due to its higher TRL and technical performance, catalytic methanation is mainly used in large-scale PtM plants [14].

However, this methanation process requires constant operating conditions to achieve a high CH₄ content in the synthetic natural gas (SNG). For this purpose, the feed gases CO₂ and H₂ 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 the thermodynamic optimum. For many commercially available fixed-bed reactors, an operating temperature of 300 °C and a pressure of 20 bar have proven to be suitable operating conditions. Nevertheless, the operating temperatures of established reactor concepts vary between 200 and 600 °C and a pressures range of 20–80 bar [18,26]. In any case, the effective cooling of the reactor is required. In the PtM system described above, this waste heat could be used for SOEC or CO₂-desorption (as can be seen in Section 2.2).

In addition to the technical properties mentioned above, the purity of CO₂ is crucial for a stable methanation process. In order to investigate the effects of CO₂ impurities on methanation, experiments investigating the direct methanation of flue gases for different types of power plants were 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, the 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 [27,28].

2.1.4. Oxyfuel Glass Melting Tank

In oxyfuel combustion processes, a fuel such as natural gas is burned in a pure oxygen atmosphere instead of ambient air. Oxyfuel combustion is more energy-efficient because the 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 [29]. 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. Nevertheless, oxyfuel furnaces are established state of the art, 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 [8].

2.1.5. Flue Gas Purification Systems

Currently, mainly fabric filters and electrostatic precipitators are established 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 hydrochloride 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, $\text{Ca}(\text{OH})_2$) 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. Fabric filters in particular are much more sensitive to high gas temperatures than electrostatic filters.

2.2. 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 natural gas or pulverized coal, in order to prevent CO_2 emissions prior to combustion. These are mainly absorption processes with solvents such as rectisol, selexol, or purisol. Such processes are widely established in the petrochemical industry, for example, to avoid the acidification of natural gas and to be able to offer a high-purity product [30].

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 [31]. 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 [30]. However, both processes are unsuitable for the glass industry. Metal oxides in the combustion chamber would also react with the glass batch, 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 3). Accordingly, the CO₂ product stream would be contaminated after the condensation of H₂O.

Post-combustion processes concern the capture of CO₂ from combustion flue gases. Membrane processes as well as the adsorption and absorption processes are discussed for this purpose. Membrane processes provide for the separation of CO₂ 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 CO₂ capture from ambient air, known as direct air capture [32]. However, de- and adsorption cycles are still too slow for application in exhaust gas streams and are not available in sufficient capacity [30]. For CO₂ capture from exhaust gases, absorption processes are currently being discussed as the main option [30]. In particular, the use of monoethanolamine (MEA) as an absorbent is widely investigated and can be considered a standard. However, MEA can suffer 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 [33,34].

Nevertheless, CO₂ 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 the separation of CO₂ from flue gases. They were also proposed for applications in other industry sectors, such as the cement industry [35].

The integration of such a post-combustion CO₂ 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 SO₂ emissions. A flowsheet for this integration is shown in Figure 2a).

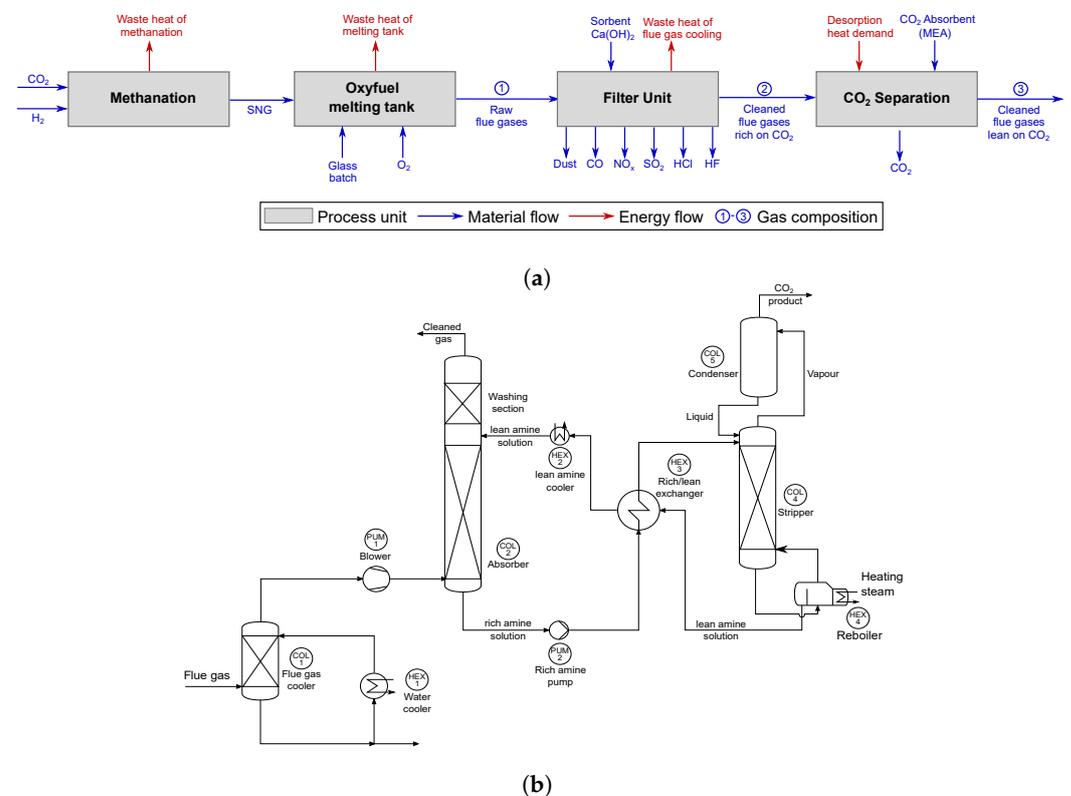


Figure 2. (a) Flowsheet for the conceptual integration of a CO₂ separation processes in glass melting systems. (b) Principal flow sheet of the components of a CO₂ absorption plant with an amine-based solvent.

CO₂ separation from flue gases using absorption process can be divided into two main process steps. First, CO₂ 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 approximately $120\text{ }^\circ\text{C}$ and consequently cools the lean solution. In the stripper column, the MEA solution is regenerated by an endothermic process. Therefore, low-pressure steam is supplied to the reboiler to maintain stable regeneration conditions. A temperature level of at least $150\text{ }^\circ\text{C}$ and a pressure of 2 bar are required to ensure an efficient desorption process.

This thermal energy demand for the desorption of CO_2 is considered to be the main cost factor during the 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 the electrical power output of the turbines and thus has a significant impact on the 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.

2.3. 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. Especially for heat-intensive processes at common temperature levels of the glass industry, the use of waste heat from these processes for steam generation is evident. Figure 2a shows the most promising waste heat sources of the PtM process for the glass industry, namely (i) methanation, (ii) melting process, and (iii) the flue gas filter unit. These options will be described in greater detail below.

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 approximately $80\text{ }^\circ\text{C}$ [19]. Therefore, the utilization of this waste heat potential for steam generation is limited. 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.

2.3.1. Waste Heat of Thermochemical Methanation

For thermochemical methanation processes, the continuous cooling of reactors is required to prevent the degeneration of established catalyst materials. Otherwise, the exothermic nature of the dominant reaction processes would lead to catalyst particle sintering. Commercially established tubular bulb reactors for methanation processes are usually operated at a temperature level of $300\text{ }^\circ\text{C}$ 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 approximately $260\text{ }^\circ\text{C}$ and a pressure of 45 bar [36]. A Sankey diagram of tubular bulb reactors for methanation and their cooling process is shown in Figure 3.

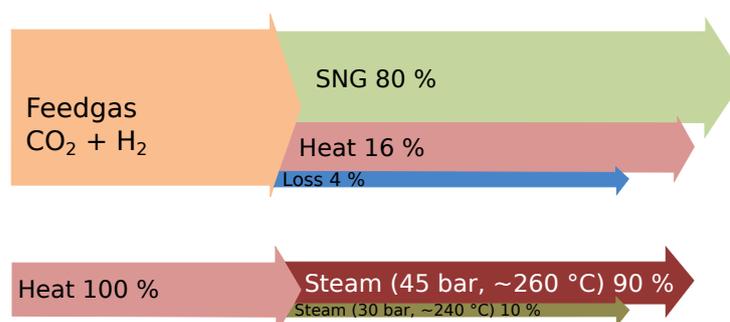


Figure 3. Sankey diagram of a tubular bundle methanation reactor [36].

2.3.2. Waste Heat of Melting Tank

Due to the melting temperatures of up to 1600 °C, as well as an efficiency of approximately 42% of oxyfuel furnaces [8], 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 exchangers close to the furnace wall is restricted, since permanent access for industrial maintenance and servicing must be ensured. 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 °C) is an obstacle for the generation of process steam at the temperature and pressure level required for CO₂ desorption [17].

2.3.3. 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 the cloth filter systems are sensitive to excessively high temperatures. Therefore, heat exchangers are 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 [37]. In some cases, however, the reheating of the waste gases is also necessary to ensure sufficient flow conditions for the removal of waste gases through pipes and stacks [17,38].

3. Flue Gas Properties and Composition

Detailed data on the flue gas composition and properties must be available to enable an evaluation of the CO₂ 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, a literature screening was conducted for data from the flue gas analyses of melting systems, suitable for the application of the PtM process presented in this paper. The exhaust gas study by Roger et al. [39] 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 the removal of boron compounds (namely boric acid (H₃BO₃) and meta boric acid (HBO₂)) 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. For the studies in this work, a conventional soda-lime glass, was assumed to be molten in this furnace. Compared to borosilicate glass, soda-lime glass changes the chemical composition of the glass batch while maintaining the technical design of the melting tank. Melting a soda-lime glass further results in the absence of boron compounds in the flue gas. The effects of boron compounds, such as H₃BO₃) and HBO₂, on currently established absorbents such as MEA have not been investigated to date and are therefore unpredictable.

Table 1. Key figures of the melting system investigated in [39].

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

The flue gas properties and composition shown in Table 2 were used for the examination of the CO₂ 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 [40,41].

Table 2. Flue gas composition and properties for the examination of the CO₂ absorption plant. Based on the experimental results of [39], additional properties are calculated using [40,41]. STP = standard temperature and pressure conditions (0 °C; 1.0135 bar). Numbers of gas compositions refer to Figure 2. ① raw gas before flue gas treatment. ② cleaned gas after flue gas treatment unit, entering the CO₂ separation process. ③ clean gas composition after CO₂ separation.

Properties	Unit	① Raw Gas	② Clean Gas Rich on CO ₂	③ Clean Gas Lean on CO ₂
Temperature	°C	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 (<i>c_p</i>)	J/(kg·K)	1219.2	1138.4	1022.5
Composition				
H ₂ O	Vol%	19.1	13.8	2.3
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	-

It should be noted that the volume fractions of CO₂ in the flue gas are very low, while N₂ and O₂ are very high for an oxyfuel combustion system. It can be concluded that in this particular case, high amounts of ambient air are entering the flue gas system. The rise of O₂ fractions from raw gas to clean gas may also be explained due to ambient air leaks in the system. It should be noted, that such flue gas compositions are based on experimental results from existing glass melting plants, with no further explanations for the presumed high ambient air content provided by the study [39]. However, for the sake of the most energy-efficient overall process, this high ambient air content should be avoided, as it can result in a higher CO₂ concentration in the flue gas. This would have a positive effect on the specific heat requirement (MJ/kg CO₂) of the CO₂ separation.

4. Materials and Methods

4.1. Simulation Approach

The technical and economic analysis of CO₂ capture from the flue gases of the glass industry requires the 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 [42,43]. The most important equipment of the CO₂ capture plant, namely the absorber and stripper column (as can be seen in 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, namely the compressor, the pump, or the heat exchanger were modeled in a simplified way. The ELECNRTL property method in Aspen is used to calculate the media data using the eNRTL model for the liquid phase and the Redlich–Kwong equation of state for the vapor phase.

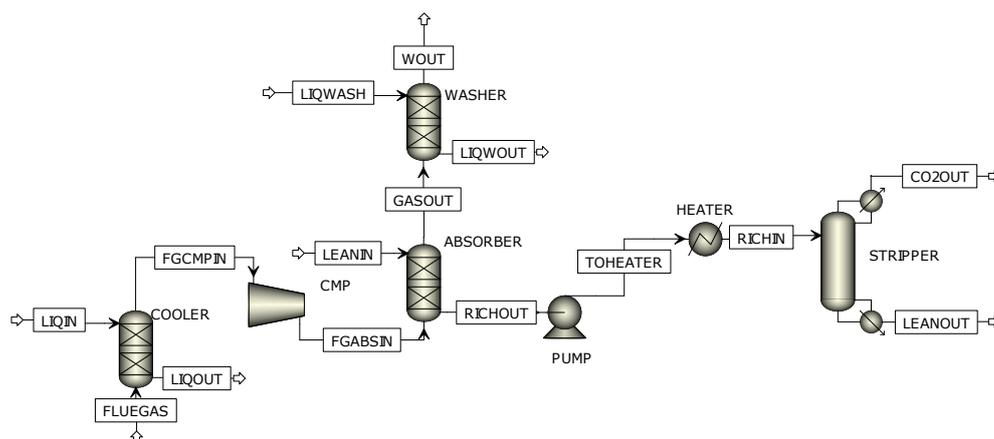


Figure 4. Main flow sheet overview of Aspen Plus Model used for the simulation.

The dissociation reactions are considered by defining the equilibrium reactions, 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 [44] are used in all columns. The mass transfer coefficients, interfacial areas, and column hold-up are calculated using the correlation of Bravo et al. [45,46]. The heat transfer coefficients were determined according to the theory of Chilton and Coburn [47].

In order to ensure the validity of the mass balance, it would be an obvious step to close the loop in the Aspen model. However, this significantly decreases the numerical stability of the model. Thus, the mass balance is controlled using the built-in optimizer in Aspen Plus. The CO_2 mass balance is controlled by the heat flow rate to the stripper. All other minor components such as oxygen or nitrogen are varied in such a way that the mass balance is fulfilled. The carbon capture rate is controlled by using the solvent flow rate. To ensure that the plant is operated at the optimal operation point, the lean CO_2 loading (CO_2 loading at absorber entry) is varied by the optimization function.

In order to calculate the column diameters, the fractional approach to maximum capacity is used in Aspen Plus [48]. 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 [42], and the relevant parameters of the plant are listed in Table 3.

Table 3. Assumed design parameters of amine scrubbing plant.

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

In order to calculate the equipment costs, a comparison with an identical system is required (as can be seen in Section 4.3). Therefore, a simulation model was developed for the CO_2 separation plant with the exhaust conditions ② shown in Table 2 and another was developed for the reference CO_2 absorption plant described in [43,49]. In addition, the operating parameters shown in Table 4 are assumed for the CO_2 separation plant. Due to

the continuously operated glass melting tank, operating hours of 8688 h/year are assumed for the CO₂ separation plant. These high operating hours are necessary to guarantee a sufficient separation rate of 90% CO₂. This assumption for operating time is higher than the reported 7500 operating hours from [49]. However, operating hours of more than 8500 h/year are a common assumption in other studies, investigating CO₂ separation from industrial processes [50].

Table 4. Operation parameter of the designed CO₂ absorption plant. All parameters are based on [49], unless indicated otherwise. * Own simulations.

Parameter	Value
Specific desorption heat demand	928 kWh/t CO ₂
Reboiler heat power	915 kW
Life time	20 years
Operating hours	8688 h/year
MEA degradation rate	1.5 kg/t CO ₂
Cooling water make up	1.0 m ³ / GJ thermal
Water cooler duty	640 kW
CO ₂ product temperature	26 °C
CO ₂ product pressure	2 bar
CO ₂ emissions before [8]	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

4.2. CO₂ Compression

After the CO₂ separation unit (Figure 1, 6), the separated CO₂ is available at a pressure of approximately 2 bar and a temperature of approximately 26 °C. Thus, to reach the operating pressure of the methanation reactor of approximately 20 bar, CO₂ compression is required. The required pressure of the methanation reactor is much lower than the usual pressures for the underground storage of CO₂ in established carbon capture and storage concepts of up to 120 bar [51]. To date, to achieve this high compression from 2 to 120 bar, four-stage compression processes have been suggested for CCS concepts [51], which might no longer be required for the reduced compression ratio from 2 to 20 bar for the PtM concept. Due to the significant deviations in the compression processes of the PtM concept described in Figure 1 and the described process in [43], a detailed evaluation of the compression energy demand and investment cost is conducted. In [51], the required power demand for CO₂ compression is calculated as:

$$P_{el} = \alpha_{el} \cdot \ln\left(\frac{P_{out}}{P_{in}}\right) \cdot \dot{m}_{CO_2} \quad (1)$$

where P_{el} is the electrical power demand in kW, α_{el} is the compressor power constant of 87.85 kW/kg CO₂, P_{in} and P_{out} are the inlet and outlet pressure in Pa, respectively, and \dot{m}_{CO_2} is the inlet CO₂ mass flow in kg/s. Moreover, the calculation of investment costs in a CO₂ compressor is suggested as [51]:

$$I = \left(\alpha_1 \cdot \dot{m}_{CO_2}^{\alpha_2} + \alpha_3 \cdot \ln\left(\frac{P_{out}}{P_{in}}\right) \cdot \dot{m}_{CO_2}^{\alpha_4} \right) \cdot \dot{m}_{CO_2} \quad (2)$$

where I is the investment cost in EUR and \dot{m}_{CO_2} is the CO₂ mass flow in kg/s. α_n are empiric constants, determined as $\alpha_1 = 0.1 \cdot 10^6$ in EUR/(kg/s), $\alpha_2 = -0.71$, $\alpha_3 = 1.1 \cdot 10^6$ in EUR/(kg/s) and $\alpha_4 = -0.60$ [51]. The results of the calculations obtained from Equations (1) and (2) were validated against vendor information [52].

4.3. Economic Analysis

The costs of the plant components were calculated using the “estimating equipment cost by scaling” method, described in [53]. For this purpose, the costs of closely related reference plant equipment k_r is compared to the equipment cost of the proposed plant k_e by an exponential factor m :

$$k_e = k_r \left(\frac{G}{G_r} \right)^m \quad (3)$$

The basis of the characteristic component parameters of the reference equipment G_r include, for example, the column volume, pump mass flow rate, or heat transfer area. These parameters are in relation to that 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 be subsequently calculated as a proportion of total equipment costs, such as that proposed by [53] and already used for similar approaches for cost calculations of CO₂ absorption plants [49]. The well-established equivalent annual cost (EAC) method was used to calculate the costs of owning and operating assets over their entire life time. Further assumptions for cost parameters are shown in Table 5.

Table 5. Assumptions for cost calculations of CO₂ absorption plant. All parameters are based on [49], unless indicated otherwise.

Parameter	Value
Interest rate	5.0% p.a.
Maintenance cost	4.0% of FCI
MEA price	1000 EUR/t
Cooling water cost	0.20 EUR/m ³
Operating labor cost, based on [54,55]	45.00 EUR/h
Electricity costs [8]	0.12 EUR/kWh

According to [54], hourly gross salaries in Germany for operating labor in maintenance and repair are between 15.89 and 30.96 EUR/h, depending on the company, industry sector, and location. Ref. [55] stated a gross annual salary of EUR 47,756 for maintenance and repair operating labor. Given the standard 40 h work week in Germany, this results in a gross hourly wage of 22.26 EUR/h. In Germany, however, companies have to pay additional insurance, taxes, and levies (ITL) on top of the gross wages of employees. To account for these costs, an additional ITL-factor of 1.7 can be considered on the gross salary. The 45 EUR/h, named in Table 5 thus results in a gross hourly wage of 26.47 EUR/h, which meets both the published data of [54,55].

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 A1) [56].

5. Results and Discussion

5.1. Simulation Results

For CO₂ 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% [49]. Therefore, relevant comparative parameters for Equation (3) were considered with particular detail during simulations. The most important results are shown in Table 6. In addition, columns 4 and 6 in Table 7 show the simulation results for the CO₂ absorption plant considered in this work, as well as the reevaluation of the reference plant, considered in [43,49].

Table 6. 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	90%

The sizing of the effective column diameters of the absorber and desorber is crucial for the adequate operation of CO₂ capture processes from flue gases. These were determined in the developed ASPEN Plus model by linear optimization with a 90% CO₂ capture rate as a boundary condition. For the desorber, this results in a diameter of 0.95 m for the flue gas volume flow rate and composition specified in Table 2. The effective desorber column diameter is determined as 0.50 m. The required heat output of the desorber of 556.5 kW, or the specific heat requirement of 3354 MJ/kg CO₂ must be provided by the reboiler. The CO₂ capture system can avoid 8641 t CO₂/year.

5.2. Cost Calculation Results

Cost calculations for the CO₂ absorption plant were determined considering the following steps: (i) calculation of equipment cost (Section 5.2.1); (ii) calculation of capital expenditures (CAPEX, Section 5.2.2); (iii) calculation of operational expenditures (OPEX, Section 5.2.2); and (iv) the calculation of resulting CO₂ separation cost (Section 5.4).

5.2.1. Equipment Cost

Table 7 gives an overview of the equipment cost for both the modeled CO₂ absorption plants. Details concerning the reference plant are given in [49]. The plant for this work was designed for the same CO₂ capture rate of 90% and stripper operating conditions of 2 bar and 130 °C.

The absorber column is the most cost-intensive plant component, both in the original CO₂ separation plant and in the newly designed plant for the glass industry. The costs for the absorber are approximately EUR 340,000. The second most important contribution to equipment costs is related to the stripper column, at approximately EUR 110,000. Accordingly, another 20% of the total costs are contributed by the stripper. To date, 85% of the total equipment costs can be attributed to the most important columns in the process. Other important and cost-relevant items are the cold water pump, blower and DC water cooler with a combined investment cost of approximately EUR 51,000. The previously described parts cover 96% of the total component costs. The remaining plant components such as rich/lean heat exchanger, solvent pumps, reboiler, or the chiller for the exhaust gas cooler cooling supply can be attributed a total cost of EUR 20,000, based on the given percentage share of [49]. Since these components account for only 4% of the total cost, this summarized cost estimate is sufficient for these components. This cost allocation pattern is consistent with the data in [49] and can thus be considered valid.

Table 7. Equipment cost calculation. Indicators (Ind.) refer to Figure 2b for the easier identification of equipment. * Remaining heat exchanger and pumps with less significant influence on the total component costs.

Ind.	Equipment	Type	Exponent		Reference Plant	Own Plant	
			m	G_r	k_r in millions of EUR	G	k_e in EUR
COL 2	Absorber	Column	0.60 [57]	12,560 m ³	10.94	39.25 m ³	340,000
COL 4	Stripper	Column	0.60 [57]	3391 m ³	3.43	8.75 m ³	110,000
–	Cold water pump	Pump, reciprocating	0.34 [53]	1.21 m ³ /s	2.04	0.0015 m ³ /s	21,000
PUM 1	Blower	Blower, centrifugal	0.59 [53]	485.44 m ³ /s	3.10	2.236 m ³ /s	13,000
COL 1	Flue gas cooler	Column	0.60 [53]	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 compressor	Compressor, rotary, two stage [52]		31.49 m ³ /s	31.73	0.07761 m ³ /s 0.167 kg/s	120,000

The total costs for CO₂ compression after separation, calculated as described in Section 4.2, were approximately EUR 153,000. Alternatively, using the second approach described in Section 4.3 would yield investment costs of EUR 69,000, but only for a single-stage compression process. A single-stage compressor, however, would have to achieve a compression ratio of 1:10. Such a high compression ratio is unfavorable for thermodynamic reasons in single-stage design and would lead to high heat generation, high compressor operating power, and thus high energy demand. Thus, the single-stage compressor is extended to a two-stage compression process, yielding a compression ratio of 1:5 in the first compressor stage. Therefore, the costs calculation of method 2 would double, yielding costs of EUR 138,000. Finally, the costs were validated against vendor information, giving estimated costs of EUR 120,000 for a two-stage reciprocating compressor [52]. For further calculations, the cost estimated by the vendor was used, as this can be considered the most practical approach.

5.2.2. Capital Expenditures

In addition to the equipment cost, additional investments for installation, control engineering, pipes, and electrical installation have to be considered. These cost are calculated according to the methodology shown in Section 4.3. The cost elements can be divided into direct costs for materials and further equipment, and indirect costs for services such as engineering and construction expenses of the plant. The direct cost elements can be further structured according to inside battery limit (ISBL) cost, and outside battery limit (OSBL) cost. ISBL includes material costs, which are directly associated to the plant, while OSBL are secondary expenses for building, yard improvement, or service facilities.

Ranges for the assumed percentages of ISBL costs are given in [53]. Table 8 shows the parameters taken into account and their influence on the total CAPEX. Due to the comparably small size of the CO₂ capture plant, the lower limits of the ranges proposed in [53] were selected. Thus, the additional costs for construction and installation, instrumentation, and control, as well as piping and electrical equipment are approximately EUR 354,000, or approximately 68% of the component costs. The total material and equipment cost of the CO₂ separation plant for the glass industry are approximately EUR 875,000.

The OSBL costs for buildings, yard improvements, and service facilities were assumed to be lower than recommended by [53]. Due to the low column volumes and the resulting compactness of the plant, a container solution for plant housing is attractive. Considering this option, OSBL costs of over EUR 100,000 building infrastructure seems justified, or even on the safe side. The excavation and concrete construction work will be limited with this simplified type of building design. Service facilities such as changing rooms and showers for personnel or workshops can be provided by the existing infrastructure at a glass manufacturer site. The total direct costs thus amount to approximately EUR 980,000.

For indirect cost, ranges suggested by [53,58] are used. This results in total indirect costs of approximately EUR 107,000. For the calculation of the total plant investment costs (FCI), the investment costs of the CO₂ compressor must also be considered. In total, the FCI add to approximately EUR 1.15 million.

In addition to the FCI, costs for labor arising in the company, start-up, and initial material costs must be taken into account. The costs for start-up and initial material costs amount to approximately EUR 92,000. The material cost is mainly for the initial loading of the solvent circuit. This consists in a 30 wt. % solution of monoethanolamine (MEA) in water.

The component costs were determined by cost comparison with a CO₂ separation plant for coal-fired power plants, which was designed as early as 2007. Since then, due to inflation, as well as global geopolitical events such as the COVID-19 pandemic, non-negligible price increases have occurred in all areas of public life, which also affected chemical engineering. In order to take these developments into account, a price index correction, based on [56] was applied. This price index correction since 2007 has a significant impact of 21% on total CAPEX. The extent to which prices developed according to current influences such as recent conflicts, should be closely examined. These effects have not yet been taken into account in the current values of [56] and may require reassessment for future works.

The CAPEX of plants for CO₂ capture from flue gases of power plants are far above the values calculated in this study. Abu-Zahra and Singh proposed EUR 147 million and EUR 179 million, respectively [49,59]. With respect to CAPEX, there is a factor of 90 between both plants, and with respect to the absorber column dimensions, there is even a factor of 300. This is mainly due to the significantly lower flue gas mass flow of the investigated glass melting tank. In the investigated case study, 0.569 kg/s are present at the glass industrial plant, while 616.0 kg/s are present at the reference plant of the coal-fired power plant. To allow a more specific comparison, the required absorber column volume per ton of CO₂ captured is calculated. The glass industry plant needs $4.54 \cdot 10^{-3} \text{ m}^3$ absorber/t CO₂ captured, while the reference plant needs only $3.44 \cdot 10^{-3} \text{ m}^3$ absorber/t CO₂ captured. Accordingly, for the specific amount of flue gas occurring in the glass industry, a larger absorber column is necessary. This is most likely due to the high levels of ambient air flowing into the investigated flue gas filter unit. The ambient air increases the exhaust gas volume while reducing the CO₂ partial pressure in the cleaned flue gases rich on CO₂.

Nevertheless, the large scaling factor between the reference plant and the designed plant is outside the limit proposed by [53] for the applicability of the cost comparison method. However, the determined cost frameworks appear plausible by comparing specific CAPEX to other literature sources. Ref. [35] investigated the CO₂ capture for carbon-intensive industrial processes and reported a specific CAPEX of 160 EUR/t CO₂ and year for amine-based post-combustion CO₂ capture in cement industry processes. The resulting specific CAPEX of this work is 188 EUR/t CO₂ per year. Considering the price increases since 2012 (approximately 16% according to [56], as can also be seen in Figure A1), the deviation of 15% calculated in this work seems justified.

It should be noted that the applied boundary conditions and assumptions of this work may have a significant impact on the implementation costs of a CO₂ capture plant in glass melting processes. However, the figures proposed herein should provide a sufficiently detailed basis for an initial cost estimation that could serve as a valuable basis for further investigations.

Table 8. Total capital investment (CAPEX) for MEA CO₂ separation plant.

	Range [53] %	Used %	Cost EUR
Direct cost			
Inside battery limit (ISBL) cost			
Total equipment cost (EC)	-	-	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:	979,480
Indirect cost			
Engineering	10	10	52,100
Construction expenses	10	10	52,100
Contractor's fee	0.5	0.5	2605
Contingency	17	17	88,570
		Total indirect cost:	106,805
CO ₂ compressor equipment [52]			120,000
		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 total direct and indirect cost		21.5	248,386
		Total capital investment (CAPEX):	1,634,278

5.2.3. Operational Expenditures

The operating expenses (OPEX) for the CO₂ capture plant can be further divided into fixed charge, direct production costs, plant overhead costs, and general expenses. Table 9 shows an overview of the calculated OPEX. According to the methodology described in [53,58], ranges or fixed percentages for the respective cost categories are given.

The fixed costs of the CO₂ capture plant include local taxes, fees, and charges, as well as insurance rates. Lower percentages of the range suggested in references [53,58] were specified for this purpose, as there are currently no specific tax rates or fees for such plants in Germany. The insurance coverage will primarily include fire insurance, the costs of which are based on the investment volume, fire risk, and the business activity. Based on the investment costs of EUR 1.63 million determined in Section 5.2.2, the low fire risk of the plant (essentially short circuit and cable fire, hardly flammable materials) and the low fire-prone glass industry, the resulting fixed costs of approximately 18,000 EUR/year (1.1% of CAPEX) appear sufficient.

Table 9. Total operating expenditures (OPEX) for MEA CO₂ separation plant. Ranges are from [53], others are named.

	Range %	Used %	Cost EUR/Year
Fixed charge			
Local taxes	1.0–4.0% of FCI	1.0%	12,063
Insurance	0.5–1.0 % of FCI	0.5%	6031
			18,094
Direct production cost			
Cooling water			4003
MEA makeup [49]		1.5 kg/t CO ₂	14,401
Maintenance (M)	1.0–10% of FCI	1.0%	12,063
Operating labor (OL) [54]	0.1 job/shift	45 EUR/h	39,096
Supervision and support labor (S)	30% of total labor cost	30%	11,729
Operating supplies	15% of maintenance	15%	1809
Laboratory charges	10–20% of operating labor	10%	3910
			87,012
Plant overhead cost	50–70% of M + OL + S	50%	31,444
General expenses			
Administrative cost	15–20% of OL	15%	5864
Distribution and marketing	2–20% of OPEX	2%	2848
Research and development cost	2–20% of OPEX	2%	2848
			Total operating expenditures (OPEX): 148,111

The direct production cost included the material requirements for plant operation, such as the cooling water, MEA makeup caused by degradation, and maintenance materials. The cooling water demand is based on simulation results. The MEA degradation was calculated based on the data given in Tables 3 and 5. The cost of the MEA makeup is approximately 14,000 EUR/year or 18% of the total OPEX. Other sources have reported an OPEX cost share of up to 32% [49,59]. Due to the size of the plant, the reduced cost share for the CO₂ separation plant in the glass industry seems plausible. However, the high O₂ content in the flue gas (see Table 2) may cause increased MEA degradation due to oxidation. For this reason, switching to more stable absorbents such as secondary or tertiary amines appears promising. These exhibit increased thermal and chemical stability against oxidative degeneration, as well as increased energy efficiency (as can be seen in Section 5.5). Besides these beneficial effects on operating parameters, the reduced energy demand and increased stability of advanced amines may result in a significant cost reduction.

The operating labor cost for the maintenance, operation, and monitoring of the plant, amounting to approximately EUR 39,000 per year, is the largest cost component (approximately 27%) in the annual OPEX. For this work, we assumed a 0.1 job per shift for the operation of the CO₂ separation plant. In the glass industry, a three-shift operation of 8 h 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 the maintenance and servicing of the CO₂ separation plant thus corresponds to a total time expenditure of approximately 2.5 h/day or 912.5 h/year. Other studies such as [49,59] 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 OL expenses seems justified. In addition, OL costs influence supervision and support labor costs, laboratory charges, and plant overhead costs. Taking all of this influence into account, OL costs contribute to approximately 60% of total OPEX.

Additionally, general expenses for distribution and marketing, as well as ongoing research and development, add up to approximately EUR 11,000 per year. As a result, the annual operating expenses (OPEX) amount to approximately EUR 148,000 in total. This represents approximately 9% of the total CAPEX. [35] assumed an OPEX range of 7–12% of total CAPEX for a post-combustion MEA separation plant in the cement industry. Therefore, the OPEX calculated in this work can be considered consistent with other literature sources and even indicate potential for further optimization.

5.3. Cost for Desorption Heat Generation

The supply of the necessary heat for desorption is crucial for the economic viability of a post-combustion CO₂ absorption plant (Section 2.3). As shown in Table 4, the heat demand for the designed CO₂ separation process is 928 kW, at a steam temperature and pressure of 150 °C 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.

5.3.1. 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. Such a reactor can provide high pressure and temperature steam (HP-steam) and low pressure and temperature steam (LP-steam, see Figure 3). Table 10 shows the power distribution of a tubular bulb reactor based on vendor information [36]. It is evident that the HP steam of the methanation reactor can already cover approximately 120% of the heat demand for CO₂ desorption. According to vendor information, a specific CAPEX of 40 EUR/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 approximately EUR 49,000 can be considered for the 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%, an exact thermal cost of 0.0004 EUR/kWh was achieved.

Table 10. Waste heat potential of the 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 °C	1.09
LP-steam	30 bar, 240 °C	0.12
Power loss	Heat losses	0.30
Methanation reactor	Combined	7.60

5.3.2. 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 approximately 850 kW for a ΔT of 247 °C. This is not sufficient to provide the required thermal power of the reboiler of 915 kW.

The flue gas needs to be cooled to approximately 130 °C to ensure sufficient heat output for adequate steam generation. At this ΔT of 340 °C, an enthalpy of 1171 kW can be used. Based on vendor information and calculations, a five 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 highly corrosive exhaust gases, stainless steel would be suitable as a construction material for this heat exchanger. A cost estimate for these material options is approximately EUR 40,000 for conventional steel, and EUR 90,000 for stainless steel, respectively, [37]. Assuming a lifetime of 20 years

and an interest rate of 5%, specific costs of 0.0007 EUR/kWh thermal for common steel and 0.001 EUR/kWh thermal for stainless steel are achieved.

The lower outlet temperature of the flue gases from the filter system would also affect the CO₂ separation system. As a result, the capacity of the flue gas cooler could be reduced, which in turn would result in lower investment costs. However, 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.

5.4. Cost of CO₂ Separation

With a lifetime of 20 years and an interest rate of 5%, the annual depreciation or EAC costs of the CO₂ separation plant designed in this work are approximately 135,000 EUR/year. As shown in Table 9, there are additional OPEX of approximately 146,000 EUR/year. The power required for CO₂ compression can be calculated to 73 kW using the method described in Section 4.2. However, vendor information named a compressor peak power of 55 kW and an operating power of 41 kW. Using the vendor information, CO₂ compression has an energy demand of approximately 357 MWh/year, or approximately 41 kWh/t of CO₂ captured, for the compressor operating hours of Table 4. Specific electricity costs are taken from [8] where 0.12 EUR/kWh were assumed for glass industry companies. The three cost factors EAC of CAPEX, OPEX, and electricity demand for the CO₂ compression add up to an annual cost of EUR 353,000 for the CO₂ separation plant designed in this work.

The heat demand for desorption in year-round operation is 8017 MWh/year, or 928 kWh/t CO₂. The described CO₂ capture plant can avoid 8641 t CO₂/year (Table 4). The three options for waste heat utilization, described in Section 5.3, were used to investigate their influence on CO₂ separation costs. The results are shown in Table 11. The CO₂ separation costs are 38.07 EUR/t CO₂ for the waste heat utilization of the methanation process. For the flue gas heat exchanger option, slightly higher costs of 38.42 EUR/t CO₂ or 38.73 EUR/t CO₂ were achieved. Above all, the lower heat output that these heat exchangers can generate, but also the higher material costs for the stainless steel version have an impact on additional costs. Nevertheless, these cost increases are very small and illustrate the potential for cost-effective waste heat utilization for absorption-based CO₂ capture in the glass industry.

Table 11. Calculation of CO₂ separation costs for different waste heat utilization options. EAC = equivalent annual costs of CAPEX.

		Specific Cost	Total Cost EUR/Year
EAC of CAPEX		189 EUR/(t CO ₂ ·year)	134,895
OPEX			146,440
Electricity for CO ₂ compression	0.12 EUR/kWh	4,95 EUR/t CO ₂	42,745
		Fixed costs:	325,751
Heat supply cost options			
	0.0004 EUR/kWh	0.37 EUR/t CO ₂	3207
1. Methanation reactor		Total annual costs:	328,958
		CO ₂ separation cost:	38.07 EUR/t CO ₂

Table 11. Cont.

2. Flue gas heat exchanger conventional steel	0.0007 EUR/kWh	0.65 EUR/t CO ₂	6235
		Total annual costs:	331,986
		CO ₂ separation cost:	38.42 EUR/t CO ₂
3. Flue gas heat exchanger stainless steel	0.0010 EUR/kWh	0.93 EUR/t CO ₂	8908
		Total annual costs:	334,659
		CO ₂ separation cost:	38.73 EUR/t CO ₂

Other studies reported CO₂ separation costs from industrial waste gases to range from 25 to 135 EUR/t CO₂. However, these reported values depend very much on the used CO₂ capture technology. Comparable process designs in the cement industry with post-combustion CO₂ capture using MEA, assume short- to mid-term CO₂ separation costs to be approximately 65 EUR/t CO₂ at a low cost for external steam import [35]. In studies investigating CO₂ capture from industrial point sources for methanation, 50 EUR/t CO₂ was found [60]. However, they assumed a lower CO₂ compression level of 10 bar and no detailed specification of industrial point sources was given.

5.5. Options for CO₂ Separation Process Improvement

The CO₂ separation costs of 38–39 EUR/t CO₂ found indicate the promising potential for low-cost and energy-efficient integration options for the post combustion CO₂ separation within the introduced PtM process for the glass industry. In addition, there are further options for optimizing the CO₂ 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 CO₂ separation costs. This can be achieved by the reduction in the energy requirement for desorption and lower investment cost for the plant equipment because of reduced liquid flow rates [49].

MEA is considered to be the standard solvent for post-combustion CO₂ absorption. However, as mentioned in Section 2.2, MEA shows disadvantages in terms of thermal and chemical stability. Chemical degradation caused by oxidation can especially be an issue for the application of MEA-based CO₂ separation processes in the glass industry. The high O₂-content of 20.5 vol-% in the investigated flue gas composition might increase oxidative degeneration (see Table 2). These effects can be avoided by changing the absorbent. The substitution of the primary amine MEA with secondary or tertiary amines such as methyl-diethanolamine (MDEA) or mixtures such as methyl-diethanolamine with piperazine (MDEA-PZ) are discussed in the literature for more stable CO₂ absorption processes. These solvents are not improved with regard to higher thermal or chemical resistance, but also enable a higher CO₂ loading capacities, while at the same time reducing the desorption heat demand. For example, the desorption heat demand of MDEA-PZ is reported as 2.6 GJ/t CO₂, while the heat demand of MEA is 3.4 GJ/t CO₂ [61,62]. Thus, a change of absorbents could not only improve the chemical stability but also further reduce the energy demand and CO₂ capture costs, respectively.

In addition to solvent replacement and changing solution properties, an advanced process design could also positively influence the costs of CO₂ separation. Improved designs for the rich/lean heat exchanger [63], vapor recompression, and split-stream processes [64], have been investigated and shown promising potential for enhancing the CO₂ removal capacity for reduced energy demand.

Combinations of these improved process designs and careful solvent selection could even further optimize the costs and performance of absorption-based CO₂ separation from flue gases of glass melting processes.

6. Conclusions

In this work, an innovative concept for the integration of a PtM process into oxyfuel glass melting processes is presented. The state of the art of the main process components is described and recommendations for suitable technology options are given. The required capture of CO₂ from the glass industry flue gases were identified as the process that requires more detailed research, as it has not been discussed before. Post-combustion CO₂ absorption capture processes, which have also been applied in power plant technology, were identified as the most promising option. 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:

- Amine-based CO₂ capture processes allow a reduction of −90% of CO₂ emissions of combustion-based glass melting processes. The designed CO₂ absorption plant for the glass industry is approximately 400 times smaller than the comparable concepts for power plants, due to the much lower volume of produced 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. For CO₂ desorption, the methanation process can be seen as the most suitable option as it provides steam at sufficient temperature and pressure, with no impact on the existing glass industry infrastructure.
- The resulting low operating costs result in low CO₂ separation costs of 38–39 EUR/t CO₂. These could be further reduced by described optimization approaches, such as changes in solvent loading, the use of advanced solvents, and/or advanced CO₂ absorption processes.

Further studies should start with these optimization approaches and can thus provide a basis for later cost-efficient experiments on real glass melting plants.

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Conflicts of Interest: The authors declare no conflict of interest.

Appendix A

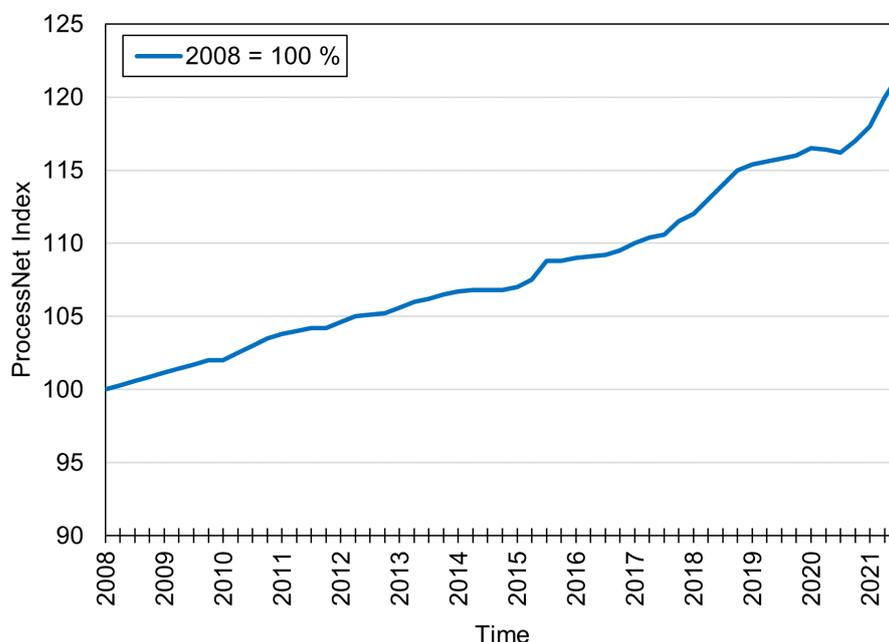


Figure A1. Quarterly development of the ProcessNet—Chemical Plant Price Index Germany since 2008.

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