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Techno-economic performance of enhanced sodium carbonate-based CO_2 capture process

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ABSTRACT

A simulation model was developed, and a techno-economic analysis (TEA) was conducted for a novel carbon dioxide capture process based on sodium carbonate solution. The key innovation in this process is the incorporation of a microbubble generator, which significantly enhances mass transfer and improves reaction rate. The model, based on prior experimental data, was used to evaluate CO_2 capture costs for the non-optimized base case, identify key cost factors, and assess potential process improvements.

The base case capture cost was estimated at EUR 114–133/tCO_2, which is higher than typical costs for amine absorption. The high costs are largely due to the high liquid flow rates required for efficient mass transfer and the need to maintain low solution concentrations to prevent salt precipitation. A notable advantage of the process is its low-temperature regeneration (~65 °C), which enables the use of waste heat. When waste heat is available at no cost, capture costs could be reduced by EUR 30–40/tCO_2. The use of a heat pump was found to be economically favorable when heat costs exceed EUR 13–20/MWh, depending on electricity prices. Increasing absorption pressure and temperature were found to be promising means for improving the process performance.

1. Introduction

Carbon capture, utilisation and storage (CCUS) is seen as one of the main means for mitigating climate change. For example, in the IEA's Sustainable Development Scenario (IEA, 2020), CO_2 emissions from the energy sector are expected to decrease to net zero by 2070 and almost 15 % of this reduction, compared to the Stated Policies Scenario, would originate from CCUS. Furthermore, capturing biogenic CO_2 and permanently storing it either underground or in products results in negative emissions. Negative emissions are an important tool for offsetting the residual emissions from hard-to-abate sectors like aviation, the cement and steel industry and agriculture to achieve overall carbon-neutrality (Hannula and Melin, 2021).

CO_2 can be captured from various sources, such as flue gases from power and waste-to-energy plants, recovery boilers, cement kilns, and even from the air. After capturing, CO_2 can be either permanently stored in geological formations, for example, or it can be used as a carbon source to produce various chemicals, chemical intermediates or fuels, when combined with renewable hydrogen, enabling sustainable routes for current petrochemical-based products. Although various technologies for CO_2 capture already exist, there is a demand for more economical and environmentally benign technologies.

Chemical absorption is the most proven and common technology for CO_2 capture (Isa et al., 2018). Chemical absorption is a mass-transfer phenomenon where, in the case of carbon capture and storage (CCS) application, CO_2 is absorbed into the liquid through dissolution followed by a chemical reaction. CO_2 is then released in the subsequent step called stripping or desorption, which takes place either at elevated temperatures or at sub-atmospheric pressure. There are various liquid absorption technologies that have been recently reviewed by (Ochedi et al., 2021).

For post-combustion CO_2 capture, amine processes have been the most widely used due to their fast reaction rates, suitability for low CO2 partial pressure applications and high CO_2 removal efficiencies (Knuutila et al., 2009). Nevertheless, several limitations hinder widespread adoption of this technology, including elevated energy needs for solvent regeneration, significant corrosion risks, toxicity, absorbent loss due to low stability, the formation of carcinogenic compounds, and substantial costs (Knuutila et al., 2009; Meng et al., 2022; Ochedi et al., 2021). Monoethanolamine (MEA) has been considered as the

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"benchmark amine" for a long time. However, nowadays more advanced proprietary capture solvents are offered by various technology suppliers (Barlow et al., 2023; Feron et al., 2020; IEAGHG, 2019). These advanced amines provide better process and environmental performance compared to MEA through the use of novel mixtures of amines and adding inhibitors to mitigate corrosion and degradation, for example (Barlow et al., 2023; IEAGHG, 2019). A conventional flowsheet including absorption and desorption is presented by (Dziejarski et al., 2023).

Carbon capture using alkali carbonates would offer several benefits compared to amine systems. Potassium and sodium carbonates, and the corresponding bicarbonates, are non-hazardous compounds that are widely used in industry (Knuutila et al., 2009) They do not form toxic by-products and their volatility is also very low, which reduces solvent loss due to degradation and eliminates the need for wash stages and reclamation units (Isa et al., 2018). In addition, carbonates are much less corrosive than amines, enabling the use of less expensive materials (Karali et al., 2022). Furthermore, carbonates are relatively low-cost chemicals. Lastly, the temperature required for releasing CO_2 from bicarbonates is significantly lower compared to amines, which could enable the utilisation of low-value waste heat from low-temperature water electrolysis, for example. (Nii et al., 1995) investigated vacuum regeneration on carbonate and concluded that regeneration at 343 K was optimal both from CO_2 desorption rate and water vapour evaporation perspectives.

The CO_2 absorption process using potassium carbonate solution has been widely applied, e.g. for synthesis gas and natural gas purification where the partial pressures of CO_2 are high. This so-called Benfield or Hot Potassium Carbonate (HPC) process was developed by Benson and Field as early as in the 1950s (Benson et al., 1954; Benson and Field, 1959). The main challenge regarding pure alkali carbonate-based CO_2 capture is the slow CO_2 absorption rate into the solution (Hu et al., 2016; Isa et al., 2018; Lu et al., 2011). The slow reaction rate is emphasised at post-combustion capture (PCC) conditions, where the partial pressure of CO_2 and temperature are low (Hu et al., 2016). This has limited the use of HPC for PCC applications as compressing the flue gases to adequate pressure has thus far been considered to lead to overly high costs (Barlow et al., 2023). Still, research has also been conducted to assess the suitability of potassium carbonate-based capture process for PCC for flue gases from coal-fired power plants (Mumford et al., 2012; Smith et al., 2014) and biomass-fired combined heat and power (CHP) plants (Gustafsson et al., 2021). Moreover, Capsol Technologies are offering their HPC-based technology CapsolEoP® for solid fuel plants and CapsolGT® for gas turbine plants (Capsol Technologies, 2023). In addition, a novel integrated vacuum carbonate absorption process (IVCAP) has been developed for PCC applications by researchers at the University of Illinois (Lu et al., 2012, 2011).

Sodium carbonate has been less commonly used for CO_2 capture compared to potassium carbonate. CO_2 capture with potassium carbonate is closely linked to the Solvay process (Steinhauser 2008), an industrial process developed in the 19th century for producing sodium carbonate. The Solvay process uses ammonia, water, carbon dioxide, and sodium chloride to produce sodium carbonate. At one stage CO_2 reacts with ammonia and brine to form sodium bicarbonate, a precursor to sodium carbonate. The Solvay process can be seen to have laid the foundation for the carbonate-based CO_2 capture technologies.

Various promoters and enzymes have been considered as a means for improving the reaction rate and therefore for reducing the size and cost of the absorption process (Borhani et al., 2015; Hu et al., 2016; Isa et al., 2019, 2018; Ochedi et al., 2021; Rochelle et al., 2007). The most widely studied promoters in potassium carbonate solutions have been amines, such as monoethanolamine (MEA), diethanolamine (DEA) and piperazine (PZ) but also acids, such as perchloric acid ($HClO_4$) or hypobromous acid (HBrO) have been considered. Carbonic anhydrase (CA) is the most studied enzyme. While significant increases in reaction rate can be achieved with CA, the harsh conditions of industrial absorption pose challenges for industrial application. (Hu et al., 2016)

Another drawback of carbonate-based capture processes, albeit less dramatic, is the relatively low water-solubility of alkali bicarbonates, especially sodium bicarbonate. The low solubility of bicarbonates into water leads to high solvent volume flows and thus high sensible energy needs in regeneration. To cope with the low solubility, concepts where carbonates are allowed to form a slurry have been suggested (Anderson et al., 2014, 2013; Knuutila et al., 2009; Pandit et al., 2014; Smith et al., 2015). In their conceptual study, (Knuutila et al., 2009) found that by allowing sodium bicarbonate to form a slurry, the energy requirement of

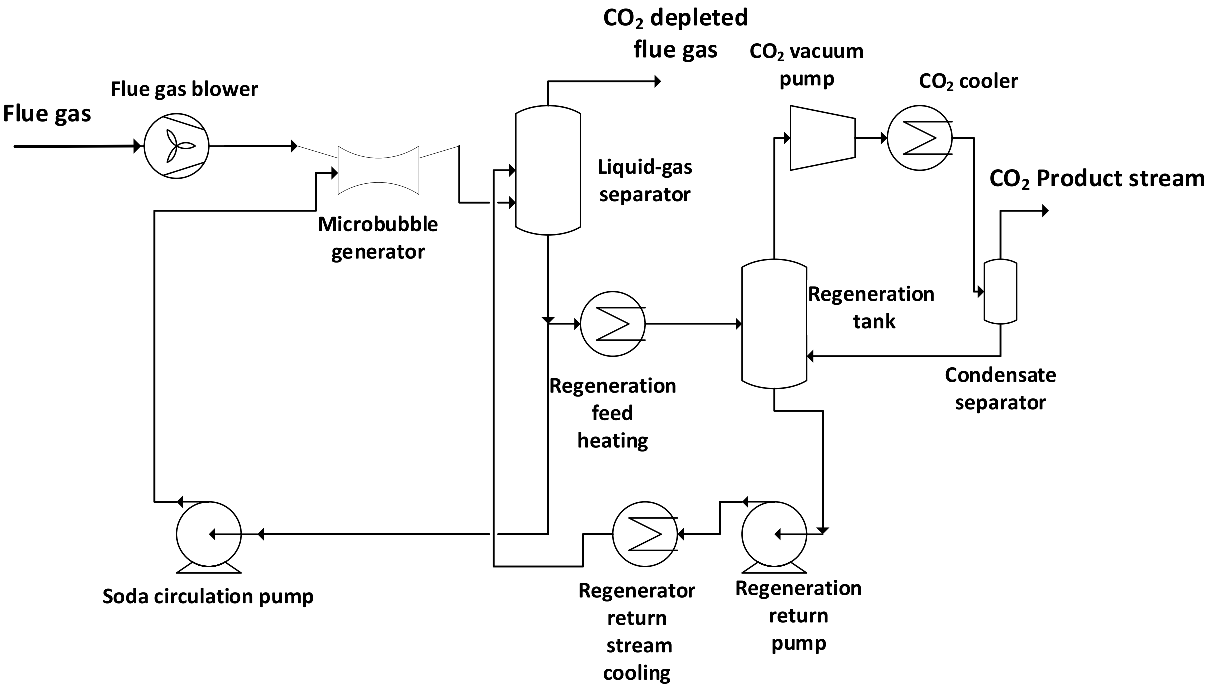

Fig. 1. Conceptual process diagram illustrating the novel Na_2CO_3 based capture process.

stripping of the solvent could potentially be around 3.22 MJ/kg of captured CO_2, which is significantly lower than with MEA-based systems whose energy consumption is typically around 3.8 MJ/kg of captured CO_2. According to (Smith et al., 2015), by allowing potassium bicarbonate to precipitate, the energy requirements can be decreased from over 3 GJ/tonne of CO_2 to <2.5 GJ/tonne of CO_2.

In practice, the aim is commonly to prevent the precipitation of bicarbonates, as this causes significant engineering challenges such as fouling or clogging of the process equipment (Fosbøl et al., 2013; Hu et al., 2016). However, KC8 Capture Technologies, for example, are commercialising their UNO MK 3 technology, which uses a proprietary precipitating potassium carbonate (K_2CO_3) solvent (Barlow et al., 2023). At the core of the process is a patented turbulent bed contactor technology that can handle precipitation while providing process intensification at the same time. The process has already been demonstrated at the pilot scale, while two demonstration-scale facilities are scheduled to begin operation in 2024, indicating that the slurry phase PCC process can also be realised in practice (Barlow et al., 2023). Slurry phase processes have already been used when the aim is to produce bicarbonate as a product. For example, the Dutch company Twence B.V. captures CO_2 from flue gases at their waste-to-energy plant using amines and combines the captured CO_2 with sodium carbonate to produce a sodium carbonate slurry in a recently developed process by Procede Gas Treating B.V. (Huttenhuis et al., 2016)

Another development has been to use highly permeable, micro-encapsulated carbon sorbents (MECS), which combine the advantages of liquid solvents (i.e., high capacity, selectivity, water tolerance) and solid sorbents (high surface area, low volatility). The idea of encapsulating the carbonate sorbents (either potassium or sodium carbonate solution) is to overcome their own main drawbacks of slow CO_2 absorption kinetics and difficulty in handling precipitated solids (Vericella et al., 2015).

In addition, process intensification technologies have been developed for improving mass transfer. For example, Carbon Clean has developed rotating packed beds in which a rotating disc of packing material creates centrifugal force far greater than the gravitational force seen in conventional columns. It is claimed that the mass transfer improvement allows up to 10 times smaller columns. Their technology also includes a proprietary amine-promoted buffer salt solvent (APBSCDRMax®) (Barlow et al., 2023).

The purpose of this paper is to provide a techno-economic analysis of another novel concept for improving the absorption rate for a sodium carbonate-based CO_2 capture system. In this concept, the absorption rate is improved by adopting a "micro bubble generator" instead of a traditional absorption column. The concept has been presented in detail by (Kajolinna et al., 2024), where the experimental results are presented, while this paper provides insights into the techno-economic feasibility. The aim is to highlight the most critical aspects affecting the capture costs, and analyse possible improvements and most favourable process integration options. Detailed optimisation of the process is out of the scope of this paper.

2. Process description

The novel carbon dioxide capture process using aqueous Na_2CO_3 as the absorbent is illustrated in Fig. 1 and described in more detail in Kajolinna et al. (2024). The feed gas is pressurised with a feed gas blower and then mixed with the aqueous solution of Na_2CO_3 and $NaHCO_3$ using a microbubble nozzle to enhance the mass transfer between gas and liquid. The idea is to overcome the traditional limitation of carbonates, which is the slow mass transfer in gas absorption by generating small microbubbles with a high surface area. After the absorption of CO_2, the CO_2-lean flue gas containing mostly nitrogen, remaining CO_2 and oxygen is separated from the soda solution in a vertical tank.

To convert sodium bicarbonate back to carbonate, a small part of the solution is fed to regeneration while the rest is recycled to the microbubble generator. The need for only a small fraction to undergo regeneration arises from the microbubble generator's demand for a high liquid-to-gas (L/G) ratio. The soda solution circulation though the regeneration is adjusted so that enough bicarbonate is regenerated to keep the level of bicarbonate constant in the absorption, assuming that only 30 % of the bicarbonate is turned into carbonate in the regeneration step according to experimental observation from earlier work. For efficient mass transfer, a higher solution flow compared to the amount required solely by the chemical reaction is needed. The regeneration of the soda solvent is conducted under medium vacuum at a temperature of 65–75 °C, also enabling the utilisation of waste heat as the heat source. After the regeneration, the pressure of the gas stream is increased by a vacuum pump and the heat from the hot gas is used as an additional heat source for the regeneration step. After cooling the CO_2 rich gas stream, the condensed water is separated and a pure stream of CO_2 is obtained. The regenerated soda solution is returned after cooling back to the liquid-gas separator so it can be reused in the absorption step.

3. Goal and scope

In this paper, we first develop a flowsheet for the novel process for industrial-scale implementation. We assume that CO_2 is captured from a typical flue gas stream of a large-scale wood-fired combined heat and power (CHP) plant equipped with a flue gas scrubber. The considered annual capture capacity is approximately 12.1 t/h, equivalent to 105 kt CO_2 per year.

To assess the mass and energy balances, we create a simulation model using Aspen Plus software. The model incorporates experimental data derived from prior research conducted by (Kajolinna et al., 2024). The prior research report results such as CO2 capture ratio, CO2 absorption rates measured during proof of concepts tests in a bench scale device. The process conditions and key parameters determined in this previous work were used as input in this paper to evaluate the techno-economic performance of the unoptimised case of the process. This assessment considers estimated mass and energy balances, equipment sizes, costs and economic parameters.

Lastly, we investigate the key factors affecting the techno-economic performance of the process the most, and potential means to improve the unoptimised case that has not yet been verified experimentally. For these cases it is assumed that the capture ratio can be maintained at the same value as in the base case. These results gives indication whether these ideas attractive from techno-economic performance point of view and should be studied by future experimental work.

4. Methods

4.1. Process simulation and technical evaluation

The objective of modelling was to create a simulation model that could be used to estimate mass and energy balances for the novel CO_2 capture process at an industrial scale. The process was simulated with Aspen Plus© V11.1. Electrolyte NRTL (ELEC—NRTL) was chosen as the property method in the simulation. For better estimation of the properties, an Aspen Plus insert ECLSCR (special data package for caustic evaporator, including parameters for H_2O- Cl_2—CO_2—HCl-NaOH—NaCl-Na_2CO_3) for a temperature range of 0–200 °C, pressure up to about 1000 atm for CO_2 and concentration below saturation point. The heat demands in absorption and regeneration were verified by hand calculation using reaction heats based on heat of formation values from literature and latent heat of water evaporation. The relative amount of water evaporated in regeneration was estimated based on partial pressure of CO2 vs. water for the soda solution at the corresponding process conditions from the data presented by Knuutila et al. (2010).

Some of the main parameters, such as the concentration of soda solution, and the conversion rates and process conditions during

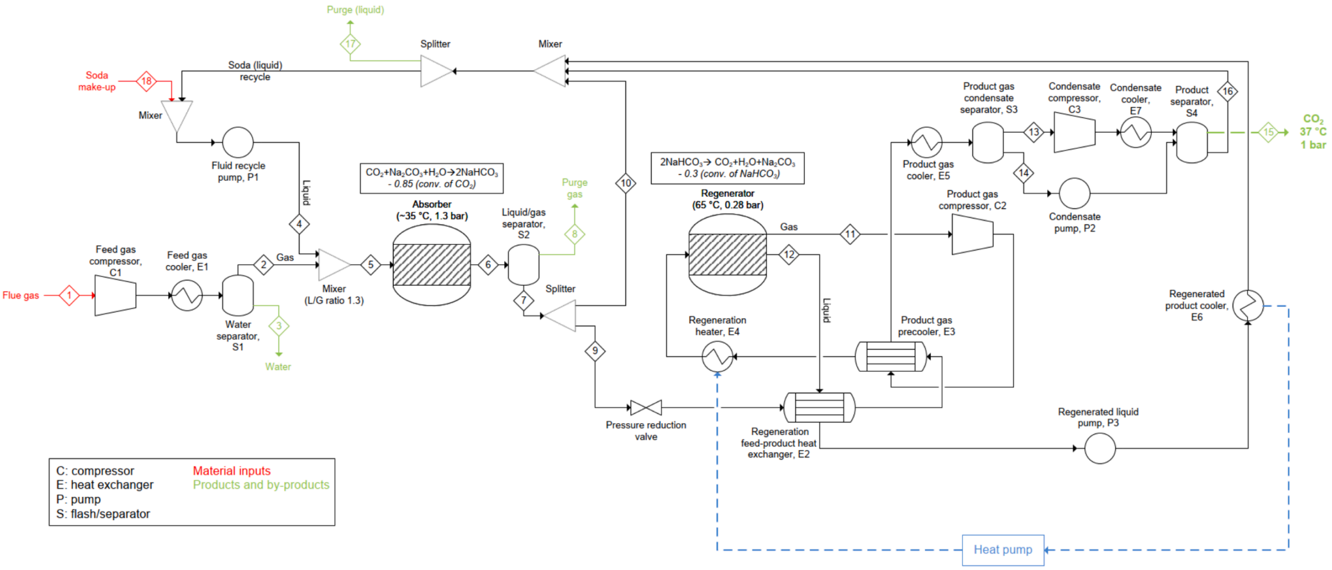

Fig. 2. Process flowsheet for the base case, including a heat pump.

Table 1
The composition of the flue gas.

Flue gas composition	mol-%
Carbon dioxide (CO_2)	12.9
Water (H_2O)	21.2
Oxygen (O_2)	3.0
Nitrogen (N_2)	62.9

absorption and regeneration steps, were adopted from the experimental work by (Kajolinna et al., 2024). Creating a rigorous model capable of predicting absorption and regeneration performances was out of the scope of this study. Efficiencies of compressors were estimated by default values of 72 % according to the isentropic method and pump efficiencies default values were used. Pressure drops in process equipment were not considered except a pressure drop of 0.5 bar between MBG, absorber and V/G gas separator.

The flowchart developed for the industrial scale implementation is illustrated in Fig. 2 and the key parameters of the model are described in this chapter.

The assumed flue gas composition (Table 1) represents a typical woody biomass-fired large-scale boiler equipped with a flue gas scrubber for heat recovery. For simplicity, the impurities (SO_2, HCl and NO_x) are not considered. The amounts of SO_2 and HCl after a flue gas scrubber are typically very low. In addition, the nitrous oxides consist mostly of NO, which is only very sparingly soluble in water, meaning that it does not markedly react with the soda solution.

In the first step, the flue gases (entering at 60 °C and 1.03 bar_a) are compressed to 1.5 bar_a and cooled to 45 °C for water removal. The pressure increase of 0.5 bar is selected based on an observed pressure drop of 0.5 bar between the MBG inlet and the purge gas exit. The gas stream is then mixed with the soda solution at a ratio of 1.3:1 (liquid-to-gas) which was shown to result in efficient mass transfer by (Kajolinna et al., 2024). The concentration of the soda solution given as pure Na_2CO_3 is ~7 wt-%. Prior to mixing, the pressure of the soda solution is raised from 1 bar_a to 1.5 bar_a to overcome the pressure drop observed in prior experimental work .

The mixed feed enters the absorber (modelled as RSTOIC component), which is assumed in the simulation of the base case to operate at 35 °C and 1.3 bar_a. It is assumed that 85 % of CO_2 is absorbed in accordance with experimental findings. The molar ratio between HCO_3^-/Na^+ was approximately 0.33 in the absorber liquid inlet and 0.34 in the outlet. The reaction occurring in the absorber is shown in Eq. (1):

$$CO_2 + Na_2CO_3 + H_2O \rightarrow 2NaHCO_3,\ \Delta H = -135\ kJ/mol_{CO2} \quad (1)$$

After the absorber, the feed is flashed to separate the CO_2-lean flue gas and soda liquid streams. The soda liquid stream proceeds to a splitter, from which 92 % of the stream is recycled back to absorption while the remaining 8 % continues to regeneration. The reason for recycling the soda liquid stream back to absorption is that the MBG requires a high liquid/vapor volumetric flowrate for efficient mass transfer and sending 8 % of the liquid to regeneration is sufficient to maintain the bicarbonate level constant in the absorption. Before the regenerator, the stream passes through a pressure reduction valve, reducing the pressure to 0.4 bar_a and through cross heat exchangers and a heater, heating the stream to 65 °C.

Regeneration takes place at 65 °C and 0.28 bar_a. It is assumed that 30 % of $NaHCO_3$ is converted based on experimental findings. The reaction equation for regeneration is shown in Eq. (2):

$$2NaHCO_3 \rightarrow CO_2 + H_2O + Na_2CO_3,\ \Delta H = 135\ kJ/mol_{CO2} \quad (2)$$

Two streams are produced in the regenerator: gas and liquid streams. The liquid stream is first cooled down in a cross-heat exchanger that provides pre-heating for the regenerator inlet stream. The pressure of the stream is then raised to 1.5 bar_a, and it is cooled down to 35 °C to then be recycled to an absorption step. The gas stream out of the regenerator is first compressed to 0.65 bar_a. It is then used for further pre-heating of the regenerator inlet stream, after which it is further cooled down to 30 °C and flashed. The pressure of gas and liquid streams is raised to 1 bar_a with a compressor and a pump, respectively. The streams are then combined to be flashed again. The gas stream is the product CO_2 stream, while the liquid stream goes to be recycled as the Na_2CO_3 fluid.

In an actual process, a small amount of recycle soda solution would be purged in order to avoid the accumulation of impurities. The purged sodium salts would be replaced with make-up soda, for example in the form of sodium carbonate. In the simulation model, the make-up is approximately 40 kg/h of fresh sodium carbonate. The make-up amount in the simulation is higher than actually needed, because using a very small amount of make-up would result in convergence challenges in the simulation.

The actual demand of the make-up was not evaluated in detail due to its negligible effect on costs. Even with the 40 kg/h assumption, the resulting make-up cost would have been only approximately EUR 1/

tonne of CO_2 captured, assuming a sodium carbonate price of EUR 330/t (Procurement Resource, 2023).

4.2. *Economic evaluation*

The main parameter used in the economic evaluation of the process is the capture cost of CO_2 (EUR/t). This cost includes capital costs (CAPEX) and operational costs (OPEX). The impact of CAPEX on the capture cost is calculated as investment annuity, using the annuity method. The annuity factor is calculated using Eq. (3), and investment annuity using Eq. (4).

$$f_a = \frac{r(1+r)^m}{(1+r)^m - 1} \tag{3}$$

where f_a is annuity factor, r weighted average cost of capital (WACC) or the target profit and m the plant lifetime.

$$a = f_a * FCI \tag{4}$$

where a is investment annuity and FCI is fixed capital investment.

The FCI is calculated using the Lang method described by (Towler and Sinnott, 2022). First, all the costs of individual equipment (C_e) are summed and then converted into fixed capital investment (FCI) using a Lang factor (F_L) of 4.74 suitable for fluid processes (Eq. (5)).

$$FCI = F_L \cdot \left(\sum C_e \right) \tag{5}$$

The equipment costs are estimated using the Aspen Process Economic Analyzer (APEA), except for the absorber, the regenerator, the liquid/gas separator (S2), the product gas compressor (C2), the fluid recycle pump (P1) and the feed gas compressor (C1). For the compressors and the pump, vendor estimates are used instead because they give a more accurate estimate, allowing the selection of the exact type of compressor/pump best suited for the intended purpose. The absorber and regenerator costs could not be estimated reliably using APEA and thus they are estimated based on literature data. The cost of the liquid/gas separator is estimated. The target level of detail of this study corresponds to an AACE class 4 cost estimate with an accuracy level of ±30 % (Towler and Sinnott, 2022).

In cases where the costs assume carbon steel as the material, material factors ($f_{material}$) are used to convert the costs to represent the costs for stainless steel SS316. This is done by using Eq. (6) and Eq. (7).

$$C_e = f_{material} \cdot C_{e,\ CS} \tag{6}$$

where $C_{e,CS}$ is the equipment cost in carbon steel and

$$f_{material} = \frac{Cost\ in\ the\ chosen\ material}{Cost\ in\ carbon\ steel} \tag{7}$$

The costs given in USD were converted to EUR using a EUR/USD rate of 0.924 according to the average exchange rate of 2023. The process equipment cost estimates from APEA, where estimated costs correspond to the year 2018, were converted to costs for 2023 by multiplying by the Chemical Engineering Plant Cost Index (CEPCI) ratio for February 2023 (806.3) vs. CEPCI 2018 (603.1).

The capacity scaling method (Eq. (8)) was used to scale the costs of the capture process from the reference capacity (S_{ref}) to other capacities (S_{new}).

$$C_{new} = C_{ref} \left(\frac{S_{new}}{S_{ref}} \right)^n \tag{8}$$

where C_{ref} = investment cost of the reference equipment

C_{new} = investment cost of the new equipment
n = cost exponent (0.67 from (Remer and Chai, 1990).

Table 2
Key technical and economic parameters.

Parameters	Value	Unit
Technical parameters		
Yearly full load hours of CO_2 capture plant	6500	h/a
Capacity of CO_2 capture plant	12.1	tCO_2/h
Coefficient of Performance (COP) for heat pump	5.5 (Marina et al., 2021)*	–
Economic parameters		
WACC	8 %	–
Lifetime	20	a
Electricity cost	45	EUR/MWh
Maintenance cost	4 %	of FCI
Specific investment cost of heat pump	400 (Marina et al., 2021)	EUR/kW heat transferred

* The estimate is based source temperature of ~ 35 °C and supply at 65 °C.

The capture cost of CO_2 is calculated using Eq. (9) and the parameters presented in Table 2.

$$CO2_{capcost} = \frac{(N_{elec}C_{elec} + N_{heat}C_{heat})O_pF + f_aFCI + \ M_c}{O_pF} \tag{9}$$

where

$CO2_{capcost}$ CO_2 capture cost, EUR/t of CO_2 captured
N_{elec} Amount of electricity, MWh/t of CO_2 captured
C_{elec} Electricity cost at CO_2 capture, EUR/MWh
N_{heat} Amount of heat needed in regeneration, MWh/t of CO_2 captured
C_{heat} Cost of heat, EUR/MWh
F Capacity of CO_2 capture plant, tCO_2/h
M_c Yearly maintenance cost, EUR
O_p Yearly full load hours for CO_2 capture plant, h/a

The assumed parameters used in the economic calculation are reported in Table 2.

5. Results

5.1. *Technical performance*

In this section the technical results determining the process performance derived from process simulation are presented. First the most important process streams are summarised in Section 5.1.1 and subsequently, the power and the heat demands are reported in Section 5.1.2.

5.1.1. *Process streams*

The most important stream flows and composition are shown for the considered capacity 12.1 t/h (see Table 3 and Table 1 in the electronic support material).

The CO_2 produced in the process has a purity of 94 mol-%. The remaining part contains 6 mol-% water and the content of O_2 and N_2 is <0.1 mol-%. Consequently, very high purity of CO_2 could be achieved by just drying the gas.

5.1.2. *Energy demands*

Based on the simulation for the base case, where 12.1 tonnes of CO_2 is captured, approximately 25 MW of external heat needs to be supplied (above 70 °C) to the process when the temperature difference between hot and cold streams is at minimum 5 °C (Fig. 3). The cooling demand (below 30 °C) is ~30 MW. This opens up a possibility to use a heat pump and make the process self-sufficient with respect to heat (Fig. 4). Thus, for the base case, a heat pump recovering heat from cooling of the soda solution after regeneration is added to the concept (see Fig. 2).

The minimum utility consumption can be compared to the actual

Table 3
The stream table illustrating flows in Fig. 2.

Stream number		1	4	7	8	9	12	15
Description		**Flue gas**	**Soda to absorption**	**Soda solution after absorption and V/L separator**	**CO_2 depleted flue gas**	**Feed to regeneration**	**Soda back from regeneration**	**CO_2 product**
Phase		Vapour	Liquid	Liquid	Vapour	Liquid	Liquid	Vapour
Temp.	°C	60.0	35.6	35.7	35.7	35.7	64.4	35.8
Pressure	bar_a	1.0	1.5	1.0	1.0	1.0	0.3	1.0
Mole %								
H2O		21.2	98.3	98.3	5.5	98.3	98.4	6.0
CO2		12.9	0.0	0.0	2.7	0.0	0.0	94.0
O2		3.0	0.0	0.0	4.2	0.0	0.0	0.0
N2		62.9	0.0	0.0	87.6	0.0	0.0	0.0
NAHCO3		0.0	0.8	0.9	0.0	0.9	0.6	0.0
NA2CO3		0.0	0.8	0.8	0.0	0.8	1.0	0.0
H2O	kg/h	9527	46,645,860	46,641,589	1790	3731,327	3716,494	313
CO2	kg/h	14,165	0	0	2106	0	0	12,052
O2	kg/h	2396	48	52	2392	4	0	4
N2	kg/h	43,965	0	0	43,965	0	0	0
NAHCO3	kg/h	0	1871,040	1917,075	0	153,366	107,356	0
NA2CO3	kg/h	0	2347,153	2318,112	0	185,449	214,474	0
Mass %								
H2O		13.6	91.7	91.7	3.6	91.7	92.0	2.5
CO2		20.2	0.0	0.0	4.2	0.0	0.0	97.4
O2		3.4	0.0	0.0	4.8	0.0	0.0	0.0
N2		62.8	0.0	0.0	87.5	0.0	0.0	0.0
NAHCO3		0.0	3.7	3.8	0.0	3.8	2.7	0.0
NA2CO3		0.0	4.6	4.6	0.0	4.6	5.3	0.0
Total Mass Flow	kg/h	70,052	50,864,100	50,876,828	50,252	4070,146	4038,324	12,369
Volume Flow	m^3/h	65,578	47,660	47,672	44,625	3814	3835	7448

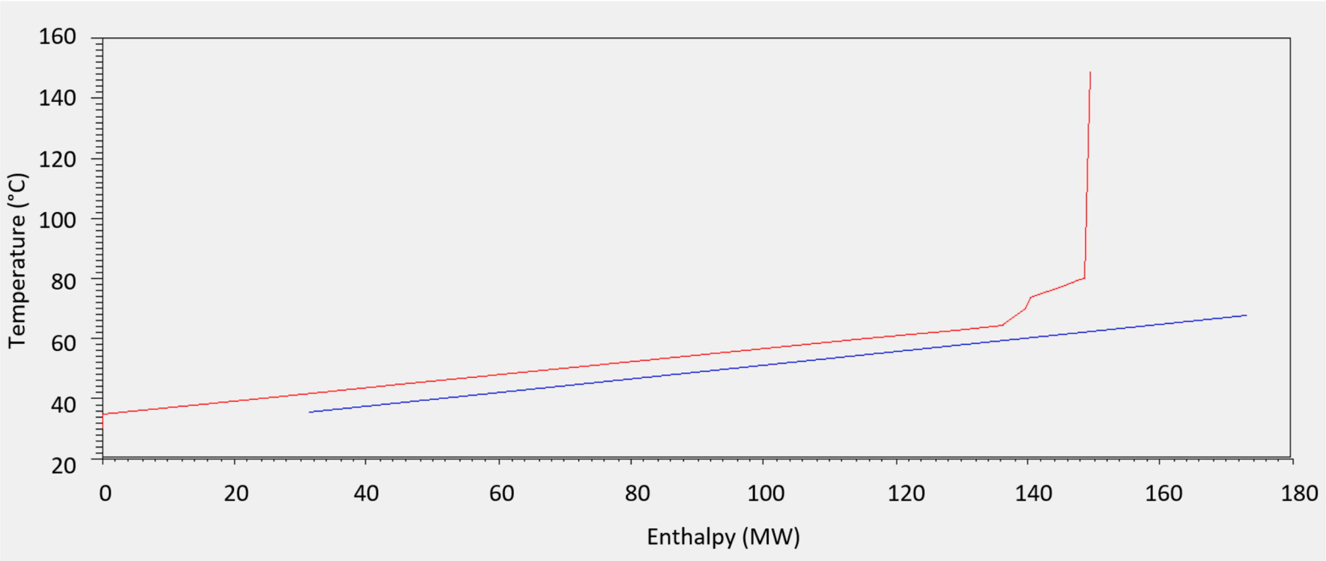

Fig. 3. Hot and cold composite curves with a minimum temperature approach of 5 °C between heat exchangers of cold and hot streams.

duties of heat exchangers in the simulated flowsheet. In the simulated flowsheet (see Fig. 2) the heater (E4) utilises heat supplied by the heat pump (25.1 MW). The heat pump uses heat from the heat exchanger (E6) and raises it to the needed temperature level for E4. Heat exchangers E7 and E5 use cooling water to cool hot process streams and E1 uses cooling air as a cooling medium. The other heat exchangers transfer heat between process streams in the process.

Table 4 shows heat exchanger details. The listed parameters of heat exchangers are duty, inlet and outlet temperature, area, and heat transfer coefficient. It can be observed that heat exchanger E2 has a high duty and very large area of >10,000 m2. The regeneration heater (E4) is the only one where external heat, 25.1 MW, is required.

A major part of the heating need is covered by the internal heat transfer in the process. The reason for this is that high liquid flow is fed

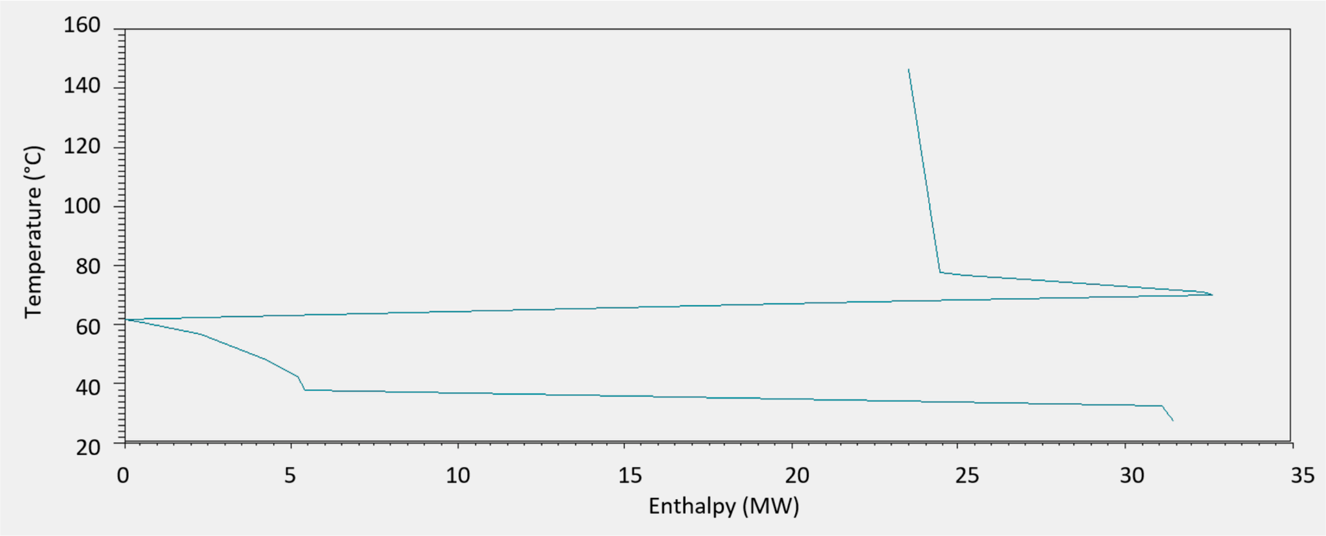

Fig. 4. Grand composite curve with a minimum temperature approach of 5 °C between heat exchangers of cold and hot streams.

Table 4
The duties and areas of heat exchangers in the process.

Heat exchanger	Duty	Hot Inlet T	Hot Outlet T	Cold Inlet T	Cold Outlet T	Area	Heat transfer coefficient
	[kW]	[°C]	[°C]	[°C]	[°C]	[m^2]	[kW/ m^2-K]
Feed gas cooler E1 (part 1)	1810	77.9	65	30	35	838.2	0.1
Feed gas cooler E1 (part 2)	3610	65.0	45	30	35	1764	0.1
Regeneration feed- product heat exchanger E2	104,000	64.4	40.7	35.7	59.3	10,760	2.4
Product gas pre-cooler E3	12,400	149	60.6	59.3	62.1	2180	0.6
Regeneration heater E4	25,100	75	75	62.1	67.8	3424	0.75
Product gas cooler E5	2318	60.6	30	20	25	65.92	1.8
Regenerated product cooler E6	25,300	40.7	35	25	25	2737	0.75
Condensate cooler E7	53	79.7	63	30	35	88.89	0

Table 5
Power consumption of different process equipment in the process.

Equipment	Duty	Unit
Feed gas compressor, C1	1004	kW
Fluid recycle pump, P1	770	kW
Product gas compressor, C2	1139	kW
Condensate pump, P2	0.2	kW
Condensate compressor, C3	155	kW
Regenerated liquid pump, P3	86	kW
All items without heat pump	3154	kW
Heat pump	4564	kW
Total electricity consumption	7718	kW
Total electricity consumption	640	kWh/t CO_2

to the regeneration, roughly 4000 t/h, and in addition, the absorption and regeneration stages are operated at different temperatures, 35 vs. 65 °C, respectively. A high liquid flow is needed to remove enough CO_2 from the soda solution in the regeneration step. In this case, a regeneration degree, i.e. (HCO_3^-/Na^+) molar ratio of 30 % (see Eq. 2), is assumed according to experimental data reported by (Kajolinna et al., 2024). Another reason is that a low salt content of ~7 wt-% expressed as pure Na2CO3 is used to prevent the precipitation of sodium bicarbonate. The low salt content has been observed to work well in experimental tests when the soda solution is cooled to outside temperatures between tests. However, in a larger-scale process, where the liquid is kept at a temperature above 30 °C, a higher salt content of ~10 wt-% could be used.

In Table 5, the process equipment using electricity is listed. It can be observed that the flue gas compressor (C1) and product gas compressor (C2) have the highest electricity demands. The fluid recycle pump (P1), although circulating a large volume of liquid in the absorption step, only has a duty of 770 kW due to the low head (5 m) required. All process equipment consumes roughly 3.2 MW of electricity in the CO_2 capture part of the process. When a heat pump is used to transfer 25.1 MW to the regeneration feed, it needs an additional ~4.5 MW of electricity, when the COP is 5.5. In total, the process needs about 7.7 MW of electricity, or 640 kWh per tonne of CO_2. When a heat pump is used, no heat from external sources is needed. The duties of pumps and heat exchangers are also shown for all process equipment in the Simulation diagram (see Fig. 2) in the electronic support material.

5.2. Economic results

5.2.1. Fixed capital investment

In Table 6, the purchase cost of individual process equipment can be seen. Also, the contribution of each piece of equipment to the total cost is reported in Fig. 5. It can be observed that the large heat exchanger E2 represents >45 % of the equipment costs. Although the slow absorption rate for sodium and potassium carbonate solution compared to amine systems is usually an important drawback, the absorber represents only ~1.6 % of the fixed capital investment. According to (Kajolinna et al., 2024) the measured value can be up to 10 times higher compared to a column system and therefore the absorber size can be significantly reduced. Although in this paper no direct comparison of techno-economic performance has been made for a column system, the cost of the absorber and the MBG have a small effect compared to other process equipment in this process. The heat exchanger network has a much bigger impact on the fixed capital investment of the process.

Table 6
Capital cost of the process equipment.

Source of cost estimate	Name in Figure 3 (Process Diagram)	Equipment cost for CS [USD]	Material factor ($f_{material}$) 316 SS	Purchased cost of equipment (316SS), [2023 EUR]	% of Total
Towler and Sinnott (2022)	CO2 absorber + MBG	99,457	1.3	161,325	1.6
Aspen	Condensate cooler, E7	10,300	1.3	16,537	0.2
Aspen	Regenerated liquid pump, P3	83,000	1.3	133,258	1.3
Aspen	Feed gas cooler, E1, 2nd part 65–45 °C	35,700	1.3	57,317	0.6
Aspen	Condensate pump, P2	5300	1.3	8.509	0.1
Aspen	Regeneration heater, E4	241,146	1.3	387,164	3.9
Towler and Sinnott (2022)	Regenerator	193,007	1.3	240,821	2.4
Own estimate	Liquid/gas separator, S2	99,457	1.3	161,325	1.6
Aspen	Product gas cooler, E5	65,700	1.3	105,482	1.17
Aspen	Regeneration feed- product heat exchanger, E2	3082,600	1.3	4949,167	49.3
Aspen	Regenerated product cooler, E6	112,443	1.3	1461,753	18.0
Vendor Atlas Copco	Product gas compressor, C2			500,000	5.0
Aspen	Feed gas cooler, E1, 1st part 77.9–65 °C	15,100	1.3	24,243	0.2
Aspen	Product gas precooler, E3	297,300	1.3	477,320	4.8
Aspen	Product gas condensate separator, S3	25,100	1.3	40,298	0.4
Own Estimate	Condensate compressor, C3			68,161	0.7
Aspen	Water separator, S1	36,700	1.3	58,922	0.6
Aspen	Product separator, S4	25,100	1.3	40,298	0.4
Vendor est. Sulzer	Fluid recycle pump, P1			300,000	3.0
Vendor est. Atlas Copco	Feed gas compressor, C1			500,000	5.0
			Total purchased equipment cost (M EUR)	10.0	100
			Fixed capital investment (M EUR)	47.6	

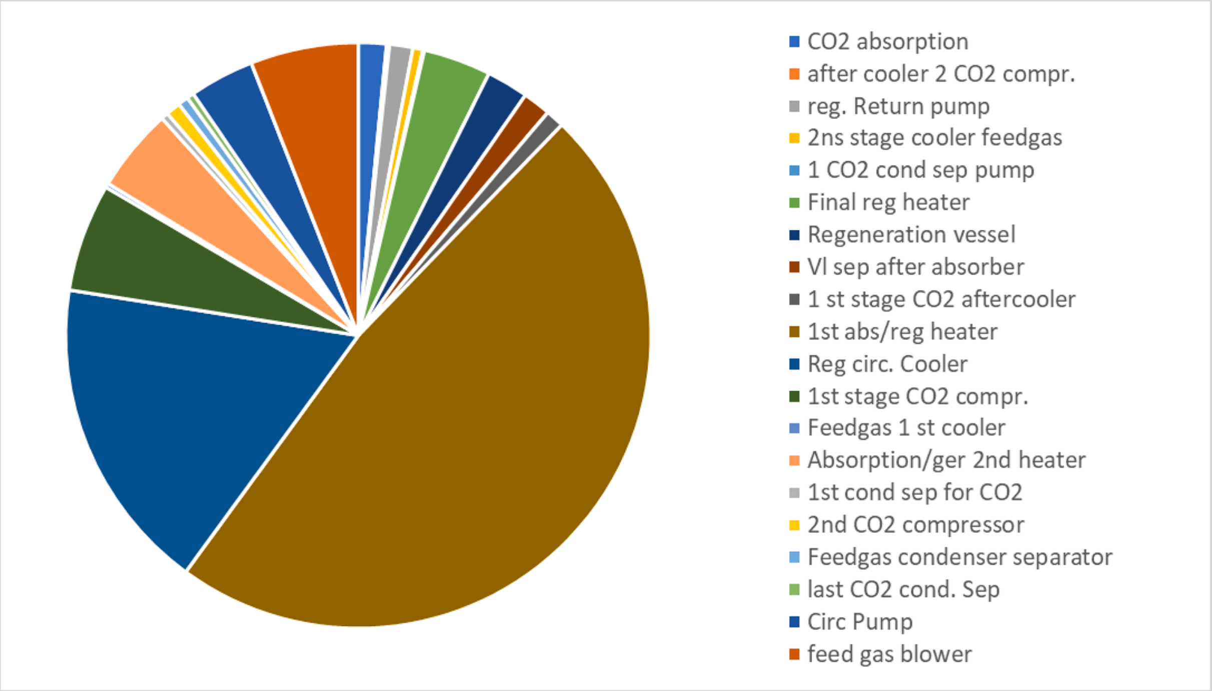

Fig. 5. The share of each equipment of total purchased investment cost.

5.2.2. CO_2 capture cost and sensitivity analysis

The CO_2 capture cost breakdowns are shown in Fig. 6 with two different full load hours and capacities. With the assumption of 12.1 tCO_2/h and 6500 h/a, representing typical full load hours for combined heat and power plants, the capture cost is EUR 133/tonne of CO_2. Even at this relatively high utilisation rate, the process is still very capital-intensive, with CAPEX accounting for more than half of the costs. The CAPEX originates mainly from the CO_2 capture process; the contribution of the heat pump is only 20 % of the total CAPEX. The share of variable costs (electricity) is only ~20 % of the total capture costs. The electricity consumption of the heat pump corresponds to ~60 % of the total electricity consumption. With annual full load hours of 8000 h/a, the

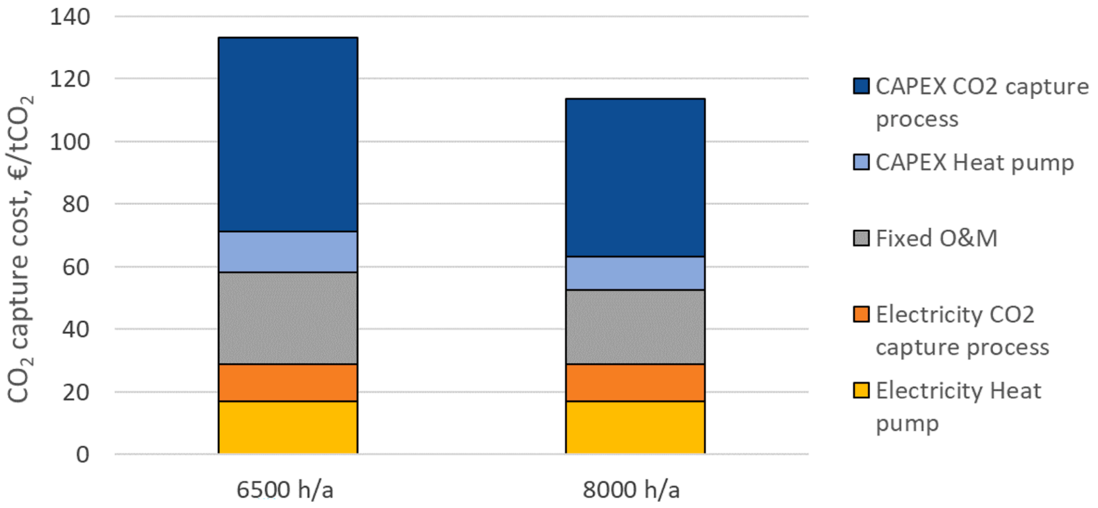

Fig. 6. CO_2 capture cost with annual full load hours of 6500 and 8000 h/a.

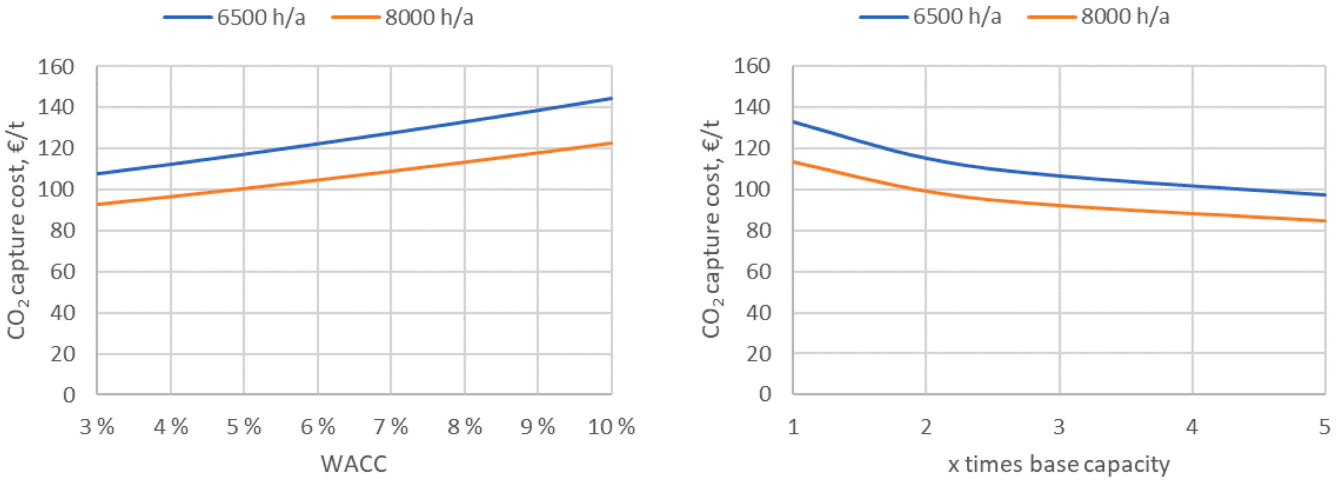

Fig. 7. The effects of WACC (left) and capacity (right) on the capture costs with two different full load utilisation hours.

capture cost decreases to EUR 114/tonne of CO_2. A utilisation rate of this range could be achieved if CO_2 were captured from a waste-to-energy plant or a pulp mill, for example.

The effects of plant capacity and WACC on capture costs with both full load hour assumptions are illustrated in Fig. 7. The capture costs for a plant with five times the base capacity would decrease to EUR 85/ tCO_2.

In Fig. 8, the impacts of heat and electricity costs on the CO_2 capture costs are illustrated. In the base configuration, in which the heat for regeneration was assumed to be produced using a heat pump, the cost of heat does not affect the overall cost. However, it might be feasible to provide the heat externally, in case there is low-cost or waste heat available. In this case, the investment in a heat pump could be avoided. For example, in pulp mills there is typically an excess of low temperature heat in the <70–80 °C temperature range ((Alfa Laval, n.d.; Axelsson et al., 2006; Bengtsson et al., 2002; Klugman, 2008). Also, if the captured CO_2 were to be converted to methanol, for example, there would typically be some waste heat available from the exothermic CO_2 hydrogenation reactions (Karjunen et al., 2021).

It can be observed that with an electricity cost of EUR 30–60/MWh, the break-even heat price, below which using a heat pump results in a higher CO_2 capture cost, is EUR 13–20/MWh. For the base case with an electricity price of EUR 45/MWh (not shown in Fig. 8), the corresponding break-even heat price would be around EUR 16/MWh. Should heat be available at lower cost than the break-even price, the utilisation of external heat would be more profitable than using a heat pump. If heat could be considered free of charge, the capture cost would decrease by EUR 30–40 /tCO_2 compared to the concept with a heat pump, assuming electricity prices of EUR 30–60 /MWh.

6. Discussion

The calculated CO_2 capture costs for the novel sodium carbonate-based capture process are higher than those reported for amine absorption. For amines, the reported capture costs vary typically between EUR 40–80/tonne of CO_2 (e.g. (Garðarsdóttir et al., 2018; Kearns et al., 2021; Onarheim et al., 2017; Panja et al., 2022; Zarei et al., 2023). However, care should be taken when comparing the reported costs between different sources as the assumptions regarding utility costs, for example, can vary a lot. Even more importantly, the other CO_2 capture technologies are also quite CAPEX-intensive and thus the considered capacity, full load hours and the assumptions regarding the lifetime of the plant and WACC will affect the results markedly. (Kearns et al., 2021) indicates a cost range of around USD 55–70/tonne for CO_2 capture from different sizes of biomass-fired power plants with similar assumptions to our study. The high-end value represents the cost for a similar capacity (130 kt/a) as that studied here (105 kt/a). They assume a 90 % utilisation rate (corresponding to annual full load hours of 7884

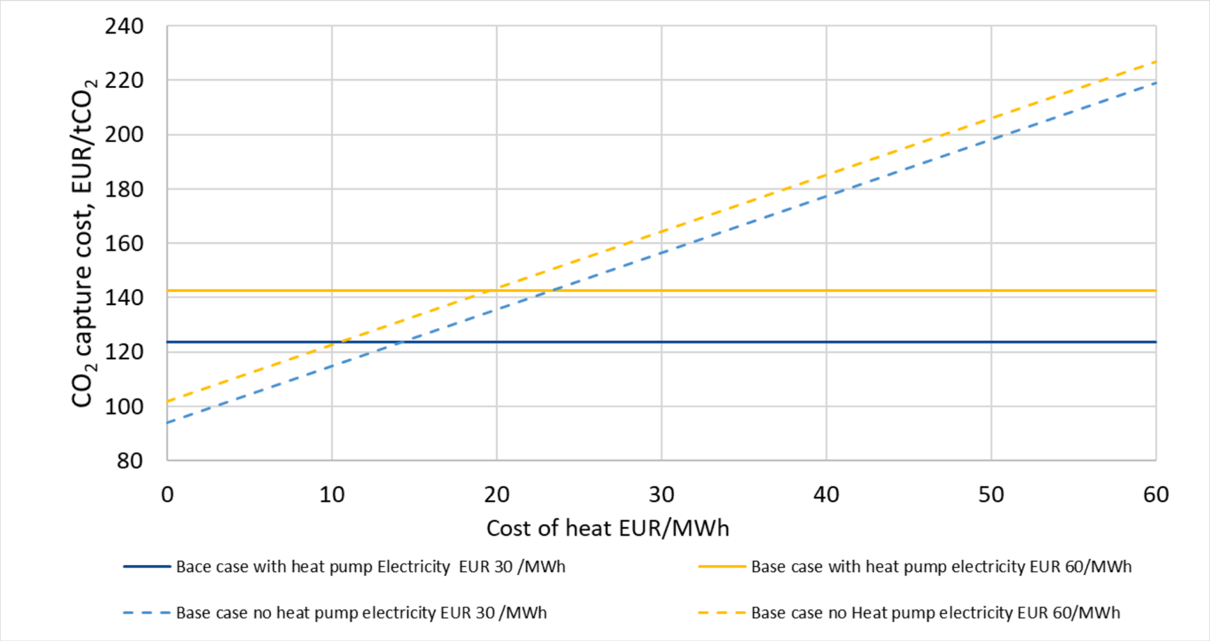

Fig. 8. CO_2 capture cost for the process with and without heat pump as a function of cost of heat with different electricity prices. The effect of a reduction of 50 % in CAPEX cost is also illustrated.

h), 8 % WACC, and a lifetime of 30 years. With the same assumptions, the capture costs for the novel process studied here would be around EUR 87/tonne of CO_2 when a heat pump is used as the heat source. Here CO2 product is obtained at 1 bar but compressing the CO2 to supercritical state > 150 bar requires approximately 100 kWh electricity per tonne of CO2, involving a cost of approximately EUR 10/t for compression and drying (Hannula and Melin, 2021) . The estimated costs are recalculated with the capacity and economic parameters used in this study.

The identified main drawbacks of the studied novel CO_2 capture process are the huge liquid flows required for absorption and regeneration. For absorption, the required liquid flow is dictated by the microbubble nozzle, which requires a high L/G ratio to achieve efficient mass transfer. For regeneration, the reason for the high required liquid flow is the low partial pressure of CO_2 in absorption, leading to a solution with a relatively low $NaHCO_3/Na_2CO_3$ ratio. Although the regeneration flow was only ~8 % of the absorption flow, it still led to enormous heat transfer requirements, causing high CAPEX. In addition, it led to high energy consumption in regeneration, as more water was evaporated than would have been with a higher $NaHCO_3/Na_2CO_3$ ratio.

The possible means to overcome these drawbacks are discussed next.

6.1. *Absorption at higher temperature*

In the base case, temperature is lower in the absorption (35 °C) than in the regeneration (65 °C). This leads to an extensive heat transfer need between the two stages. If absorption and regeneration could be operated at the same temperature, the need to transfer heat would decrease dramatically. The capital cost of the heat exchanger E2, which transfers 104 MW of heat between the absorption outlet and regeneration inlet streams, represents almost 50 % of the capital cost of the process, which equals EUR 31/t_{CO2} of the CO_2 capture cost. However, one disadvantage is that the absorption rate of CO_2 could decrease as described by (Spigarelli, 2013). When the absorption and regeneration temperatures are set as equal in the used simulation model, the results show that the big expensive heat exchanger E2 is not needed, but still a significant amount of heat is needed as an input to the process. As in the base case, this heat could be supplied by a heat pump.

Although this process alternative seems to be more optimal than the base case in terms of investment cost, we have not yet confirmed a high capture rate for these conditions in earlier work, due to limitations to the experimental setup to use high absorption temperatures. On the other hand, higher absorption temperatures have been used for sodium carbonate by (Knuutila et al., 2009) and others. Furthermore, using a higher absorption temperature is a common practice in potassium carbonate processes, such as the hot carbonate process. However, in those cases absorption is carried out at elevated pressure to increase the partial pressure of CO_2 and thus also the absorption rate (Borhani, et al., 2015).

6.2. *Higher pressure*

We investigated the effect of pressure by increasing the outlet pressure of the feed gas compressor from 1.5 bar_a to 10 bar_a. For this analysis, a two-stage compressor was used. The duty of the feed gas compressor (C1) increased from ~1 MW to approximately 5.4 MW and the duty of the fluid recycle pump (P1) decreased from 776 kW to 116 kW. On the other hand, the duty of the regenerated liquid pump (P3) increased significantly from 86 kW to 1196 kW. The higher pressure significantly reduces the gas volume. If the CO_2-depleted flue gas were to be heated with the compressor intercooler to 120 °C and expanded to 1 bar(a), almost 1.9 MW of electric power could be recovered. Consequently, the amount of the liquid circulated also reduces, as we have the fixed volumetric L/G ratio. The smaller amount of liquid reduces the size of the absorber, the fluid recycle pump, and so on. On the other hand, the pressurised equipment is more expensive per volume. However, pressurised operation has a very beneficial effect on regeneration. The higher partial pressure of CO_2 in absorption leads to a more saturated solution, having a higher $NaHCO_3/Na_2CO_3$ ratio dictated by the equilibrium behaviour. In the regeneration step, there is an equilibrium between the solution and gas phase. The partial pressure of CO_2 determines the ratio of CO_2 and water in the gas phase, i.e. the lower the partial pressure of CO_2, the more water vapour is evaporated, which increases energy consumption. Thus, with the solution with a high proportion of $NaHCO_3$, less water is evaporated. This would also decrease the size and power consumption of the heat pump. Furthermore, the absorption rate of CO_2 is also proportional to the partial

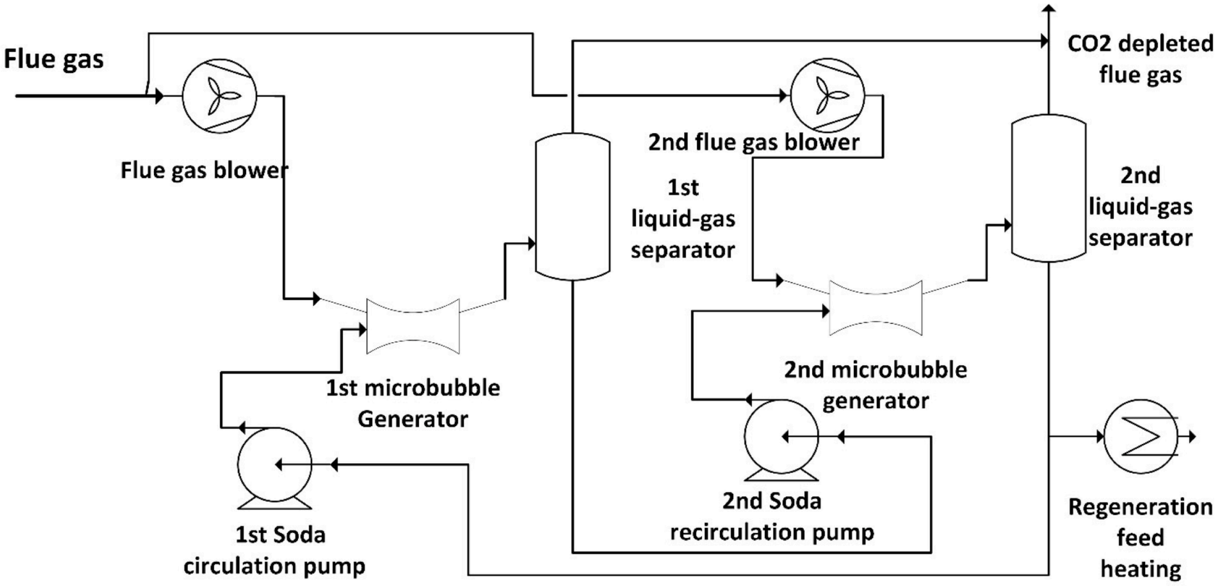

Fig. 9. Schematic diagram illustrating the process with several parallel stages.

pressure of CO_2 in the gas phase. Thus, a combination of higher absorption temperature and pressure could possibly improve the feasibility of the process significantly.

6.3. *Several parallel stages*

One way to reduce liquid circulation would be to divide the gas between parallel stages so that the soda liquid flows through each stage (see Fig. 9). This would enable lower liquid flow while keeping the same L/G ratio, as the gas flow per stage is less. The pressure of the liquid would need to be increased after each stage to compensate the pressure loss, but this would have only a minor effect on costs. In this concept, the stages could also be operated at different pressures to reduce the electricity consumption of compression but still produce a more saturated absorbent. For example, absorption could be conducted first at low pressure, e.g., 1.5 bar_a, for part of the gas. Next, the absorbent could be fed to the following stages at higher pressures. After the last stage the absorbent could be fed to the regeneration and the first stage could be performed with solution from the regeneration.

7. Conclusions

An enhanced post-capture process based on a sodium carbonate solution was evaluated for industrial-scale implementation (105 kt of CO_2 capture/a) for typical flue gas from a wood-fired power plant. A flowsheet of the process was developed, and a simulation model was built in Aspen Plus. Consequently, based on the estimated mass and energy balances and cost data, techno-economic analysis was performed, and the most significant cost factors were determined.

Capture cost in the unoptimised base case, based on direct scale-up from previous experimental work, was EUR 114–133/t, which is higher than the costs reported for amine absorption. The high costs were mainly due to the huge liquid flows required, necessitated by using a low salt concentration (<8 wt-%) to prevent precipitation. This led to very high heat transfer requirements and energy demands, but the high cost is mainly due to non-optimal heat integration. Increasing the absorption temperature would significantly reduce the heat transfer needs and thus the CAPEX of the process. The heat consumption could be reduced by increasing the absorption pressure.

One of the benefits of the process is that regeneration can be conducted at low temperature (~65 °C) which could enable the use of waste heat. If heat could be considered free of charge, the capture cost was found to decrease by EUR 30–40/tCO_2 compared to supplying heat with a heat pump. It was found that using a heat pump was optimal when the price of heat was more than EUR 13–20/MWh, depending on the electricity price.

Due to the improved absorption rate, the absorber cost was only a small part (<2 %) of the investment cost. According to the simulations the CO_2 product stream was also relatively pure, containing <0.1 vol-% N_2 and O_2, and 6 vol-% water which can be removed by conventional gas drying systems.

CRediT authorship contribution statement

Kristian Melin: Writing – review & editing, Writing – original draft, Visualization, Validation, Supervision, Software, Resources, Project administration, Methodology, Investigation, Funding acquisition, Formal analysis, Data curation, Conceptualization. **Markus Hurskainen:** Writing – review & editing, Writing – original draft, Validation, Investigation, Formal analysis, Data curation, Conceptualization. **Miia Nevander:** Writing – review & editing, Writing – original draft, Validation, Project administration, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Tuula Kajolinna:** Writing – review & editing, Data curation.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.ijggc.2025.104310.

Data availability

we include electronic support file in submission

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