Submitted for publication

Techno-economic evaluation of integrated NO_x and SO_x removal in pressurized flue gas systems for carbon capture applications

Sima Ajdari [a], Stefanìa Òsk Gardarsdòttir [b], Fredrik Normann [a], Klas Andersson [a]

[a] Division of Energy Technology, Department of Space, Earth and Environment, Chalmers University of Technology, Gothenburg, 41296, Sweden
[b] SINTEF Energy Research, Gas Technology, Trondheim, Norway

Abstract

This study evaluates the technical and economic performances of an integrated NO_x and SO_x removal unit in pressurized flue gas systems that are applicable to oxy-fuel and chemical looping combustion plants. The cost is estimated using a bottom-up and discounted cash-flow approach based on a flowsheet model that includes a detailed description of the pH-dependent gas- and liquid-phase interactions of nitrogen and sulfur. The results show that the proposed integrated removal process is able to achieve emission control efficiencies comparable to the best-available techniques, albeit at about one-third of the cost for a system involving flue gas desulfurization plus selective catalytic reduction.

1. Introduction

Systems for simultaneous control of nitrogen oxides (NO_x) and sulfur oxides (SO_x) emissions from power and industrial sources have attracted much attention since the implementation of public legislation of emission control standards in the 1970s. Reduced footprint and investment costs, currently comparable to those for conventional wet flue gas desulfurization (WFGD) and selective catalytic reduction (SCR), have provided the motivation for developing processes for multi-pollutant control (MPC), with absorption being one alternative for MPC.

Sulfur dioxide (SO_2) and nitric oxide (NO) are the main SO_x and NO_x species formed during combustion. SO_2 has a high solubility, and the low solubility of NO may be overcome by oxidizing it to nitrogen dioxide (NO_2) using an oxidizing agent in the gas phase (e.g., chlorine dioxide or ozone)[1-2] or in the liquid phase (e.g., sodium chlorite or hydrogen peroxide)[3-4]. However, MPC technologies still need to achieve commercial breakthrough, mainly due to their complexity, consumption of chemicals, and difficulties associated with disposal of the waste streams.

Along with the development of carbon capture and storage (CCS) technologies, and in particular oxy-fuel combustion and chemical looping combustion (CLC), there is renewed interest in the absorption-based removal of acid gases. In CCS systems, the conditioning of the carbon dioxide (CO_2) before transportation and storage requires cooling and compression, as well as the removal of NO_x and SO_x species. Although the levels of NO_x and SO_2 permitted in the product CO_2 are not defined, issues related to corrosion owing to acid formation in the compressor section have highlighted the importance of acid gas control during CO_2 conditioning. The increase in pressure, brought about by compression, inherently increases the rate of NO oxidation to NO_2, due to the increased partial pressures of the reactants. This in turn enables absorption-based MPC of NO_x and SO_x without the use of an oxidizing agent. A series of process designs based on the concept of NO_x and SO_x absorption has been proposed[5-7]. These processes are based on the oxidation of NO to NO_2 by the oxygen present in the flue gas and the subsequent absorption of NO_2 either with simultaneous removal of SO_2[5] or downstream of the SO_2 absorption[7]. These concepts have been tested in pilot studies and have been reported to achieve >90% removal of NO_x and SO_2[8]. However, a detailed techno-economic evaluation of these processes relative to conventional emission control systems is lacking in the literature.

The techno-economic evaluations of the oxy-fuel combustion process available in the literature (see references[9-10]) are based on first-generation power plants that apply conventional flue gas technologies that have been developed for air-fired power plants (i.e., WFGD and primary NO_x control, sometimes in combination with SCR). Although the aforementioned studies provide valuable information regarding the performance and cost of CO_2 conditioning, they risk being too conservative in outlook or overlooking important characteristics of the complex chemistry occurring in oxy-fuel/CLC plants. The present work combines a detailed process assessment tool[11] with a detailed cost analysis to estimate the costs of an integrated system for NO_x and SO_x control that is suitable for oxy-fuel/CLC plants. Plant performance is evaluated with respect to both removal efficiencies and wastewater composition. The design and performance indicators for each concept form the basis for the estimations of the capital and operating costs for the removal processes. The performances of integrated NO_x and SO_x removal processes are compared to those of conventional flue gas-cleaning techniques.

2. Methods

Techno-economic evaluations of NO_x and SO_x removal during CO_2 conditioning in oxy-fuel and CLC plants are performed for two design concepts under various set conditions (cases). Figure 1 gives schematic overviews of the oxy-fuel and CLC plants and the two concepts considered here. The main focus of this study is on the removal of NO_x and SO_2 in connection with CO_2 conditioning in the gas processing unit (GPU) indicated in Figure 1a. The main difference between the CO_2 conditioning of CLC and oxy-fuel flue gases is that the CLC flue gas requires a post-oxidation reactor (POR) for oxygen polishing downstream of the fuel reactor, to ensure complete fuel conversion[12]. The reference design concept (Concept A) of this study involves single-column, pressurized removal of SO_2 and NO_x or only NO_x (in the absence of SO_2 in the flue gas). An alternative design (Concept B), which entails a dual column with atmospheric removal of SO_2 and pressurized removal of NO_x, is also investigated. The technical evaluation is performed through a process simulation of the flue gas compression and NO_x and SO_2 removal concepts for the different cases. The cases include various flue gas compositions (in terms of the S/N ratio) to cover a range of fuels (with different sulfur contents), combustion technologies, and/or extents of upstream flue gas treatment. Table 1 shows the gas compositions and properties of the flue gases at the inlet of the GPU for the cases investigated. The flue gas is assumed to contain 4% O_2. However, two cases with lower levels (1% O_2) of excess oxygen are also included. Low levels of excess oxygen are beneficial in terms of the process economics, and extremely low levels of excess O_2 (down to a stoichiometric ratio of 1.05 or 2,000 ppm O_2 in the flue gas) have been proposed in particular for CLC process[12]. However, such low concentrations of O_2 are deemed not to be feasible with respect to fuel burnout or not relevant to the proposed process, as they will not suffice for the oxidation of NO to NO_2.

The economic evaluation involves estimations of the capital and operating costs for each case. Capital costs are calculated based on a bottom-up approach, which entails breaking down the GPU into basic items of equipment and adding installation and indirect costs. The sizing of each item of equipment is derived from the process optimization simulations for each case to achieve the required removal rates. The removal rates are set so as to represent those commonly achieved using the "Best Available Techniques"[13], i.e., SCR and WFGD. This implies 90% removal of NO_x and 95% removal of SO_2[13-14]. The equipment cost and installation cost for each item of equipment are estimated using the Aspen Process Economic Analyzer software. Finally, the derived costs are discussed relative to those for conventional flue gas cleaning.

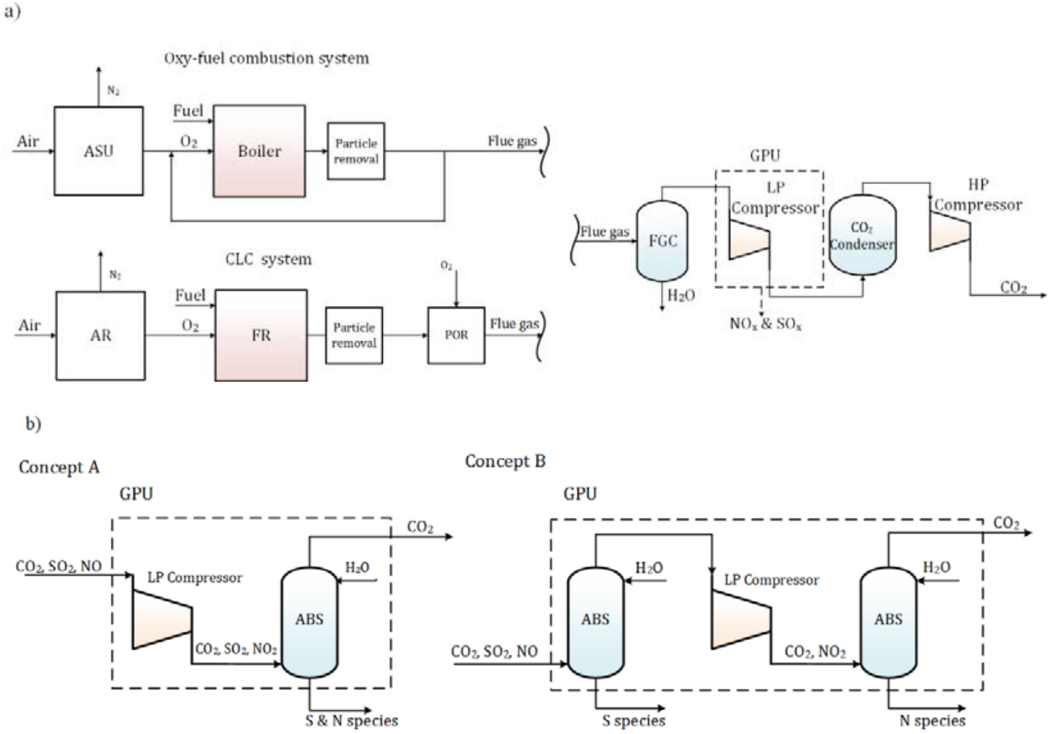

Figure 1. Schematics of: a) the oxy-fuel and CLC plant, including the combustion system and CO_2 conditioning; and b) the options for integrated control of NO_x and SO_2 emissions included in the techno-economic evaluation. Concept A: Single-column pressurized absorption of NO_x and SO_2. Concept B: Dual-column with atmospheric SO_2 absorption and pressurized NO_2 absorption. ABS, Absorber; ASU, air separation unit; AR, air reactor; FR, fuel reactor; POR, post-oxidation reactor; LP Compressor, low-pressure compressor; HP Compressor, high-pressure Compressor.

Table 1. Compositions and properties of the gases at the GPU inlet for the investigated cases.

	Case 1	Case 2	Case 3	Case 4	Case 5	Case 6	Case 7
Technology	Oxy-fuel	Oxy-fuel	Oxy-fuel	Oxy-fuel	Oxy-fuel	CLC	CLC
Example Fuel	Coal	Coal	Coal	Biomass	Coal	Coal	Biomass
Flue gas Composition							
CO_2 (%)	87	87	87	87	87	92	92
O_2 (%)	4	4	4	4	4	1	1
SO_2 (ppm)	1,000	400	2,000	0	1,000	400	0
NO_x * (ppm)	440	440	440	440	440	440	440
N_2	Balance	Balance	Balance	Balance	Balance	-	-
H_2O(g) (%)	4	4	4	4	4	4	4
Removal technology							
Design	Concept A	Concept A	Concept A	Concept A	Concept B	Concept A	Concept A
Chemical(s)	NaOH	NaOH	NaOH	NaOH	NaOH	NaOH	NaOH, Na_2SO_3

* Mole $_{NO}$/mole $_{NO2}$=10

2.1. Designs for the concepts studied

Figure 2 shows a schematic of the GPUs designed for the two concepts. The multistage, low-pressure compressor (indicated as LPC in Figure 2), which includes three-stage compression (CMP 1–3) with intercooling (HX 1–3) to 30 bar, is the same in both concepts. The condensed liquids from the intercoolers are separated in the knock-out drums (KO 1–3), and collected after depressurization in flash drums (D 1–3). The condensate from the flash drums is sent to a holding tank (T-3) prior to water treatment (not considered in this study).

In Concept A (Figure 2a), the gas from the LP compressor enters the pressurized-removal block (PRB in Figure 2). Initially, there is a pressurized gas reactor (R-1), which increases the rate of NO to NO_2 conversion to >90%. The flue gas is then introduced into the packed column (ABS-1). The solvent comprises a mixture of recycled absorbing solution, makeup water, and sodium hydroxide (NaOH). In cases with low or no concentrations of inlet SO_2 (i.e., Case 7), sodium sulfite (Na_2SO_3) may be added to increase the concentration of sulfur in the solution, thereby improving the absorption of NO_2.

For Concept B (Figure 2b), the flue gas is introduced into the atmospheric-removal block (ARB in Figure 2b) before entering the LPC and the PRB. In the ARB, most of the SO_2 and some of the NO_2 are removed from the flue gas through contact with a mixture that contains recycled absorbing solution, makeup water, and sodium hydroxide (NaOH), in a packed column (ABS-1). The LPC and PRB (wherein most of the NO_x is removed) are similar to those used for Concept A. The general operating conditions and design parameters for the concepts are presented in Table 2. Detailed equipment lists and operating conditions for all cases are presented in Tables A1–A7 in the Appendix.

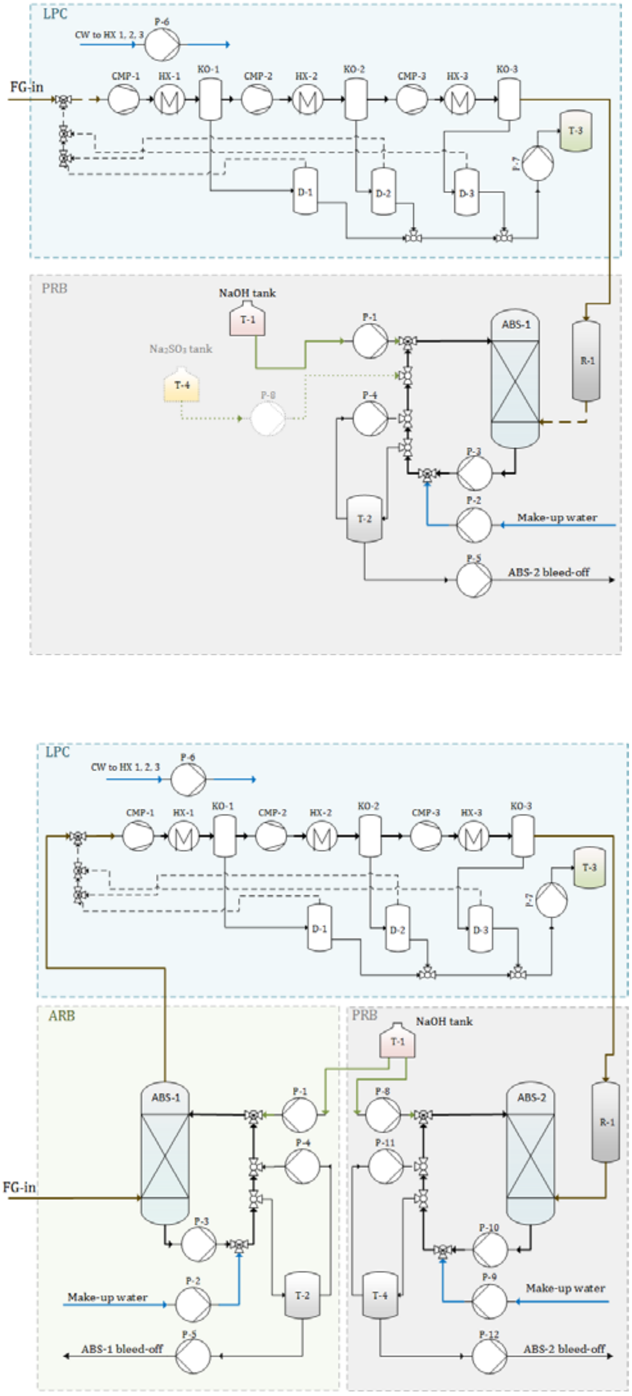

Figure 2. Schematics of the GPUs for: a) Concept A; and b) Concept B. ABS, Absorber; CMP, compressor; CW, cooling water; D, flash drum; FG, flue gas; HX, intercooler; KO, knock-out drum; P, pump; R, reactor; T, tank; LPC, low-pressure compressor; ARB, atmospheric-removal block; PRB, pressurized-removal block.

Table 2. General operating conditions and design parameters. (The abbreviations in parentheses are those used in Figure 2)

Columns (ABS)		LP Compressors (CMP)	
Column packing	Mellapak 250Y	Isentropic efficiency	0.88
SO_2 removal efficiency	>98%	Mechanical efficiency	0.9
NO_x removal efficiency	90%	Compression stages	3
		Discharge pressure	30 bar
Pumps (P)		Intercooling temperature	30°C
Isentropic efficiency	0.75	Discharge temperature	30°C
Cooling water temperature	10°C	Intercoolers (HX)	
		Type: Shell and tube	
		flue gas in tubes	
Flue gas		Pressure drop	3% of inter-stage pressure
Mass flow	110 kg/sec		
NO/NO_2 ratio	10		
Temperature to GPU	30°C		

2.2. Process modeling

Two modeling frameworks are used for the process simulations. For the sizing of the equipment and evaluation of the performance of the GPU for each case, the process simulation software Aspen Plus ver. 8.8 was used. The process model is discussed in detail elsewhere[11]. To predict the composition of the wastewater emanating from the process, a detailed chemistry model developed in Matlab[15] was used. Compared to the reaction mechanism applied in the absorption column, this mechanism may be applied over a broader pH-range and for longer liquid residence times. This is an important feature, as the residence times in the liquid handling system are in the order of hours (the liquid residence times in columns are in the order of seconds to minutes).

The process model of the GPU considers the reactions and condensation in the intercoolers, in addition to the mass transfer and reaction kinetics in the absorber. The chemistry in the absorber is based on the previously developed reduced mechanism[16]. The condensates and absorber bleed-off collected in the intermediate storage tanks (indicated as T in Figure 2) will contain several nitrogen and sulfur species, and the chemistry of these mixtures are of the utmost importance for the process. In the storage tanks, the liquid phase is assumed to be perfectly mixed. The tanks are sealed (i.e., the gas phase is not open to the atmosphere), interactions with the gas phase are limited, and the mass transfer description is simplified in this model. For more details regarding the model, see our previous publication[15].

2.3. Cost estimation

Capital costs are estimated using a bottom-up and discounted cash-flow approach[17]. The cost estimate for the GPU is performed for the nth-of-a-kind plant, i.e., for a commercial plant built after successful demonstration of the technology, and on a pre-taxation basis. The capital costs are estimated as so-called "total plant costs". The main calculation steps include an estimation of the equipment costs followed by the addition of installation costs, such as those for piping, erection, instrumentation etc. Total direct costs (TDC) result from adding process contingencies to the equipment and installation costs. The total direct costs plus indirect costs, owner's costs, and project contingencies generate the total plant costs.

The main assumptions made in the economic analysis are summarized in Table 3. Contingency factors for the GPU are derived based on the maturity of the technology, in line with the AACE guidelines for process contingencies[18]. Since related processes have been demonstrated on a small-pilot-plant scale[19-20], and to account for uncertainties in the level of detail of the equipment list, the process contingencies are set at 35% of the sum of the equipment and installation costs. The indirect costs include cost elements such as yard improvements, service facilities, and engineering and consultancy costs, as well as building and sundries [17]. Owner's costs and project contingencies for the nth-of-a-kind cost estimates are chosen according to the AACE guidelines for cost estimates.

Material selection is important in relation to the equipment cost. Stainless steel 304L is the default material for the process equipment. The components that operate under more-severe conditions, i.e., the absorber(s), the intercoolers in the compression train, and the buffer tank (T-2 and T-4 in Figure 2), are constructed from duplex-grade stainless steel 2205.

The fixed operating costs include maintenance, insurance, and labor costs and are based on assumptions related to material replacement and a factor-based approach[17], as listed in Table . The variable operating costs for the GPU include the costs for utilities and chemicals and are based on the energy and material balances derived from the process simulations. The unit costs of the required utilities and chemicals are also listed in Table 3. The costs are calculated for a flue gas corresponding to 350MW$_e$ with an electrical efficiency of 39%.

Table 3. Main assumptions made in the economic analysis.

Cost basis	€2015
Operational lifetime	25 years
Construction time	1 year
Discounted cash flow rate	8%
Tax rate	0%
Inflation rate	0%
Annual operating hours	8,000
Process contingencies	35% of total direct costs without process contingencies
Indirect costs	14% of TDC*
Owner´s costs	7% of TDC
Project contingencies	15% of TDC
Insurance and location tax	2% of TDC
Maintenance cost	2.5% of TDC
Operative labor salary	60 k€/year
Operative labor, number of persons	5
Electricity	58.1 €/MWh
Cooling water	0.39 €/m^3
Process water	6.65 €/m^3
NaOH 50/50 wt% solution	370 €/tonne
Na_2SO_3	

*TDC, Total Direct Costs.

The reference cost for NO$_x$ reduction is estimated using the method described previously[21] for an aqueous ammonia-based SCR system. The method is based on regression to cost data available from vendors. Here, it is assumed that the SCR reactor is placed downstream of the flue gas recycle and dust removal (low-dust) units. This means that the volume of the SCR reactor may be smaller than an air combustion unit of similar power, owing to the removal of N_2 from the flue gas. The temperature into the SCR reactor is assumed to be 300°C, applying the high-temperature dust removal system recommended previously[22]. The methods described in the literature for calculation of an SCR unit are based on the heat flow in the boiler of a conventional, coal-fired plant[23]. As the reduced flue gas flow rate of the oxy-fuel/CLC plant has a significant impact on the cost of the SCR unit[22], the calculated cost is scaled using a general scaling factor (0.6) to the flue gas flow rate of the oxy-fuel plant. Thus, the design of an SCR system will differ from the designs of conventional power plants, and the method mentioned above gives an order-of-magnitude estimation of the SCR unit, so as to provide a basis for discussions and comparisons with the absorption-based system. The estimated total investment cost of the SCR unit includes the cost of the SCR reactor, reagent preparation cost, and piping and auxiliary power costs. The operating costs include the costs for ammonia, electricity, catalyst replacement, and annual maintenance. The assumptions made for the cost estimation are summarized in Table 6.

Table 6. Main assumptions made for the estimation of the reference cost for NO_x reduction by SCR.

Parameter	Value	Reference
Fuel input	815 MW	
Flue gas inlet temperature	300°C	
NO_x removal efficiency	90%	
Stoichiometric ratio	1.05	[23]
Reagent	30 wt% NH_3	[23]
Reagent cost	400 €/tonne	[24]
Process contingencies	5%	[23]

The reference cost for SO_x reduction is estimated using the method described previously[25] for a wet limestone process with forced oxidation. In a conventional power plant, air is added to the FGD unit to support oxidation of the hydrogen sulfite. However, in an oxy-fuel/CLC plant, the addition of air results in undesirable dilution of the CO_2. Thus, oxygen from the ASU is used instead of air, entailing an additional cost of oxygen production[26]. The capital cost of the FGD system includes the costs for the reagent feed system (receiving, storage, and grinding), SO_2 removal (absorbers, tanks, pumps), and waste/byproduct handling, in addition to the annual costs for maintenance, labor, reagents, and electricity. The cost of the absorber is mainly determined by the volume flow of flue gas and, thus, is lower for oxy-fuel conditions. Table 7 summarizes the main assumptions made for the cost estimation of FGD for SO_x removal. The flue gas conditions are taken from Case 1.

Table 7. Main assumption for the estimation of the reference cost for SO_x removal by FGD.

Parameter	Value	Reference
Removal efficiency	95%	[25]
L/G, l/m^3	17	[25]
Reagent	Limestone slurry	
Limestone purity	95.3%	[25]
Flue gas inlet temperature	180°C	
Flue gas outlet temperature	57°C	[25]
Process contingencies	5%	[25]

3. Results

The technical and economic performances of the investigated processes are presented in separate sections. As the uncertainties related to the disposal of the liquid effluent are high, disposal issues related to the composition of the liquid effluent are discussed only in the technical performance section. The economic performance includes estimations of the required investments, as well as the annual costs, for the GPU.

3.1. Technical performance of the GPU

The primary criteria set for the technical design are >95% removal of SO_2 and >90% removal of NO_x. These criteria are met for all the investigated cases. NO_x removal is dependent upon the amount of NO oxidized to NO_2 and the NO_2 absorption rate in the absorber. An NO oxidation rate of around 40% is achieved in the intercoolers, and the reactors ensure the residence time that is required to achieve >90% NO_x removal downstream of the absorber. The secondary objective is to minimize the amount of freshwater used in the absorption process, thereby limiting the amount of wastewater for treatment and disposal. The required amount of freshwater is related to the solvent pH, the residence time in the absorber (i.e., the packing height), and the amount of sulfite (e.g., Na_2SO_3) added to enhance NO_x removal. The pH in the absorber is maintained at 5–7, which ensures almost complete absorption of SO_2. The influence of the solvent pH was investigated in our previous papers[11, 15]. A pH level <5, in addition to reducing the amount absorbed per kg of solvent, will lead to the formation of undesired products in the liquid. A pH level >7 will give excessive rates of CO_2 absorption and chemical consumption. In the presence of sulfur, the desired removal efficiencies can be achieved using shorter columns. For example, Cases 1 and 2 with $SO_{2,\ \mathrm{inlet}}$ of 1,000 ppm and 400 ppm, respectively, have 90% and 77% less packing material than Case 4, which does not have any SO_2 in the flue gas. The effect of increasing the packing height is strongest for an NO_x-only system[11] (Case 4). Figure 3 shows the removal efficiencies achieved by varying the makeup flow rate (and consequently, the L/G ratio in the column) and the packing height while maintaining a pH level >5 for Case 4. Furthermore, in multi-control systems for SO_x and NO_x, the rate of NO_x absorption is generally the factor determining the packing height, as the absorption of SO_2 is rapid compared to the absorption of NO_2. In the dual-column design, the atmospheric column is designed for SO_2 absorption and may be considerably shorter than the other columns.

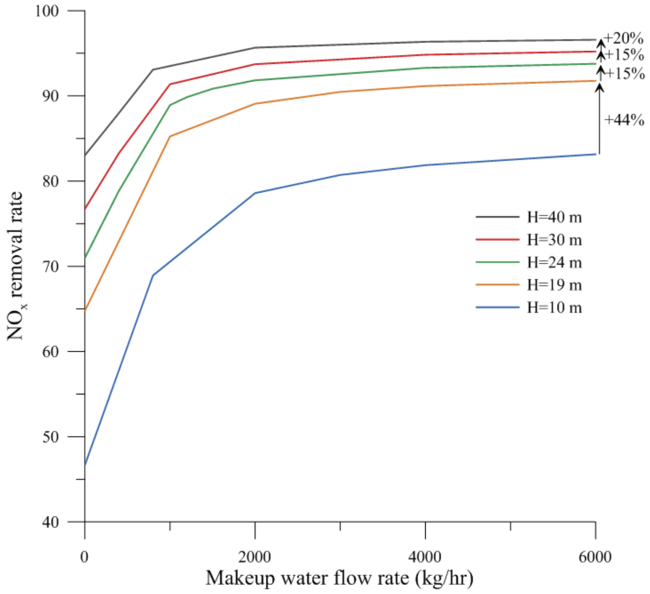

Figure 3. NO_x removal efficiency as a function of the flow rate of makeup water for various packing heights for Case 4. The relative increase in the installed cost for the absorber resulting from an increase in the packing height is shown on the right-hand side of the graph.

Wastewater may be a major issue for the proposed process. The availability and quality of freshwater are important factors, and the management of water and wastewater in heat and power generation plants is crucial. Power plants consume large volumes of water, and wastewater treatment is required so that all plants remain within the proscribed disposal limits. The plants typically aim to recycle water, and ideally, to achieve zero liquid disposal (ZLP). Depending on location, different (national) quality standards and operating permits for the discharge of wastewater are applicable. The proposed absorption systems for NO_x and SO_2, and the condensates from the compressor intercoolers in oxy-fuel and CLC plants will create new types of wastewater streams. This section discusses the volumes and quality levels of these wastewater streams, the potential issues and limitations associated with wastewater handling, and wastewater treatment options.

The key performance indicators (KPIs) for evaluating wastewater generation from the proposed process include the: pH of the solution (allowable range for disposal, 6–9); volumes of liquid (m^3/hr); and concentrations of nitrates/nitrites and sulfates/sulfites (mg/L). Table 4 lists the KPIs for the condensates after 0 hours (as received), 1 hour, and 10 hours in the condensate holding tank, for two inlet concentrations of SO_2 delivered to the compressors. The condensate from the intercoolers contains unstable products, such as nitrites and sulfites, which will react if allowed a residence time, for example in the condensate holding tank (see Figure 2). As the pH is <2, NO and N_2O will be emitted from the condensate according to Reactions R1 and R2.

$$2HNO_2 \rightarrow 2NO + HNO_3 + H_2O \qquad (R1)$$

$$2HNO_2 + 2HSO_3^- \rightarrow N_2O + 2HSO_4^- + H_2O \qquad (R2)$$

The liquid-phase chemistry is strongly pH-dependent, and gaseous emissions of NO and N_2O may be avoided by neutralizing the pH of the condensate. Regarding sulfur, the dissolved SO_2(aq) may desorb to the vapor phase depending on the pH of the solution, although it may be reabsorbed completely within 1 hour. Furthermore, the chemistry of the interactions between nitrites and sulfites may result in the formation of sulfates, N-S complexes, and even hydroxylamines. While the formation of sulfate is favorable in terms of the wastewater characteristics (as it is more stable), the two latter products may complicate the wastewater treatment process if present at high levels. As the interactions between nitrites and sulfites slow down at pH 7, immediate neutralization of the pH of the condensate may be beneficial for wastewater treatment. This might also make it possible to re-use the condensate for the absorption process, wherein the presence of sulfites enhances NO_2 removal.

Table 4. Characteristics of the condensate transported from the intercoolers into the condensate holding tank for cases in which there is: 1) both NO_x and SO_2 present during compression, $SO_{2\text{-in}}$ = 400 ppm (Case 1); and 2) only NO_x present during compression (Cases 4, 5, and 7).

	Case 1				Cases 4, 5 and 7		
Parameter	After 0 hr (As received)	After 1 hr	After 10 hr	After 10 hr (pH-controlled)	After 0 hr (As-received)	After 1 hr	After 10 hr
pH	2.0	1.8	1.8	7.0	2.3	2.2	2.2
Flow, m^3/hr	6.6				6.6		
Sulfite[1], mg/L	530	1	0	563 (no SO_2)	-	-	-
Nitrite, mg/L	343	38	34	334	343	224	158
Nitrate, mg/L	296	310	311	296	296	348	378
Sulfate, mg/L	0	536	623	0	-	-	
Other, mg/L	-	HADS[2]: 65 HAMS[3]<1 NH_2OH <1	HADS<1 HAMS<1 NH_2OH <1	HADS<10			
Gaseous emissions (as % total-N)	-	NO (4%), N_2O (23 %)	NO(4%), N_2O(24%)	-	-	NO (13%)	NO (20%)

[1]Including dissolved SO_2

[2]Hydroxylamine disulfonic acid

[3]Hydroxylamine monosulfonic acid

Table 5 shows the KPIs for the absorber bleed-off stream for all the cases and for WFGD wastewater as a reference[27]. The treatment of wastewater from the WFGD is considered one of the most challenging issues for conventional power plants, due to the complexity and variability of composition of the wastewater streams[27]. For WFGD processes, a limit is usually imposed on the disposal of wastewater that contains sulfates and sulfites. According to the European Commission[28], the limits are 1,300–2,000 mg/L for sulfates and 1–20 mg/L for sulfites (daily average values). In addition, disposal limits as low as 4.4 mg/L (as total nitrogen, daily average values) for nitrites/nitrates are applied in the USA for WFGD wastewater[29]. The amounts of nitrites/nitrates in the liquid are naturally the largest difference compared to WFGD. Moreover, the sulfites/sulfates and nitrites/nitrates ratios in the absorber bleed-off as-received are higher than commonly detected for WFGD. Given the instability of sulfites, it is desirable to oxidize them to sulfates. Residence time in combination with access to air is commonly used to enhance oxidation. As shown in Table 4, sulfates and nitrates dominate already after 1 hour in the liquid hold-up. Nitrogen- and sulfur-containing complexes, such as hydroxylamine disulfonic acid (HADS), may also form in the liquid. The presence of HADS in the wastewater at a significant concentration could necessitate additional treatment. Precipitation and thermal decomposition have been proposed as a possible treatment[30], although they require further investigation. Removal of SO_2 prior to compression may reduce the formation of nitrogen- and sulfur-containing complexes. However, as seen for Case 5, even the presence of a low level of NO_2 leads to the formation of HADS. It is likely that HADS is present also in WFGD systems[30].

Table 5. Characteristics of the absorber bleed-off for all the cases investigated (as-received).

Parameter	Case 1	Case 2	Case 3	Case 4	Case 5 ABS-1	Case 5 ABS-2	Case 6	Case 7
pH range	5.0–6.0	5.0–6.0	5.0–6.0	5.0–6.0	5.0–6.0	5.0–6.0	5.0–6.0	5.0–6.0
Volume flow, m^3/hr	1.1	1.0	2.3	1.3	2.0	1.3	1.1	2.3
%S-in FG as Sulfite	16.5%	2%	54%	-	90%	0	1%	-
%S-in FG as Sulfate	20%	50%	10%	-	2%	<1%	48%	-
%S-in FG as HADS	63%	48%	36%	-	7%	0	51%	-
%N-in FG as Nitrite	19%	69%	9%	46%	0	47%	66%	60%
%N-in FG as Nitrate	0	<1%	0	46%	0	44%	1%	22%
%N-in FG as HADS	72%	22%	82%	0	8%	0	23%	9%

In summary, the liquid waste stream generated from the proposed process will require treatment before being recycled within the plant or released to the environment. While new challenges will arise in regard to the treatment of nitrogen components, conventional wastewater treatment technologies should be applicable. Such technologies include:

- Effluent sump with pH control to promote conversion to stable components.
- Precipitation with calcium salts to remove sulfates
- Sedimentation, flotation or filtration to separate precipitates
- Ion exchange processes with resin beds (commonly the final polishing step for boiler feedwater) to remove the remaining ions, such as sodium, nitrates, and sulfates
- Physical, chemical or biological removal of nitrates/nitrites.

3.2.Economic performance of the GPU

The total plant costs for all the investigated cases are presented in Appendix B. As expected, the compressor is the major contributor to the total cost of the GPU. The total direct cost of the compressor is around 59 M€ for each of the integrated designs, and also for a design with a conventional emissions control system. Electricity consumed by the compressors makes up the largest fraction of the utility cost, and is similar for all the cases (14.0–14.6 M€/year). The compressor cost shows a small variation (<0.3%) depending on the presence of SO_2 during the compression process, which also slightly increases the cost of electricity (<0.5%). The same material is used in all the cases, i.e., stainless steel 304L for the compressor and duplex-grade stainless steel 2205 for the intercoolers, where acid formation and low pH values are expected. If all the acidic gases are removed prior to compression the use of stainless steel 304L in the intercoolers could be feasible. In that case, the total direct cost of the intercoolers, which account for around 5% of the total direct cost of LPC, would be reduced by around 15%.

Figure 4a presents the total direct costs (without process contingencies) for the NO_x and SO_x removal processes (excluding the compressor) in all the cases investigated. The dual-column design (Design B) requires a considerably higher investment than the single-column design (Design A). The total direct cost of Case 5 is almost 3-fold higher than that of Case 1, which has the same flue gas conditions. The flue gas composition exerts some influence on the total direct cost, which varies in the range of 3–5 M€ for Design A, depending on the case. Using the process solely for NO_x removal, e.g., in the case of a sulfur-free fuel (Cases 4 and 7), increases the required investment by 50%–90% relative to the cases with SO_2 in the flue gas (Case 1). In Case 4, the lack of sulfur is compensated for by a considerably taller absorption column, and in Case 7 by the addition of sulfite to the absorber. In Case 7, the cost of the tank required to hold the sulfite is considerable. Otherwise, no significant differences in total direct costs are observed for the various SO_2 concentrations in the flue gases, i.e., Cases 1-3, as it is mainly the control of NO_x that determines the dimensions of the units. The oxygen concentration in the flue gas affects the required investment in the reactor to achieve the desired rate of NO to NO_2 conversion. Cases 6 and 7, which have 1% O_2 in the flue gas, have a 3-fold higher cost for the reactor than Case 1 with 4% O_2 in the flue gas. Figure 4b shows the total annual specific costs (in €/tonne of removed NO_x and SO_x) for all the cases, excluding the costs associated with compressors, as compared to the specific costs of the SCR and WFGD systems. The cost of the chemicals, with a major contribution from NaOH for pH control, is the factor that contributes most to the specific cost of the removal process. Comparing Concept A (Case 1) and Concept B (Case 5), for the same flue gas compositions, it may be concluded that the specific cost of NO_x and SO_x control is 36% lower when the single-column design (Concept A) is used, due to lower capital and fixed operating costs. Case 3 has the lowest specific cost, due to the high S/N ratio in the flue gas and the consequently increased effectiveness of the acid gas removal, yielding lower capital and operating costs. The conventional removal processes, i.e., SCR and WFGD, have high specific costs relative to Concepts A and B, mainly due to the higher capital cost. The total direct cost of the SCR system is 58 M€ and of the WFGD system is 26 M€. It should, however, be noted that it is difficult to compare novel techniques with established techniques, and that different methodologies have been applied to estimate the cost for each individual system. For a flue gas that contains both NO_x and SO_x (same concentrations as in Case 1), a combination of WFGD and SCR or a combination of WFGD and Concept A will result in a 300% or 98% increase, respectively, in the specific cost of removal, as compared to Concept A (Case 1). A combination of WFGD and Concept A has a 46% higher specific cost than Concept B (Case 5).

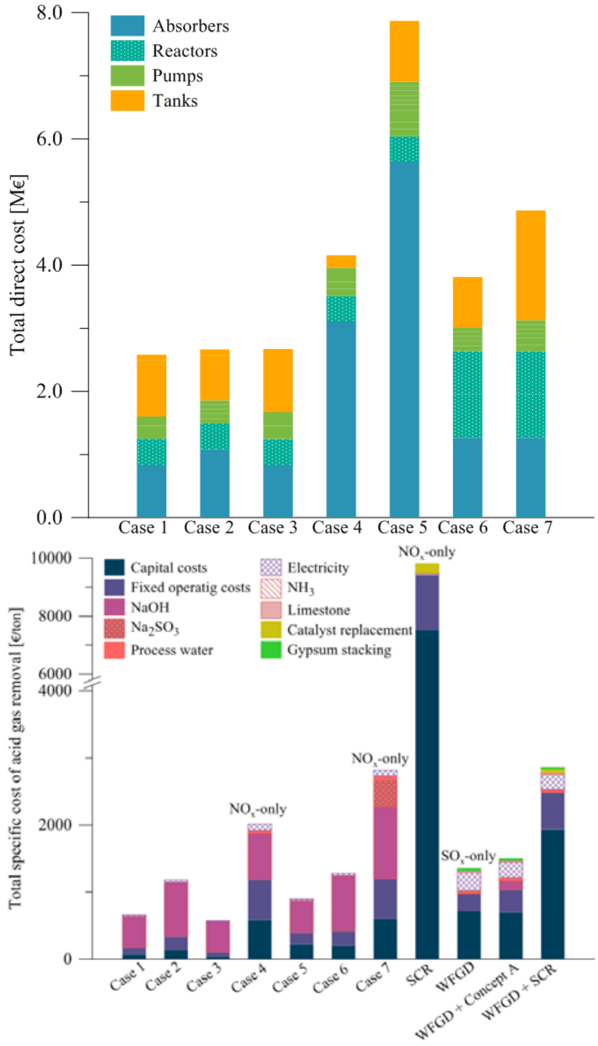

Figure 4. a) Total direct cost for the NO_x and SO_x removal processes for Cases 1–7 (cost of the LPC is not included). b) Comparison of the total specific costs for removal of acid gases (NO_x + SO_x) for Cases 1–7, in addition to the FGD and SCR systems.

4. Discussion

To discuss the sensitivity of the obtained results, the cost assumptions that most strongly affect the total plant costs are studied. Figure 5 shows the sensitivity to cost of the NaOH, total direct costs, interest rate on the total specific costs for Concepts A (Case 1) and B (Case 5) and for WFGD+Concept A for the same flue gas. The consumption of chemicals, and specifically for pH control, is a key component of the operating cost of the proposed process. The sensitivity of the total specific cost of removal to the cost of NaOH is weakest for the WFGD+Concept A system. If the cost of NaOH is doubled, use of the WFGD system for SO_x removal might be as attractive as using the atmospheric SO_x absorber system. However, the volume of wastewater from the WFGD+Concept A system is about 3-fold greater than that from the Concept B system, which implies a higher cost for wastewater treatment in the former case. The WFGD+Concept A system has the highest sensitivity, with around 20% change in the total specific cost as a result of 50% change in the total specific costs and the interest rate, The predicted total specific cost of removal in Cases 1 and 5 is not sensitive (<5% change in total specific cost caused by a 50% change) to either the equipment costs or the interest rate.

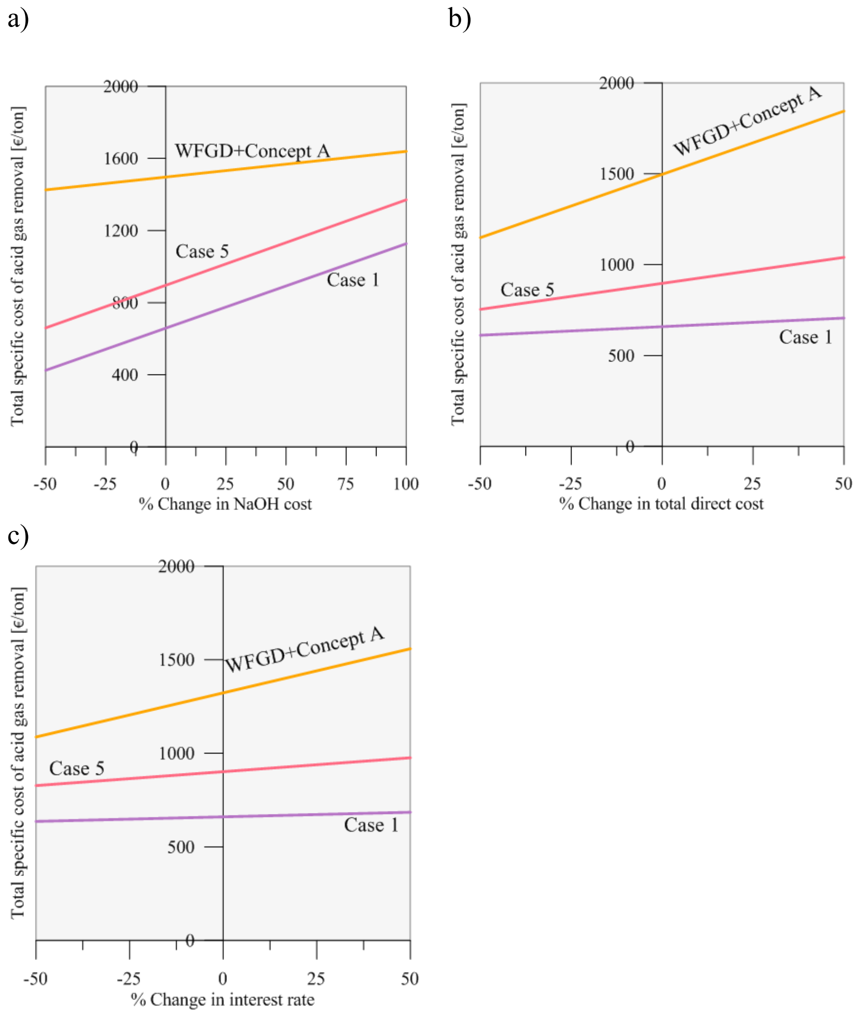

Figure 5. Sensitivities of the total specific cost of acid gas removal to: a) the cost of NaOH; b) the total direct cost of the gas removal system, and c) the interest rate.

Lastly, the choice of the removal system will not only be affected by the total costs discussed here, but also by the requirements of the upstream and downstream processes. For example, in the case of a high-sulfur fuel and to avoid sulfur retention in the boiler, the use of a sulfur removal process in the recycle loop of an oxy-fuel system might be needed to avoid corrosion in the boiler. In that case, the capital cost of the WFGD and SO_2 removal absorber will be around 1.5–2.0-times higher than that calculated here, due to the greater volume flow of flue gas to be treated. Furthermore, the design of the removal process, more specifically the required volume of the reactor to provide a sufficient residence time for NO oxidation prior to absorption, can be affected by the design of the compression process, in terms of pressure levels, intercooling temperature etc. For instance, a decrease of 10°C in the intercooling temperature enhances NO oxidation and decreases by approximately 15% the cost of the reactors. However, in such a case, the cost of cooling water will increase, and this should be considered when optimizing the process.

5. Conclusions

We evaluated the technical and economic performances of a NO_x and SO_x removal unit integrated with flue gas compression in oxy-fuel and chemical looping combustion systems. This entailed the establishment of basic design flowsheets for a reference design concept, i.e., a single-column pressurized absorber, and an alternative design with a dual column setting for atmospheric removal of SO_2 and pressurized removal of NO_x. Removal efficiencies comparable to those of BAT (>98% for SO_2 and >90% for NO_x) can be achieved by the proposed process. The investment costs for the proposed concepts are significantly lower (by about one-third) than for a corresponding conventional flue gas cleaning system that employs wet flue gas desulfurization and selective catalytic reduction.

The risks identified for the proposed absorption-based processes include the sensitivity to cost-efficient pH control and the generation of considerable volumes of a new type of wastewater, as compared to a WFGD system. The wastewater treatment must be capable of handling higher concentrations of nitrogen acids and sulfites. The costs of wastewater treatments for the proposed process need to be investigated further.

The proposed process works for a wide range of flue gas compositions. The presence of sulfur in the flue gas enhances NO_x removal and decreases the specific cost of emissions control. Low concentrations of O_2 in the flue gas increase the capital costs.

6. References

1. Yoon, H. J.; Park, H.-W.; Park, D.-W., Simultaneous Oxidation and Absorption of NOx and SO2 in an Integrated O3 Oxidation/Wet Atomizing System. *Energy & Fuels* **2016,** *30* (4), 3289-3297.

2. Hultén, A. H.; Nilsson, P.; Samuelsson, M.; Ajdari, S.; Normann, F.; Andersson, K., First evaluation of a multicomponent flue gas cleaning concept using chlorine dioxide gas – Experiments on chemistry and process performance. *Fuel* **2017,** *210*, 885-891.

3. Hutson, N. D.; Krzyzynska, R.; Srivastava, R. K., Simultaneous Removal of SO2, NOX, and Hg from Coal Flue Gas Using a NaClO2-Enhanced Wet Scrubber. *Industrial & Engineering Chemistry Research* **2008,** *47* (16), 5825-5831.

4. Jr, E. B. M.; Overcamp, T. J., Hydrogen Peroxide Scrubber for the Control of Nitrogen Oxides. *Environmental Engineering Science* **2002,** *19* (5), 321-327.

5. Allam R. J., W. V., Miller J. Purification of Carbon Dioxide. May 31, 2007, 2007.

6. Liémans, I.; Alban, B.; Tranier, J.-P.; Thomas, D., SO_x and NO_x absorption based removal into acidic conditions for the flue gas treatment in oxy-fuel combustion. *Energy Procedia* **2011,** *4* (0), 2847-2854.

7. Court Ph., B. A., Darde A., Trainer J.P., Perrin N., Callide CO_2 capture pliot plant design. In *2nd IEA GHG Oxyfuel Combustion Conference*, Yeppoon, Australiua, 2011.

8. White, V.; Wright, A.; Tappe, S.; Yan, J., The Air Products Vattenfall Oxyfuel CO2 Compression and Purification Pilot Plant at Schwarze Pumpe. *Energy Procedia* **2013,** *37*, 1490-1499.

9. NETL *Pulverized Coal Oxycombustion Power Plants* DOE/NETL-2007/1291; United States Department of Energy's National Energy Technology Laboratory: 2008.

10. Shah K., E. L., Spörl R., Belo L., Wall T. Coal quality impacts and gas quality control in oxy-fuel technology for carbon capture and storage – cost impacts and coal value; University of Newcastle, Australia: January 2014, 2014.

11. Ajdari, S., Normann F., Andersson K., Evaluation of operating-and design parameters for pressurized flue gas systems with integrated removal of NOx and SO_2. 2019.

12. Lyngfelt, A.; Leckner, B., A 1000MWth boiler for chemical-looping combustion of solid fuels – Discussion of design and costs. *Applied Energy* **2015,** *157*, 475-487.

13. Lecomte Th., F. d. l. J. F., Neuwahl F., Canova M., Pinasseau A., Jankov I., Brinkmann Th., Roudier S., Delgado Sancho L., Best Available Techniques (BAT) Reference Document for Large Combustion Plants (JRC), J. R. C., Ed. Publications Office of the European Union: Luxembourg, 2017.

14. R., G. *Wet flue gas desulfurization technology evaluation*; Sargent and Lundy: Chiago, IL, USA, 2003.

15. Ajdari, S.; Normann, F.; Andersson, K.; Johnsson, F., Modeling the Nitrogen and Sulfur Chemistry in Pressurized Flue Gas Systems. *Industrial & Engineering Chemistry Research* **2015,** *54* (4), 1216-1227.

16. Ajdari, S.; Normann, F.; Andersson, K.; Johnsson, F., Reduced Mechanism for Nitrogen and Sulfur Chemistry in Pressurized Flue Gas Systems. *Industrial & Engineering Chemistry Research* **2016,** *55* (19), 5514-5525.

17. Anantharaman, R.; Bolland, O.; Booth, N.; van Dorst, E.; Ekstrom, C.; Fernandes, E. S.; Franco, F.; Macchi, E.; Manzolini, G.; Nikolic, D.; Pfeffer, A.; Prins, M.; Rezvani, S.; Robinson, L. *European best practise guidelines for assesment of CO_2 capture technologies*; D1.4.3 in DECARBit Project; 2011.

18. NETL Quality guidelines for energy system studies: Cost estimation methodology for NETL assessments of power plant performance; National Energy Technology Laboratory: 2011.

19. White, V.; Wright, A.; Tappe, S.; Yan, J., The Air Products Vattenfall Oxyfuel CO2 Compression and Purification Pilot Plant at Schwarze Pumpe. *Energy Procedia* **2013,** *37* (0), 1490-1499.

20. Lockwood, F.; Granados, L.; Leclerc, M.; Lesort, A.-L.; Beasse, G.; Delgado, M. A.; Spero, C., Oxy-combustion CPU – From Pilots Towards Industrial-scale Demonstration. *Energy Procedia* **2014,** *63*, 342-351.

21. Daniel Mussatti, R. S., Paula M. Hemmer, Randy Strait *Selective Catalytic Reduction*; U.S. Environmental Protection Agency: 2000.

22. Normann, F.; Andersson, K.; Leckner, B.; Johnsson, F., Emission control of nitrogen oxides in the oxy-fuel process. *Progress in Energy and Combustion Science* **2009,** *35* (5), 385-397.

23. Agency, U. S. E. P. EPA Air Pollution Control Cost Manual; 2016.

24. Jilvero, H.; Normann, F.; Andersson, K.; Johnsson, F., Ammonia-based post combustion – The techno-economics of controlling ammonia emissions. *International Journal of Greenhouse Gas Control* **2015,** *37*, 441-450.

25. Srivastava, R. K. *CONTROLLING SO2 EMISSIONS: A REVIEW OF TECHNOLOGIES* EPA/600/R-00/093; U.S. Environmental Protection Agency: Washington, D.C., 2000.

26. Darde, A.; Prabhakar, R.; Tranier, J. P.; Perrin, N. In Air separation and flue gas compression and purification units for oxy-coal combustion systems, 2009; pp 527-534.

27. Carpenter, A. M. *Waste water regulations and issues for coal-fired power plants*; IEA Clean Coal Center: London, UK, 2018.

28. Commission, T. E., Best Available Techniques (BAT) Coclusions. Official Journal of the European Union: Brussels, 2017.

29. Agency, U. E. P., Effluent Limitations Guidelines and Standards for the Steam Electric Power Generating Point Source Category Agency, E. P., Ed. United State of America 2015; Vol. 212.

30. Littlejohn, D.; Chang, S. G., Modeling of the chemistry of wet limestone flue gas desulfurization systems. *Energy & Fuels* **1991,** *5* (2), 249-254.

Appendix A. Equipment lists

Tables A1-A7

Table A1. Equipment list for Case 1

Equipment	Type/material	Size	Equipment ID
Columns and packing			
Absorber column	SS2205	D = 2.4 m, H=4.3 m	ABS-1
Absorber packing	Sulzer Mellapak 250Y	H=3 m	
Pumps			
NaOH pump	Centrifugal	1.1 kW	P-1
Makeup water pump	Centrifugal	0.8 kW	P-2
Absorber re-circulation pump	Centrifugal	0.7 kW	P-3
Absorber buffer pump	Centrifugal	0.7 kW	P-4
Absorber bleed-off pump	Centrifugal	0.1 kW	P-5
Cooling water pump	Centrifugal	230 kW	P-6
Condensate pump	Centrifugal	0.3 kW	P-7
Tanks and Vessels			
NaOH tank	Vertical cylinder, SS304L	H= 3.7 m, D = 14.8 m	T-1
Absorber buffer tank	Vertical cylinder, SS2205	H= 1 m, D = 0.3 m	T-2
Condensate storage tank	Vertical cylinder, SS304L	H= 11 m, D = 2.8 m	T-3
Knock-out drum 1	Vertical cylinder, SS304L	H= 8.3 m, D = 5.3 m	KO-1
Knock-out drum 2	Vertical cylinder, SS304L	H= 6.3 m, D = 3.9 m	KO-2
Knock-out drum 3	Vertical cylinder, SS304L	H = 3.6 m, D = 2 m	KO-3
Flash drum 1	Horizontal drum, SS304L	H= 2.6 m, D = 0.9 m	D-1
Flash drum 2	Horizontal drum, SS304L	H= 1.8 m, D = 0.6 m	D-2
Flash drum 3	Horizontal drum, SS304L	H= 1.5 m, D = 0.4 m	D-3
Reactor	Vertical cylinder, SS304L	H= 11.3 m, D = 2.8 m	R-1
Heat exchangers			
Intercooler 1	SS2205	1426	HX-1
Intercooler 2	SS2205	1200	HX-2
Intercooler 3	SS2205	1230	HX-3
Compressor			
Three-stage compressor	Centrifugal, SS304L	31 MW	CMP-1-3

Table A2. Equipment list for Case 2

Equipment	Type/material	Size	Equipment ID
Columns and packing			
Absorber column	SS2205	D = 2.4 m, H=10 m	ABS-1
Absorber packing	Sulzer Mellapak 250Y	H=7 m	
Pumps			
NaOH pump	Centrifugal	0.7 kW	P-1
Makeup water pump	Centrifugal	0.3 kW	P-2
Absorber re-circulation pump	Centrifugal	1 kW	P-3
Absorber buffer pump	Centrifugal	1 kW	P-4
Absorber bleed-off pump	Centrifugal	0.1 kW	P-5
Cooling water pump	Centrifugal	230 kW	P-6
Condensate pump	Centrifugal	0.3 kW	P-7
Tanks and Vessels			
NaOH tank	Vertical cylinder, SS304L	H= 3.7 m, D = 14.7 m	T-1
Absorber buffer tank	Vertical cylinder, SS2205	H= 1.4 m, D = 0.3 m	T-2
Condensate storage tank	Vertical cylinder, SS304L	H= 11 m, D = 2.8 m	T-3
Knock-out drum 1	Vertical cylinder, SS304L	H= 8.3 m, D = 5.3 m	KO-1
Knock-out drum 2	Vertical cylinder, SS304L	H= 6.3 m, D = 3.9 m	KO-2
Knock-out drum 3	Vertical cylinder, SS304L	H = 3.6 m, D = 2 m	KO-3
Flash drum 1	Horizontal drum, SS304L	H= 2.6 m, D = 0.9 m	D-1
Flash drum 2	Horizontal drum, SS304L	H= 1.8 m, D = 0.6 m	D-2
Flash drum 3	Horizontal drum, SS304L	H= 1.5 m, D = 0.4 m	D-3
Reactor	Vertical cylinder, SS304L	H= 8.5 m, D = 2.1 m	R-1
Heat exchangers			
Intercooler 1	SS2205	1426	HX-1
Intercooler 2	SS2205	1200	HX-2
Intercooler 3	SS2205	1230	HX-3
Compressor			
Three-stage compressor	Centrifugal, SS304L	31 MW	CMP-1-3

Table A3. Equipment list for Case 3

Equipment	Type/material	Size	Equipment ID
Columns and packing			
Absorber column	SS2205	D = 2.4 m, H=4.3 m	ABS-1
Absorber packing	Sulzer Mellapak 250Y	H= 3 m	
Pumps			
NaOH pump	Centrifugal	1.2 kW	P-1
Makeup water pump	Centrifugal	0.8 kW	P-2
Absorber re-circulation pump	Centrifugal	0.6 kW	P-3
Absorber buffer pump	Centrifugal	0.6 kW	P-4
Absorber bleed-off pump	Centrifugal	0.1 kW	P-5
Cooling water pump	Centrifugal	230 kW	P-6
Condensate pump	Centrifugal	0.3 kW	P-7
Tanks and Vessels			
NaOH tank	Vertical cylinder, SS304L	H= 4.4 m, D = 17.6 m	T-1
Absorber buffer tank	Vertical cylinder, SS2205	H= 1.2 m, D = 0.3 m	T-2
Condensate storage tank	Vertical cylinder, SS304L	H= 11 m, D = 2.8 m	T-3
Knock-out drum 1	Vertical cylinder, SS304L	H= 8.3 m, D = 5.3 m	KO-1
Knock-out drum 2	Vertical cylinder, SS304L	H= 6.3 m, D = 3.9 m	KO-2
Knock-out drum 3	Vertical cylinder, SS304L	H = 3.6 m, D = 2 m	KO-3
Flash drum 1	Horizontal drum, SS304L	H= 2.6 m, D = 0.9 m	D-1
Flash drum 2	Horizontal drum, SS304L	H= 1.8 m, D = 0.6 m	D-2
Flash drum 3	Horizontal drum, SS304L	H= 1.5 m, D = 0.4 m	D-3
Reactor	Vertical cylinder, SS304L	H= 11.3 m, D = 2.8 m	R-1
Heat exchangers			
Intercooler 1	SS2205	1426	HX-1
Intercooler 2	SS2205	1200	HX-2
Intercooler 3	SS2205	1230	HX-3
Compressor			
Three-stage compressor	Centrifugal, SS304L	31 MW	CMP-1-3

Table A4. Equipment list for Case 4

Equipment	Type/material	Size	Equipment ID
Columns and packing			
Absorber column	SS2205	D = 2.8 m, H=42.3 m	ABS-1
Absorber packing	Sulzer Mellapak 250Y	H=30 m	
Pumps			
NaOH pump	Centrifugal	0.2 kW	P-1
Makeup water pump	Centrifugal	4.3 kW	P-2
Absorber re-circulation pump	Centrifugal	4.5 kW	P-3
Absorber buffer pump	Centrifugal	4.5 kW	P-4
Absorber bleed-off pump	Centrifugal	0.2 kW	P-5
Cooling water pump	Centrifugal	230 kW	P-6
Condensate pump	Centrifugal	0.3 kW	P-7
Tanks and Vessels			
NaOH tank	Vertical cylinder, SS304L	H= 2.4 m, D = 9.6 m	T-1
Absorber buffer tank	Vertical cylinder, SS2205	H= 1.9 m, D = 0.5 m	T-2
Condensate storage tank	Vertical cylinder, SS304L	H= 11 m, D = 2.8 m	T-3
Knock-out drum 1	Vertical cylinder, SS304L	H= 8.3 m, D = 5.3 m	KO-1
Knock-out drum 2	Vertical cylinder, SS304L	H= 6.3 m, D = 3.9 m	KO-2
Knock-out drum 3	Vertical cylinder, SS304L	H = 3.6 m, D = 2 m	KO-3
Flash drum 1	Horizontal drum, SS304L	H= 2.6 m, D = 0.9 m	D-1
Flash drum 2	Horizontal drum, SS304L	H= 1.8 m, D = 0.6 m	D-2
Flash drum 3	Horizontal drum, SS304L	H= 1.5 m, D = 0.4 m	D-3
Reactor	Vertical cylinder, SS304L	H= 8.5 m, D = 2.1 m	R-1
Heat exchangers			
Intercooler 1	SS2205	1426	HX-1
Intercooler 2	SS2205	1200	HX-2
Intercooler 3	SS2205	1230	HX-3
Compressor			
Three-stage compressor	Centrifugal, SS304L	30 MW	CMP-1-3

Table A5. Equipment list for Case 5

Equipment	Type/material	Size	Equipment ID
Columns and packing			
SO_2 absorber column	SS2205	D = 5.5 m, H=2.9 m	ABS-1
SO_2 absorber packing	Sulzer Mellapak 250Y	H= 2 m	
NO_x absorber column	SS2205	D = 2.8 m, H=42.3 m	ABS-2
NO_x absorber packing	Sulzer Mellapak 250Y	H= 30 m	
Pumps			
NaOH pump to SO_2 Absorber	Centrifugal	0.5 kW	P-1
Makeup water pump to SO_2 Absorber	Centrifugal	0.1 kW	P-2
SO_2 absorber re-circulation pump	Centrifugal	0.5 kW	P-3
SO_2 absorber buffer pump	Centrifugal	0.5 kW	P-4
SO_2 absorber bleed-off pump	Centrifugal	0.1 kW	P-5
Cooling water pump	Centrifugal	230 kW	P-6
Condensate pump	Centrifugal	0.3 kW	P-7
NaOH pump to NO_x Absorber	Centrifugal	0.1 kW	P-8
Makeup water pump to NO_x Absorber	Centrifugal	4.3 kW	P-9
NO_x absorber re-circulation pump	Centrifugal	4 kW	P-10
NO_x absorber buffer pump	Centrifugal	4 kW	P-11
NO_x absorber bleed-off pump	Centrifugal	0.2 kW	P-12
Tanks and Vessels			
NaOH tank	Vertical cylinder, SS304L	H= 3.7 m, D = 14.7 m	T-1
SO_2 absorber buffer tank	Vertical cylinder, SS2205	H= 1.5 m, D = 0.4 m	T-2
Condensate storage tank	Vertical cylinder, SS304L	H= 11 m, D = 2.8 m	T-3
NO_x absorber buffer tank	Vertical cylinder, SS2205	H= 1.9 m, D = 0.5 m	T-4
Knock-out drum 1	Vertical cylinder, SS304L	H= 8.3 m, D = 5.3 m	KO-1
Knock-out drum 2	Vertical cylinder, SS304L	H= 6.3 m, D = 3.9 m	KO-2
Knock-out drum 3	Vertical cylinder, SS304L	H = 3.6 m, D = 2 m	KO-3
Flash drum 1	Horizontal drum, SS304L	H= 2.6 m, D = 0.9 m	D-1
Flash drum 2	Horizontal drum, SS304L	H= 1.8 m, D = 0.6 m	D-2
Flash drum 3	Horizontal drum, SS304L	H= 1.5 m, D = 0.4 m	D-3
Reactor	Vertical cylinder, SS304L	H= 8.5 m, D = 2.1 m	R-1
Heat exchangers			
Compressor			
Three-stage compressor	Centrifugal, SS304L	30 MW	CMP-1-3

Table A6. Equipment list for Case 6

Equipment	Type/material	Size	Equipment ID
Columns and packing			
Absorber column	SS2205	D = 2.4 m, H=14.3 m	ABS-1
Absorber packing	Sulzer Mellapak 250Y	H= 10 m	
Pumps			
NaOH pump	Centrifugal	0.7 kW	P-1
Makeup water pump	Centrifugal	0.3 kW	P-2
Absorber re-circulation pump	Centrifugal	2.2 kW	P-3
Absorber buffer pump	Centrifugal	2.2 kW	P-4
Absorber bleed-off pump	Centrifugal	0.1 kW	P-5
Cooling water pump	Centrifugal	230 kW	P-6
Condensate pump	Centrifugal	0.3 kW	P-7
Tanks and Vessels			
NaOH tank	Vertical cylinder, SS304L	H= 3.6 m, D = 14.3 m	T-1
Absorber buffer tank	Vertical cylinder, SS2205	H= 1.8 m, D = 0.4 m	T-2
Condensate storage tank	Vertical cylinder, SS304L	H= 11 m, D = 2.8 m	T-3
Knock-out drum 1	Vertical cylinder, SS304L	H= 8.3 m, D = 5.3 m	KO-1
Knock-out drum 2	Vertical cylinder, SS304L	H= 6.3 m, D = 3.9 m	KO-2
Knock-out drum 3	Vertical cylinder, SS304L	H = 3.6 m, D = 2 m	KO-3
Flash drum 1	Horizontal drum, SS304L	H= 2.6 m, D = 0.9 m	D-1
Flash drum 2	Horizontal drum, SS304L	H= 1.8 m, D = 0.6 m	D-2
Flash drum 3	Horizontal drum, SS304L	H= 1.5 m, D = 0.4 m	D-3
Reactor 1	Vertical cylinder, SS304L	H= 14.4 m, D = 3.6 m	R-1
Reactor 2	Vertical cylinder, SS304L	H= 14.4 m, D = 3.6 m	R-2
Heat exchangers			
Intercooler 1	SS2205	1426	HX-1
Intercooler 2	SS2205	1200	HX-2
Intercooler 3	SS2205	1230	HX-3
Compressor			
Three-stage compressor	Centrifugal, SS304L	31 MW	CMP-1-3

Table A7. Equipment list for Case 7

Equipment	Type/material	Size	Equipment ID
Columns and packing			
Absorber column	SS2205	D = 2.4 m, H=14.3 m	ABS-1
Absorber packing	Sulzer Mellapak 250Y	H= 10 m	
Pumps			
NaOH pump	Centrifugal	0.7 kW	P-1
Makeup water pump	Centrifugal	0.3 kW	P-2
Absorber re-circulation pump	Centrifugal	2.2 kW	P-3
Absorber buffer pump	Centrifugal	2.2 kW	P-4
Absorber bleed-off pump	Centrifugal	0.1 kW	P-5
Cooling water pump	Centrifugal	230 kW	P-6
Condensate pump	Centrifugal	0.3 kW	P-7
Tanks and Vessels			
NaOH tank	Vertical cylinder, SS304L	H= 3.6 m, D = 14.3 m	T-1
Absorber buffer tank	Vertical cylinder, SS2205	H= 1.8 m, D = 0.4 m	T-2
Condensate storage tank	Vertical cylinder, SS304L	H= 11 m, D = 2.8 m	T-3
Na_2SO_3 tank	Vertical cylinder, SS304L	H= 18.2 m, D = 4.6 m	T-4
Knock-out drum 1	Vertical cylinder, SS304L	H= 8.3 m, D = 5.3 m	KO-1
Knock-out drum 2	Vertical cylinder, SS304L	H= 6.3 m, D = 3.9 m	KO-2
Knock-out drum 3	Vertical cylinder, SS304L	H = 3.6 m, D = 2 m	KO-3
Flash drum 1	Horizontal drum, SS304L	H= 2.6 m, D = 0.9 m	D-1
Flash drum 2	Horizontal drum, SS304L	H= 1.8 m, D = 0.6 m	D-2
Flash drum 3	Horizontal drum, SS304L	H= 1.5 m, D = 0.4 m	D-3
Reactor 1	Vertical cylinder, SS304L	H= 14.4 m, D = 3.6 m	R-1
Reactor 2	Vertical cylinder, SS304L	H= 14.4 m, D = 3.6 m	R-2
Heat exchangers			
Intercooler 1	SS2205	1426	HX-1
Intercooler 2	SS2205	1200	HX-2
Intercooler 3	SS2205	1230	HX-3
Compressor			
Three-stage compressor	Centrifugal, SS304L	31 MW	CMP-1-3

Appendix B. Cost estimations of the integrated removal process

Table B1. Total plant costs and operating costs for all cases

	Case 1	Case 2	Case 3	Case 4	Case 5	Case 6	Case 7	FGD	SCR
Total direct cost w/o process contingencies (M€)	2.6	2.7	2.7	4.26	7.87	3.8	4.9	26	58.3
Process contingencies	0.9	0.9	0.9	1-5	2.8	1.3	1.7	1.3	2.9
Total direct cost (M€)	**3.5**	**3.6**	**3.6**	**5.8**	**10.6**	**5.1**	**6.6**	**27.3**	**61.2**
Indirect costs	0.5	0.5	0.5	0.8	1.5	0.7	0.9	5.5	12.2
Owner’s costs	0.2	0.3	0.3	0.4	0.7	0.4	0.5		
Project contingencies	0.5	0.5	0.5	0.9	1.6	0.8	1.0	4.9	11.0
Total plant cost (M€)	**4.7**	**4.9**	**4.9**	**7.8**	**14.4**	**7.0**	**8.9**	**37.8**	**84.5**
Annualized CAPEX (M€/year)	**0.44**	**0.46**	**0.46**	**0.73**	**1.35**	**0.66**	**0.84**	**3.53**	**7.91**
Fixed OPEX (M€/year)	**0.6**	**0.6**	**0.6**	**0.8**	**1.1**	**0.7**	**0.8**	**1.3**	**2.0**
Variable OPEX (M€/year)	**3**	**2.8**	**5.1**	**1**	**3.1**	**2.8**	**2.2**	**1.8**	**0.4**