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Energy and exergy analyses for the carbon capture with the Chilled Ammonia Process (CAP)

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Abstract

Post-combustion carbon capture in existing power plants is a strategic technology that can reduce emissions from power generation. The proven approach is scrubbing with amines. However, its drawbacks are energy requirement, 3 to 5 MJ per kg of captured CO₂, as well as solution corrosion and solvent degradation. An alternative approach is scrubbing with chilled aqueous ammonia. This technology aims at mitigating energy usage and solving corrosion and degradation issues. Here an approximate model of the CO₂-H₂O-NH₃ system is coupled with a proposed process to evaluate mass, energy and entropy flows. For 1 kg of captured CO₂, the simulation yields a steam extraction of 0.59 kg, equivalent to a heat duty exceeding slightly 1.5 MJ and a generation loss approaching closely 0.1 kWh, an auxiliary consumption of 0.1 kWh and a delta of almost 0.18 kWh with respect to the ideal case. Assuming a cost of electricity of 7c€/kWh, the sole operation of the capture system totals 14 €/ton_{CO₂}.

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Chilled Ammonia Process (CAP); Carbon Capture and Storage (CCS); coal-fired power plant; natural gas-fired power plant

1. Introduction

Despite the concerns about rising concentrations of greenhouse gases in the atmosphere, fossil fuels are likely to remain the main source of primary energy for long. Nevertheless, an important contribution towards the reduction of their emissions from fossil fuel-fired power plants may be from Carbon Capture and Storage (CCS) meaning that the CO₂ formed by combustion is captured and stored over an indefinite period. In principle, the CO₂ can be captured according to one of three basic ways: (1) pre-combustion, (2) post-combustion and (3) oxy-fuel combustion. All these ways will play probably a similar role in the short- and the mid-term future, for each of them has peculiar characteristics that makes it appropriate in specific circumstances. Post-combustion capture is strategic for the retrofit of the many existing power plants that will be in service for decades. Traditionally, the sweetening of gases was accomplished with aqueous monoethanol amine (MEA). As it is demanding from the standpoints of energy consumption, corrosion requirement and solvent degradation and as carbon dioxide separation is becoming popular

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in diverse industrial sectors, secondary and tertiary amines have been investigated. Currently, advanced amines, which comprise upload promoters and corrosion inhibitors, are under research. A different approach is to chemically absorb the carbon dioxide into aqueous ammonia at chilled conditions, as patented by EIG Inc. [1]. Among others, Alstom is engaged in an intensive development of the Chilled Ammonia Process (CAP) based on that patent. This paper analyzes a CAP-type process. It begins with a bibliographic review that covers the background of CO₂-H₂O-NH₃. Then, it outlines the thermochemical model used to compute mass, energy and entropy flows for a the scheme developed during the investigation. Finally, it reports first- and second-law results from the computer simulation. As to the knowledge of the writers, very little has been written regarding energy balances for complete systems and nothing at all regarding entropy balances. Entropic analysis has the capability of identifying those processes that are responsible for the greater irreversibilities and hence that require an alternative design, if possible.

2. Bibliographic review

CO₂-H₂O-NH₃ systems has been investigated by a number of scientists in the last decades. At first the main scope was modeling the chemistry to prepare numerical tools for sour-water treatment. Only in the last decade the interest has shifted onto capturing carbon dioxide. The process is being studied at either room temperature or chilled conditions. The following gives a review of the chemistry of the solution, separating the publications that regard mainly the description of experiments from those that consider primarily the development of models. Thereafter is the review of general manuscripts on ammonia scrubbing and lastly of Alstom's publications on its CAP.

2.1. Chemistry of the CO₂-H₂O-NH₃ system

According to the majority of the publications located in the open literature, the species present in the ternary system at working conditions of CAP are: CO₂, H₂O and NH₃, in both the vapor and the liquid phases, as well as the aqueous ions H₃O⁺, OH⁻, NH₄⁺, HCO₃⁻, CO₃²⁻, NH₂COO⁻ and, if precipitation occurs in the solution, the pure salt NH₄HCO₃. In contrast, not all authors agree on the possible formation and combination of the salts made of (NH₄)₂CO₃ and NH₂COONH₄. The widely proposed reactions pertinent to the capture and regeneration stages are:



Eqs. 1 and 2 describe the capture of carbon dioxide that is sought inside the absorber whereas eqs. 3 and 4 represent the precipitation of the ammonium bicarbonate salt that occurs at low temperature. On the contrary, the formation of the ion carbamate, eq. 5, may lead to an undesirable capture carbon dioxide, eq. 6, which has a greater enthalpy of reaction for the regeneration. Alternatively, carbamate may combine with ammonium ion, eq. 7.

In 1982 Pawllkowski et al. [2] reported the experimental vapor-liquid equilibrium data of ammonia and carbon dioxide aqueous system, optionally added with salts, at temperatures of 100°C and 150°C. These data were then utilized to calibrate semi-empirical thermodynamic routines (detailed by Edwards et al. and reviewed below).

Almost half a decade later, with the same sake of supporting the design of the equipment for water treatment, Goppert and Maurer [3] conducted an extensive empirical campaign on the equilibria of ammonia and carbon

dioxide aqueous solution in the 333 K to 393 K interval and up to 7 MPa. They re-calibrated those semi-empirical correlations. In 1995 Kurz et al. [4] continued the work both including the solid phase in the equilibrium and expanding the investigation between 313 K and 473 K. They updated the previously-calibrated parameters. Shortly later, Rumpf et al. [5] measured the enthalpy changes upon partial evaporation of aqueous solutions containing ammonia and carbon dioxide and compared them with the predictions from the most-recently regressed models revealing that deviations laid mostly within experimental uncertainty.

Ten years ago, Bai and Yeh [6] provided preliminary experimental data on what was, at that time, going to be referred to as the novel study of ammonia scrubbing. They pointed out the remarkable potential of achieving high removal efficiencies, over 95%, and absorption capacities, as high as 0.9 kg of CO₂ per kg of NH₃. Shortly after, Yeh and Bai [7] completed another experimental campaign with the scope of comparing amine and ammonia scrubbing and they confirmed the potential of the second over the first solvent. Experiments were conducted at room temperature in their first work and between 10°C to 40°C in the later one.

Hsu et al. [8] reported the absorption reaction kinetics of amines and ammonia solutions with carbon dioxide in flue gases. The temperature of investigation was though 50°C, which is relatively high for the CAP. Similarly, Diao et al. [9] also investigated the removal efficiency of the sole ammonia solution in the 25–55°C interval and regressed the parameters of the rate constant, in the Arrhenius form, for the capture reaction.

Interestingly, Mani et al. [10] applied ¹³C NMR spectroscopy at CO₂-NH₃ aqueous solution at room conditions and proved it is a reliable method to investigate the speciation of the ammonium salt of bicarbonate, carbonate and carbamate. They did not detect other species in the solution and determined that NH₂CO₂⁻ is the main species in presence of excess NH₃; in contrast, HCO₃⁻ prevails when most of NH₃ has reacted with CO₂ at lower pH; finally, the CO₃²⁻ anion is always present in solution but at a concentration always lower than carbamate.

In the '70 and '80, the most common approach to the numerical modeling of CO₂-H₂O-NH₃ systems was that developed jointly by two groups lead by the famous scientists Prausnitz and Maurer. As described by Edwards et al. [11], such approach implied combining the molecular-thermodynamic principles with Pitzer's semi-empirical correlation. Bieling et al. [12] introduced in that framework a technique to determine the most important parameters out of the many adjustable ones and applied such technique to ammonia and carbon dioxide aqueous solutions.

A recent approach is the use of the UNIQUAC model for the activity coefficients of the species in the liquid phase. An extended version of the model was started by Sander et al. [13] and later applied by Thomsen and Rasmussen to the CO₂ and/or NH₃ solutions [14,15]. Based on that work, a first assess of a chilled ammonia plant has been lately published (and reported in the next section). In addition to the extended version, Pazuki et al. [16] proposed a UNIQUAC-Non Random Factor (NRF) model for the description of CO₂-H₂O-NH₃ systems.

2.2. Scrubbing of CO₂ with aqueous NH₃

In 2005, Yeh et al [17] published the results of three-cycle absorption-regeneration tests conducted on MEA and ammonia in a batch reactor maintained at about 25°C. They also reported an approximate estimate of energy usage in a cyclical process. According to them, the energy necessary to heat the solution exiting the recuperator and to regenerate is 1017 cal per g of CO₂, which is high with respect to the figures provided by later authors.

Recently, Darde et al. [18] presented a numerical investigation of the phase equilibria for isothermal absorber and regenerator in the range of the working conditions from the original patent by EIG Inc. [1]. In the study, they employed the accurate thermochemical model by Thomsen and Rasmussen [15]. The results include the equilibrium composition of vapor, liquid and solid phases as a function of the CO₂ loading, i.e. the ratio of the number of moles of carbon dioxide and ammonia, the energy requirements for cooling the absorber and, lastly, the energy requirement for heating the regenerator as a function of CO₂ loading and NH₃ initial mass fraction. In short, the CO₂ loading shall be in the 0.33 and 0.67 for the lean solution, 0.67 and 1 for the rich solution. Ammonia initial concentration shall be about 28wt%.

2.3. Alstom's Chilled Ammonia Process

Alstom commenced the development of the CAP in 2006 establishing a 5-year program aiming at commercialization by the end of 2011. The program consisted of 4 phases: (1) small and (2) large scale testing at SRI Int. in the San Francisco Bay Area, CA, (3) field pilot testing of capture on coal-fired exhausts at We Energies' plant in Pleasant Prairie, WI, and at E.ON's plant in Karlshamn, Sweden, and (4) commercial demonstration of capture as well as of storage at the American Electric Power (AEP)'s Mountaineer coal plant. Bench testing is now completed and described in the Electric Power Research Institute (EPRI) technical report [19]. The first pilot testing began in July 2008 and will continue throughout the 2009 whereas the second pilot is under construction, as reported by Black et al. [20,21]. The commercial demonstration is in the engineering design, as detailed by Sherrick et al. [22]. In addition, full-size installations are being programmed for North America and North Europe. As explained in all publications, carbon dioxide is absorbed in an ammoniated solution at temperature below ambient level, 0-20°C, and at almost ambient pressure, generating a slurry containing ammonium bicarbonate. Captured CO₂ is desorbed by turning ammonium bicarbonate into ammonium carbonate at elevated temperatures, moderately above 100°C, and elevated pressures, 20-40 bar. The completed experiments allowed for a first comparison with conventional MEA based on the 460 MW super critical pulverized-coal plant investigated in the Department of Energy (DOE)/EPRI Parson's study [23]: (1) energy consumption for chilling the flue gases, removing absorption enthalpy of reaction and washing the streams can be relatively inexpensive, (2) operating the boiler with minimum excess air and cooling the exhausts reduces the flow volume by a third and increases the CO₂ concentration by a fourth, (3) capture efficiency can exceed 90%, (4) high pressure in the regeneration stage reduces substantially CO₂ compression work, (5) heat duty, and thus steam extraction, in the reboiler is small due to the low regeneration enthalpy of reaction, to the low water concentration in the vapor phase and to the low sensible enthalpy of the rich solution thanks to a high CO₂ loading, (6) direct contact cooling reduces further the SO₂, SO₃, NO₂ and particulate presence, (7) ammonia slip can be minimized to few ppm level by cold-water wash. In contrast to bench results, pilot testing preliminary results are not yet available to the public. However, they are expected to confirm also other key features like: reactants are stable, ammonia can be reintegrated into the process by various forms (anhydrous ammonia, aqueous ammonia, ammonium carbamate) and the system is reliable and cost-effective.

3. Thermochemical model

The thermochemical model adopted in this work is an approximate yet robust one because the scope of the work is to estimate mass, energy and entropy flows with a fair accuracy. In the next stage of the ongoing research the modeling will be brought to greater detail and precision. For that matter, the vapor phase is modeled as a mixture of ideal gases. Information on enthalpy and entropy of formations at standard state of all species are taken from the CRC Handbook [24]. Ideal gas heat capacities are defined as the common NASA Polynomials. The liquid phase is described in either two ways, depending on whether it is chemically reacting or not. In all areas of the process in which liquid water is employed to cool a gaseous stream, liquid phase is treated as pure water. Vapor and liquid equilibrium is computed by means of Raoult's Law assuming that water is the only condensable component. If the liquid phase is chemically reacting, it is assumed that it behaves as an incompressible ideal solution and that all captured carbon dioxide and all ammonia exist only in ionic forms. Moreover, in the lean solutions the only ions present are CO₃²⁻ and NH₄⁺ that react in the absorber with CO₂ and H₂O to form exclusively the ion HCO₃⁻, according to reactions 1 and 2. The lean solution is assumed to be preloaded hypothetically with (NH₄)₂CO₃. Depending upon the temperature, solid ammonium bicarbonate in the rich solution may precipitate, according to reactions 3 and 4. At regeneration conditions the reversal path is followed. Ammonia equivalent concentration in the initial solution, carbon capture efficiency and carbon solution loading are not calculated yet imposed in agreement with experimental results, mainly from Yeh et al. [17]. Being the adopted ammonia concentration relatively low (as stated in the next section), the formation of ion carbamate is neglected, in agreement with Mani et al. [10]. In addition, the model is not structured to compute the ammonia evaporation from the reacting liquid into the vapor phase and the dissolution of components other than carbon dioxide into the liquid. With regards to ions, enthalpy and entropy of formation at standard state are from the CRC [24] and heat capacity as a function of temperature from Thomsen and Rasmussen [15]. With regards to the ammonium bicarbonate salt, standard state data as well as temperature-dependent solubility and specific volume are from the CRC [24] while temperature-independent heat

capacity from Kopp's Rule. Finally, Refprop is used to compute properties of the extracted steam and of the carbon dioxide in the sole compression subsystem (described later), neglecting in the later case that a small quantity of water is included in the flow and making sure the properties are continuous at the boundary with those calculated from the above-described model.

4. Process description

The process analyzed in the present work comprises five subsystems, as illustrated with dashed rectangles in Fig. 1: (1) gas cooling, (2) carbon dioxide absorption and regeneration, (3) treated flue gas wash, (4) regenerated carbon dioxide wash and (5) carbon dioxide compression. It is assumed that upstream of the capture system is a wet desulphurization system. Flue gas cooling is achieved by way of three direct contact coolers, indicated with cooling columns CC1 thru CC3, out of which the first one utilizes water from dry coolers, DC1, while the other two from chillers, CH1 and CH2, operating at different evaporation temperatures. Flue gas chilling yields appreciable condensation of water and thus requires purges, PR1 to PR3, from the columns. After the refrigeration subsystem, the water content and the specific volume of the flow are appreciably reduced hence, at this point, is optimally placed the fan necessary to offset all pressure losses, named FN. Subsequently, the flow enters the absorption column, AB, where it encounters, sprayed from the top, a rich solution recycle, AB4, the lean solution, AC3, and a water make-up, WW4. The recycled stream and the lean stream are refrigerated by a chiller, CH3, in order to counteract the exothermic reaction occurring in the absorber and maintain it at a low temperature at which a moderate quantity of solid ammonium bicarbonate precipitates. Part of this slurry pumped from the bottom of the column, by PS1, is recycled whereas the remainder is boosted to regeneration pressure, by PS2, prior to entering a recuperator, RC, where, as it is heated, the precipitated salt dissolves. At the exit of the heat exchanger, the stream is preheated, in PH, by low pressure steam, ST3, extracted from the turbine and then directed into the regeneration column, RG. Sensible enthalpy and reaction enthalpy inside the column is provided by higher pressure steam, ST1, through an external reboiler, RB. The treated flue gas leaving the absorption column are washed, in WC1, by refrigerated so to minimize ammonia slip to the atmosphere. Similarly, regenerated carbon dioxide is washed, in WC2, so to simultaneously recover the ammonia from the vapor phase, reduce the water content and chill the flow going to the compression subsystem. The treated gas wash has a small water purge, PR4, while carbon dioxide wash a small water make-up. The carbon flow is compressed to slightly supercritical pressure by CC, cooled by dry coolers DC2, and ultimately pumped to transportation conditions by PC. The assumed parameters for the boundary conditions and the working conditions of all operational units are included in Tab. 1.

5. Simulation results

The thermochemical model was implemented in VBA and called by a spreadsheet document containing the first- and second-law analyses of the considered process. As indicated in Tab. 2 in terms specific to 1 kg of captured CO₂, the total heat duty, summing both pre-heater and reboiler, is about 1.5 MJ which is met by a steam extraction of 0.59 kg that results in a loss of electrical production from the turbine of about 0.1 kWh. Tab. 3 summarizes the results for the five subsystems: the first column shows the electrical consumption of the auxiliaries added to the original power plant as well as the electricity penalty related to the two steam extractions, while the second column shows the losses of reversible work related to the irreversible processes occurring in the subsystems along with the discharges of water purges and cold treated gas into the ambient. Referring to the energy analysis, in terms of unit mass of captured CO₂, auxiliaries consume globally almost 0.1 kWh (i.e a value very close to the electricity penalty quoted above for the steam extracted from the turbine), the most of which is for the chiller serving the absorber. As a whole, the absorption-regeneration subsystem takes over almost 58% of all electrical usage. The chillers of the gas refrigeration subsystem are the second most consuming ancillaries, as this subsystem is responsible for more than 20% of the usage. Carbon dioxide compression subsystem has a small share of about 13%, much smaller than in capture systems separating CO₂ at atmospheric pressure. The remainder is equally distributed over the two washing sections. In total, the electrical penalty of carbon capture is 0.2 kWh/kg CO₂. From a second-law perspective, gas cooling and absorption/regeneration subsystems have a similar irreversibility, respectively 39% and 45% of a total work increase of 0.18 kWh per kg of captured CO₂. Interestingly, compression entropy production, 4.4%, is between that of the treated gas wash, 2.9%, and that of the carbon dioxide wash, 6.2%, indicating that is advisable to improve

the second wash by employing a dry cooler in addition to the chiller and allow washing water to return hotter from the column. Entropy production from the discharge of purges and cold gas is very low, about 1% each, as expected, since their discharge temperature is close to the ambient one.

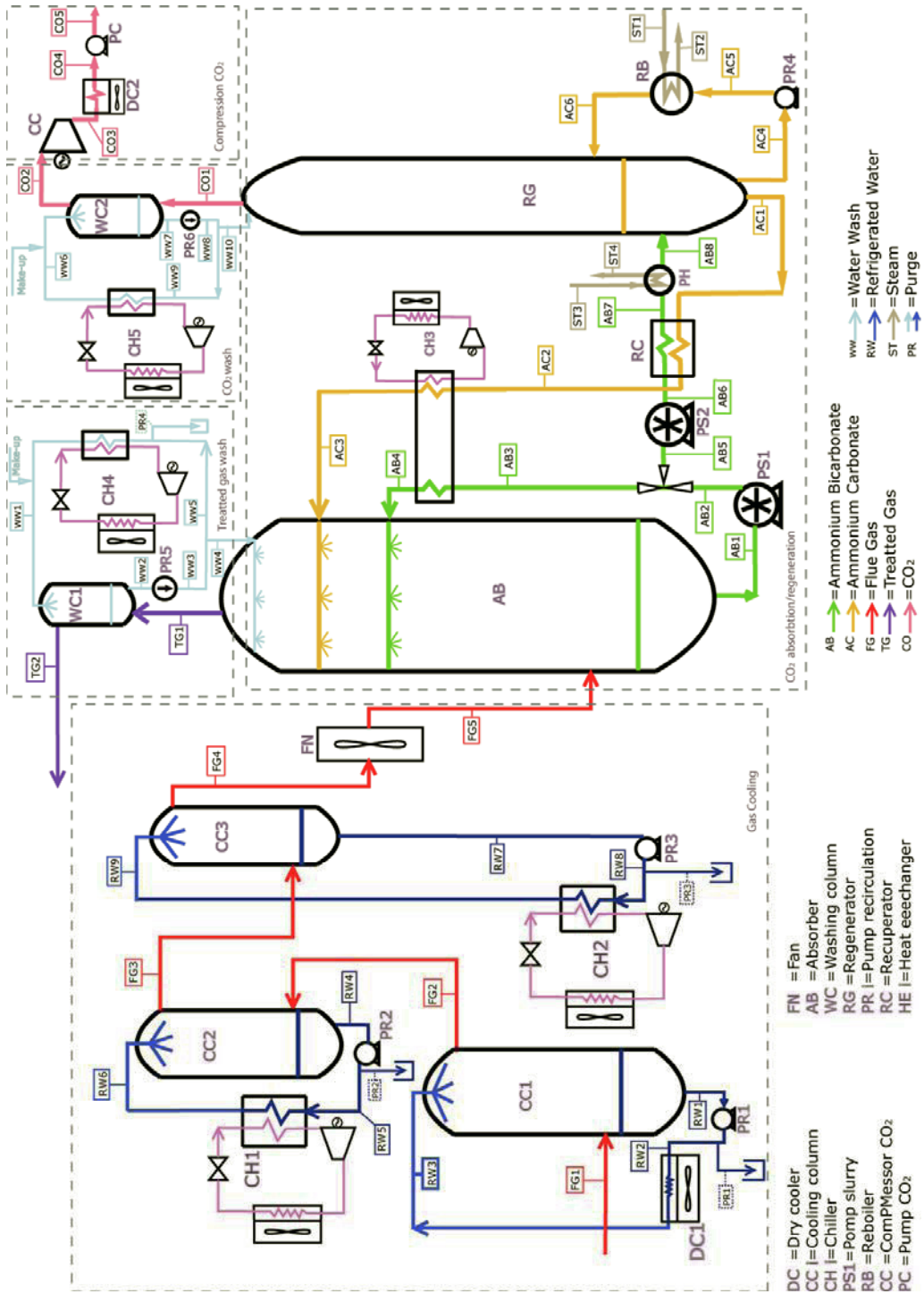


Fig. 1. Schematic of the process analyzed in this work and simulated from first- and second-law points of view.

Tab. 1 – List of the main parameters assumed for the simulation of the process illustrated in Fig. 1.

Ambient		Dry coolers	
Pressure, bar	1.01	Reference thermal power, kW	727
Temperature, °C	15	Reference ΔT inlets, °C	15
Flue gases		Reference electrical power, kW	5.94
Pressure, bar	1.025	ΔT minimum cold side, °C	5
Temperature, °C	60	Chillers	
Ar mol%	0.43	ΔT condensation, °C	10
CO ₂ mol%	11.27	ΔT evaporation, °C	3
H ₂ O mol%	saturated	COP / COP of Carnot cycle	0.65
N ₂ mol%	64.55	Recirculation pumps	
O ₂ mol%	4.24	Hydraulic efficiency, %	80
Reboiler steam		Electro-mechanical efficiency, %	90
Pressure, bar	3.5	Head pressure (PR1,PR2,PR3,PR5), bar	2.50
Temperature, °C	330	Head pressure (PR4,PR6), bar	42.50
Pre-heater steam		Slurry pumps	
Pressure, bar	1	Hydraulic efficiency, %	75
Temperature, °C	200	Electro-mechanical efficiency, %	95
Steam turbine		Head pressure (PS1), bar	5
Adiabatic efficiency, %	90	Head pressure (PS2), bar	40
Electro-mechanical efficiency, %	98	Carbon dioxide pump/compressor	
Exhaust pressure, bar	0.05	Hydraulic/adiabatic efficiency, %	85
Exhaust vapor quality, %	95	Electro-mechanical efficiency, %	92
Absorption reactor		Head pressure (CC/PC), bar	80/150
NH ₃ concentration initial solution, wt%	11.5	Cooling and washing columns	
Capture efficiency, %	90	ΔP , bar	0.015
CO ₂ loading, kg_CO ₂ / kg_solution	0.10	Hot stream outlet temp (CC1), °C	23
Extracted / regenerated flows, kg / kg	6	Hot stream outlet temp (CC2,WC2), °C	15
Recycle and lean sol inlet temp, °C	10	Hot stream outlet temp (CC3,WC1), °C	6
Regeneration reactor		ΔT cold side, °C	3
Pressure, bar	40	ΔT hot side (CC1), °C	5
Temperature, °C	120	ΔT hot side (CC2), °C	4
Heat exchangers		ΔT hot side (CC3,WC1), °C	3
ΔT min, °C	5	ΔT hot side (WC2), °C	95

Tab. 2 – Steam extraction and loss of electrical production from the steam turbine. All values are given specific to the kilogram of captured CO₂.

	Heat duty MJ / kg CO ₂	Steam extraction kg steam/kg CO ₂	Electrical loss kWh/kg CO ₂
Pre-heater	0.5813	0.2283	0.0271
Reboiler	0.9535	0.3661	0.0688
Total	1.5348	0.5944	0.0959

Tab. 3 – Electrical penalty due to auxiliaries, loss of production from turbine and work increase with respect to ideality due to irreversibilities.

	Electrical penalty		Delta work w/t ideality	
	kWh/kg CO ₂	%	kWh/kg CO ₂	%
Gas cooling	0.0207	20.75	0.0685	38.97
CO ₂ absorption/regeneration	0.0579	57.98	0.0796	45.33
Treated gas wash	0.0039	3.95	0.0051	2.89
CO ₂ wash	0.0039	3.86	0.0109	6.18
CO ₂ compression	0.0134	13.47	0.0077	4.41
Purge discharge	0.0000	0.00	0.0021	1.22
Treated gas discharge	0.0000	0.00	0.0017	0.99
Total auxiliary electrical usage	0.0998	100.00	0.1757	100.00
Total loss from steam turbine	0.0959			
Total electrical penalty	0.1957			

6. Conclusions

The CAP appears to be a promising technology. Compared to MEA, the present simulation confirms it has a reduced reboiler heat duty and a moderate electrical need for auxiliaries, in particular chillers: for 1 kg of captured

CO₂, a steam extraction of 0.59 kg, equivalent to a heat duty exceeding 1.5 MJ and a generation loss approaching 0.1 kWh, an auxiliary consumption of 0.1 kWh and an increase of work with respect to ideality of 0.18 kWh. The total electrical penalty is 0.2 kWh so, at a cost of electricity of 7 c€/kWh, the sole operation adds about 14 € per captured ton of CO₂. Referring to a modern PC-USC with net electrical output of 600 MW and efficiency of 45.0%, with respect to a coal containing 62wt% of C and having a LHV of 25 MJ/kg, the decrease in output is 76.9 MW, which reduces the efficiency to 39.2%, penalizing the performance less than most of the competing techniques.

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