ANTAL KERPELY DOCTORAL SCHOOL OF MATERIALS SCIENCE & TECHNOLOGY

Carbon Capture and its Energy Intensification for Emission Reduction in Industrial Sectors

A Ph.D. dissertation submitted to Antal Kerpely Doctoral School of Materials Science & Technology for the degree of Doctor of Philosophy in the subject of Materials Science and Technology

by

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Summary

This thesis explores carbon capture and energy intensification technologies as key strategies to mitigate CO_2 emissions from industrial sectors. It assesses the feasibility, efficiency, and economic viability of carbon capture processes, aiming to reduce energy consumption in chemical manufacturing. The research employs a combination of theoretical analysis, case studies, and techno-economic assessments to provide insights into the practical implementation of carbon capture and energy-efficient solutions.

The study focuses on two primary carbon capture technologies: air-combustion and oxyfuel-combustion. It compares their regeneration duties across five different industrial flue gases, including those from power plants, aluminum production, and cement manufacturing. A thermodynamic evaluation further examines the efficiency of these processes in gas-fired power systems, using exergy analysis to identify optimal points for energy integration, heat recovery, and process improvement.

Finally, a techno-economic assessment contrasts the power demands of a water-based carbon capture system with a conventional MEA solution. The findings emphasize the need to optimize carbon capture alternatives to balance emission reduction and economic feasibility, guiding the selection of the most effective approach for industrial applications.

1. Introduction

The issue of global warming stems from the continual rise in the concentration of greenhouse gases (GHG) within Earth's atmosphere, presenting a significant environmental concern worldwide [1,2]. Key gas contributing to this phenomenon is carbon dioxide. This gas accumulates in the atmosphere and creates a barrier that traps heat emitted from the sun, a phenomenon commonly referred to as the greenhouse effect [3–5]. The greenhouse effect plays an important role in maintaining Earth's livable temperatures, ensuring conditions suitable for life as we know it. However, human activities such as fossil fuel combustion, deforestation, and industrial processes have markedly increased the presence of greenhouse gases in our atmosphere [7,8]. As depicted in Figure 1, atmospheric CO_2 concentrations surged from approximately 335 parts per million (ppm) in 1980 to almost 425 ppm in 2024. Projections indicate a continuation of this trend, with CO_2 concentrations expected to reach 550 ppm by 2050 [6]. This relentless rise underscores the urgency for concerted efforts to mitigate emissions and transition toward sustainable energy alternatives to mitigate the intensification of climate change [7,8].

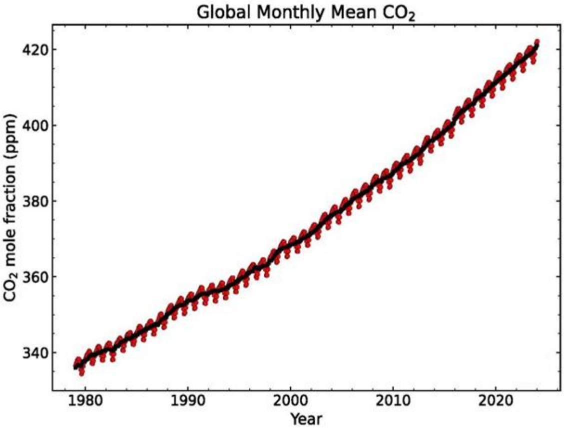

Figure 1. Atmospheric carbon dioxide emissions [6].

1.1.The Aims of this work

This thesis aims to assess the feasibility, efficacy, and economic viability of integrating carbon capture technologies into chemical manufacturing operations with the primary goal of reducing the industry's carbon footprint and energy demands.

The study examines various strategies for energy optimization, such as heat integration and the adoption of energy-efficient designs, to minimize energy consumption within industrial sectors and, consequently, decrease carbon emissions.

1.2.Approach

This work is divided into three different cases as follows:

Case 1: Carbon capture evaluation of air-combustion and oxyfuel-combustion.

Case 2: Thermodynamic evaluation of heat-integrated carbon capture process for oxyfuel and air combustion systems.

Case 3: Techno-economic analysis of water-based and amine-based carbon capture in air-combustion and oxyfuel-combustion technologies.

The basis of this study lies in the examination of flue gases for two air and oxyfuel-combustion technologies through the evaluation of energy-intensification methods. Employing the process modeling software ASPEN Plus, all aspects of model building, verification, and analysis were conducted. Carbon Capture Utilization or Storage (CCUS)

Three different types of CO_2 capture technology are demonstrated in Figure 2. The concentration of CO_2 in the gas stream, the pressure of the gas stream, and the fuel type (solid or gas) are important factors in selecting the capture system.

Post-combustion capture of CO_2 in power plants is economically feasible under specific conditions [9]. It is used to capture CO_2 from part of the flue gases from some existing power plants.

The technology required for pre-combustion capture is widely applied in fertilizer manufacturing and hydrogen production. Although the initial fuel conversion steps of pre-combustion are more elaborate and costly, the higher concentrations of CO_2 in the gas stream and the higher pressure make the separation easier.

In the case of oxyfuel-combustion there can be two options [10]:

- Clean oxygen is used; in this case, the flue gas contains only CO_2 and water, and no selective carbon capture is required.
- Oxygen with artificially mixed CO_2 as inert is used; this alternative is for existing air-based technologies where the inert effect of nitrogen is replaced with the application of CO_2.

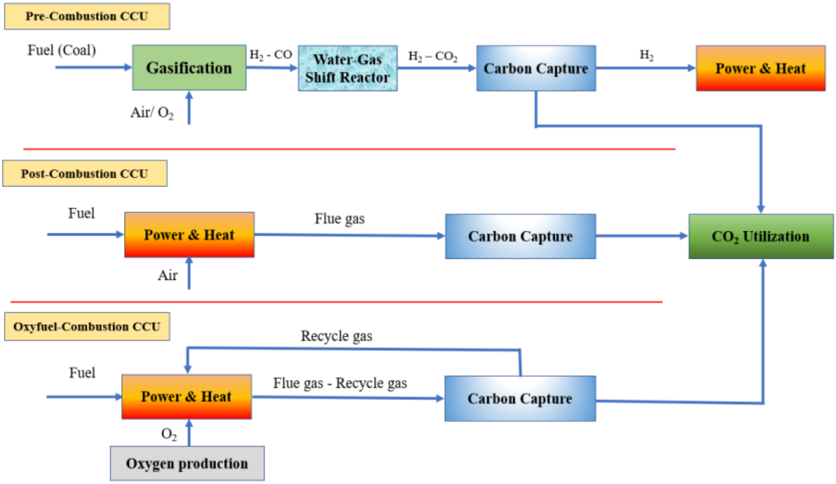

Figure 2. Schematic representation of capture systems [10].

2.Methodology

To model the absorber-stripper system in my simulation, I employ a rate-based model, which proves more suitable than the equilibrium phase model [11]. In the liquid phase, the presence of dissolved CO_2 in various chemical forms such as carbonate and carbamate induce electrolyte behavior. Thus, the electrolyte NRTL (Non-Random Two-Liquid) thermodynamic model is employed in the liquid phase. Conversely, in the vapor phase, the presence of ions is typically negligible, simplifying the thermodynamic description. Here, the SRK (Soave-Redlich-Kwong) equation of state (EoS) is chosen to characterize the behavior of the vapor phase.

2.1. Configuration of case 1

Figure 3 illustrates the carbon capture process employed in this study, with its accuracy verified against experimental data by

Nagy and Mizsey [11]. The 120 °C flue gas after the pretreatment section should be cooled to 40 °C before reaching the absorber column, then it enters the absorber column from the bottom and contacts with the countercurrent lean MEA solution from the top of the absorber. The rich MEA, destined for regeneration, is directed into the stripper, obtaining heat input from it reboiler.

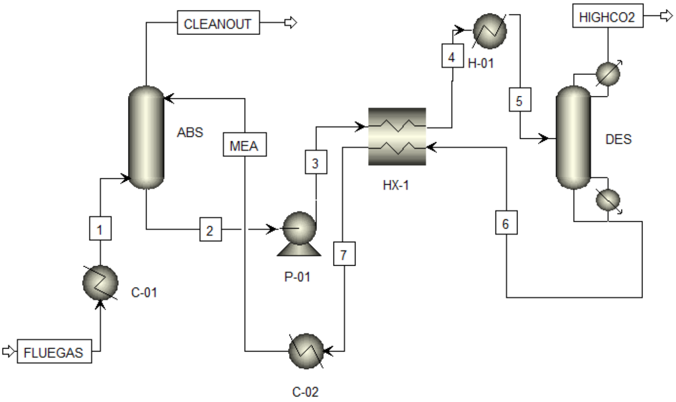

Figure 3. Carbon capture process based on chemical absorption technology.

2.2.Configuration of case 2

In this part of my study, I explored two distinct process configurations, namely a non-integrated configuration (C.1) and an integrated configuration (C.2), each illustrated in Figure 4.

In the integrated configuration, a heat-integrated system is implemented. Here, the rich MEA absorbent, laden with captured CO_2, is brought into contact with the lean but hot absorbent in a dedicated heat exchanger. This thermal exchange optimizes the overall efficiency of the capture process by leveraging the temperature gradient between the two streams.

To ensure the effectiveness of the integrated system, auxiliary heat exchangers are strategically employed. These heat exchangers play a critical role in maintaining the optimal input temperatures of the absorbent flows, thereby enhancing the performance and reliability of the CO_2 capture process.

In contrast, the non-integrated configuration operates without the integration of heat exchange between the rich and lean absorbent streams. Consequently, the thermal energy present in the hot lean absorbent is not utilized to preheat the incoming rich absorbent. This approach leads to inefficiencies and higher energy consumption compared to the integrated configuration.

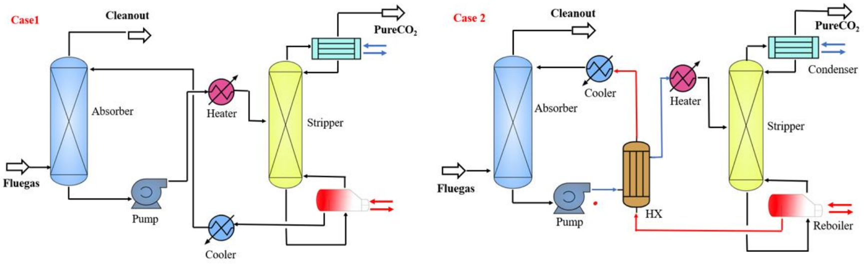

Figure 4. Different configurations of carbon capture process; (C.1) non-integrated and (C.2) integrated.

2.3.Configuration of case 3

In this case, I conducted a techno-economic evaluation comparing the power demands of novel water-based carbon capture in two different strategies with those of a conventional aqueous MEA solution, as employed in previous cases to achieve 90% CO_2 removal from gas-fired flue gas employing both MEA and H_2O as solvents. as follows:

- MEA-CO_2 capture, which used a chemical MEA solution in a chemical absorption process.

- H_2O-CO_2 capture (strategy 1), in which CO_2 is captured using water as a physical solvent.
- H_2O-CO_2 capture + gas dehydration (strategy 2), in which flue gas dehydration occurred first, followed by CO_2 capture using water as a physical solvent.

A typical amine-based CO_2 absorption system is the same as former cases (Figure 3).

The H_2O-CO_2 capture process is depicted in Figure 5, contains a series of steps. Initially, the flue gas pressure, operating at 120°C, increases through a 2-stage compression to elevate it to 26 bar. To prevent overheating within the compressor, the outlet stream underwent initial cooling within a heat exchanger, followed by further cooling to 30°C post the second compression stage. Subsequently, the cooled stream entered a counter-current 15-stage absorber, adhering to an equilibrium-based model. Water (H_2O) entered the absorber at 25.6 bar and 5°C from above, with its mass flow rate adjusted according to design specifications until approximately 90% of CO_2 was captured within the absorber. The resultant high-pressure clean gas (CL-GAS) exited from the top of the absorber. The CO_2-rich water, exiting the absorber at the bottom, maintained a pressure of 26 bar and a temperature of 30°C. Subsequently, its pressure underwent a drop to 2 bar within the turbine (T-01), facilitating electricity generation. Within the stripper, operating at 2 bar and 30°C, a high portion of absorbed N_2 and O_2 transitioned into the gas phase.

The CO_2-containing water's pressure was further reduced to 1.1 bar through a two-phase separation process (SEP) to segregate CO_2 from water. The water was then reintroduced into the cycle, with its pressure increased to 25.9 bar via a pump (P-01) and its temperature reduced to 5°C via a cooler (C-02).

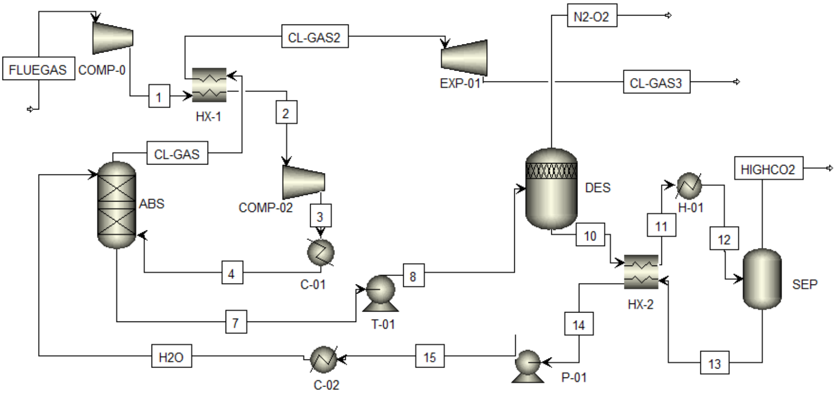

Figure 5. Process diagram of water-based carbon capture approach.

To enhance H_2O-CO_2 capture process, gas dehydration upstream integrates with the initial proposed H_2O-CO_2 capture. The schematic representation of the flue gas dehydration process is delineated in Figure 6. Initially, the flue gas, sourced from the power plant undergoes a cooling process to reach near-ambient temperatures of 30°C within a dedicated cooler. Subsequently, a two-phase separation, known as a Flash system, is initiated. The CO_2-rich stream resulting from this separation is channeled into the subsequent capture process.

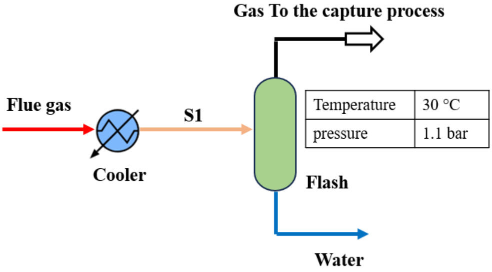

Figure 6. Schematic diagram of flue gas dehydration process.

In Figure 7, the H_2O-CO_2 capture process is illustrated with a particular focus on the stage of gas dehydration. The red dotted box serves to emphasize the significance of the dehydration phase, strategically positioned before the compression stage. This deliberate arrangement is designed to optimize the overall efficiency of the capture process. Following the dehydration process, carbon capture occurs under the same conditions as explained for the former configuration (Figure 6).

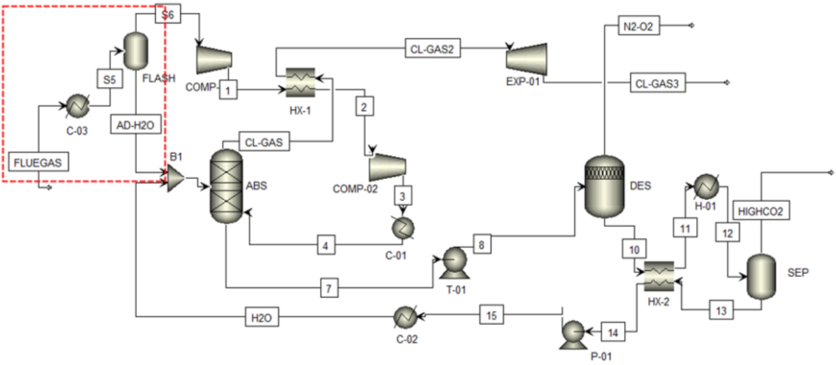

Figure 7. Process diagram of carbon capture by pressurized water + gas dehydration.

3. Scientific Theses

Thesis 1

I confirmed , based on ASPEN modelling, that oxyfuel-combustion technologies have significant savings of up to 84 % regeneration energy requirement at carbon capture rates above 90 % than that of the air-combustion alternatives. I also confirmed that 100 % carbon capture is also possible. Moreover, the energy requirement is linearly increasing with the capture rate as long as it is exponential in the case of air-combustion alternative. I demonstrated that the reason is the higher mass transfer driving force for CO_2 capture.

Thesis 2

I demonstrated the energy and exergy of carbon capture processes in the cases of air-combustion and oxyfuel-combustion technologies for both heat-integrated and nonintegrated configurations. I confirm that in the case of a heat-integrated configuration, the installed heat exchanger. It leads to about 3 times lower energy consumption as well as reduction in exergy destruction by almost 35% for oxyfuel-combustion technology and around 40% for air-combustion alternative below the 90% CO_2 removal rates. However, in 95% CO_2 removal for air-combustion, there is only about 25% reduction in exergy destruction.

Thesis 3

Based on my thermodynamic investigations, I confirmed that implementing oxyfuel technology increases the thermodynamic efficiency by 10 - 50 % of the capture process due to the higher mass transfer driving force for CO_2 capture.

Thesis 4

Comparing the application of water and MEA absorbents, I concluded that in the cases of both air- and oxyfuel-combustion technologies, the total utility demands of the capture method with water exceeds that of the capture method with MEA solvents by approximately 30 – 45 %.

Thesis 5

I concluded that the total utility demands of the water-based alternative can be significantly reduced with dehydration of the flue gas and that becomes lower than that of the MEA alternative. Moreover, utilizing a water-based carbon capture system with the application of gas dehydration the capture process requires mainly electricity. The application of this system might be considered if the electricity is relatively cheap.

4.List of publications

4.1. Scientific publication

1- Talei S, Szanyi A, Mizsey P. Improving the energy efficiency of carbon capture process: The thermodynamic insight. *ENERGY*. 2024 August 26. **Q1 (IF:9).**

2- Talei S, Szanyi A, Mizsey Comparison of Water- and Amine-Based Carbon Capture Processes for Air and Oxyfuel Combustion Technologies. *I&EC Research*. 2024 September 10. **Q1 (IF:3.9).**

3- Talei S, Fozer D, Varbanov PS, Szanyi A, Mizsey P. Oxyfuel Combustion Makes Carbon Capture More Efficient. *ACS omega*. 2024 January 10. **Q1 (IF:4).**

4- Saeed Talei, Peter Mizsey Investigation of utility demand in CO_2 liquefaction amine-based carbon capture, *PhD Students*

Almanach, University of Miskolc, Hungary, volume 1, 2024, ISSN 2939-7294.

5- Saeed Talei, Peter Mizsey. Simulation of Carbon capture process with MEA solution. *Symposium on Polyurethane Innovation 2022 (SPI)*, University of Miskolc, Hungary, 2022, ISBN 978-963-358-254-1.

6- Saeed Talei, Peter Mizsey. Comparison of Carbon Capture process for Coal-fired and Gas-fired Power Plants, *PhD Students Almanach*, University of Miskolc, Hungary, volume 1, 2022, ISSN 2939-7294.

7- Talei, Saeed, Rachid Hadjadj, Péter Mizsey, and Michael C. Owen. "Analysis of Molecular Dynamics Simulation of Carbonic Anhydrase." *Hungarian Materials and Chemical Sciences and Engineering* 47, no. 1 (2023): 109-117.

4.2.Oral and Poster Presentations

1- Saeed Talei, Peter Mizsey. Comparison of CO2 capture alternatives: MEA or Water. A Symposium on Polyurethane Innovation (SPI) 2024, Miskolc, Hungary, October 9, 2024.

2- Saeed Talei, Peter Mizsey. Oxyfuel technology for more efficient use of electric cars. 27th Conference on Process Integration, Modelling, and Optimization for Energy Saving and Pollution Reduction, Xian, China, August 25-28, 2024, Online presentation.

3- Saeed Talei, Peter Mizsey. Comparison of Carbon Capture for Air-combustion and Oxyfuel-combustion Technologies, 38th Borsod Chemical Industry Day, Miskolc, Hungary, November 17, 2022, Presentation.

4- Saeed Talei, Peter Mizsey Comparison of Carbon Capture for Air-combustion, and Oxyfuel-combustion Technologies.

XXVI. Spring Wind Conference. Miskolc, Hungary, May 5-7, 2022, Poster.

5- Saeed Talei, Peter Mizsey. Analysis of Three Main Negative Emission Technologies; Bioenergy with Carbon Capture and Storage, Absorption and Adsorption of Atmospheric Carbon Dioxide.26th Conference on Process Integration, Modelling, and Optimization for Energy Saving and Pollution Reduction, Thessaloniki, Greece, October 8-10, 2023, Online presentation.

5. References

[1] Azadi M, Northey SA, Ali SH, Edraki M. Transparency on greenhouse gas emissions from mining to enable climate change mitigation. Nat Geosci 2020;13:100–4.

[2] Metz B, Davidson O, De Coninck HC, Loos M, Meyer L. IPCC special report on carbon dioxide capture and storage. Cambridge: Cambridge University Press; 2005.

[3] Yang Y, Wu L, Guo Y, Gan B, Cai W, Huang G, et al. Greenhouse warming intensifies north tropical Atlantic climate variability. Sci Adv 2021;7:eabg9690.

[4] Keith DW, Holmes G, Angelo DS, Heidel K. A process for capturing CO2 from the atmosphere. Joule 2018;2:1573–94.

[5] Talei S, Soleimani Z. Comparative Analysis of Three Different Negative Emission Technologies, BECCS, Absorption and Adsorption of Atmospheric CO. Civil and Environmental Engineering Reports 2021;31:99–117.

[6] Change C. Atmospheric Carbon Dioxide. NOAA Climate Gov Available Online: Www Climate Gov (Accessed on 16 November 2020) 2020.

[7] Oberthür S, Khandekar G, Wyns T. Global governance for the decarbonization of energy-intensive industries: Great potential underexploited. Earth System Governance 2021;8:100072.

[8] OECD / IEA. World Energy Model Documentation 2020 Version. 2020.

[9] Zhang S, Shen Y, Wang L, Chen J, Lu Y. Phase change solvents for post-combustion CO2 capture: Principle, advances, and challenges. Appl Energy 2019;239. https://doi.org/10.1016/j.apenergy.2019.01.242.

[10] Talei S, Fozer D, Varbanov PS, Szanyi A, Mizsey P. Oxyfuel Combustion Makes Carbon Capture More Efficient. ACS Omega 2024.

[11] Nagy T, Mizsey P. Model verification and analysis of the CO2-MEA absorber–desorber system. International Journal of Greenhouse Gas Control 2015;39:236–44.