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Simulation studies of the solvent diethylamine for post-combustion CO_2 capture at the WACEM cement manufacturing plant in southern Togo at Tabligbo

Solagnon Edoh Koevidjin[1], Yawovi Nougbléga[1,2*], Yawovi Mignanou Amouzouvi[3], Milohum Mikesokpo Dzagli[2,3], Messanh Agbéko Mohou[3]
[1]Solar Energy Laboratory/ Group of Transfer Phenomenon and Energetic, University of Lomé; Lomé P.O. Box 1515, Togo.
[2]Regional Centre of Excellence on Electricity Management; Lomé P.O. Box 1515, Togo; nycogl@yahoo.fr (Y.N.).
[3]Laboratory for the Physics of Materials and Semiconductor Components'Physics (LPMCS), University of Lomé,; Lomé P.O. Box 1515, Togo.

Abstract: Industrial CO2 emissions continue to rise despite global reduction efforts, driving climate change and global warming. Post-combustion carbon capture using aqueous diethylamine (DEA) is a promising strategy to mitigate these emissions. This study aims to simulate CO2 absorption from the Western African Cement (WACEM) industry's flue gases using the Hysplit model. The tray column's key parameters, the gas-liquid mixture's thermophysical properties, and the treated emissions' composition were investigated. Results indicate optimal CO2 absorption occurs when the flue gas mass flow rate does not exceed 12.5% of the liquid mixture entering the column. The process is most effective at a DEA flow rate of 250 L/h, with a furnace temperature of 160°C and a pressure of 17 bars. These findings provide valuable insights for policymakers and industry stakeholders in optimizing post-combustion carbon capture for emission reduction.
Keywords: *Chemical absorption, Diethylamine, Hysplit, Post-combustion CO2 capture, Tray efficiency.*

1. Introduction

Air quality has recently become a significant public health concern in urban areas [1, 2]. Over the past century, human activities have gradually increased the atmospheric concentration of greenhouse gases such as CO_2, methane, nitrous oxide, and chlorofluorocarbons [3]. Anthropogenic activities including automobile traffic, industrial processes, and rapid urbanization, are identified as the primary sources of gaseous and particulate emissions into the air, with high concentrations observed in urban environments [1]. Industrial, agricultural, and human activities pose issues to the balance of ecosystems, including groundwater, soil, plants, animals, and human populations. Anthropogenic activities are considered the major source of pollutants released into the atmosphere that directly impact climate change and human health [4]. These activities contribute to ecosystem degradation, with ambient air pollutants including gases such as nitrogen oxide, ozone, sulfur dioxide, and organic compounds, as well as particles of varying sizes like PM10 and PM2.5 suspended in the atmosphere [5, 6]. Carbon capture, utilization, and storage are the most effective and efficient ways to control CO_2 emissions [7]. There are various methods for capturing CO_2 such as the chemical absorption method which is the most used for CO_2 capture using a usually amine-based chemical solvent; the post-combustion-CO_2-capture technology, which aims to absorb CO_2 from the flue gases of power plants via an amine-scrubbing process [8-11]. The amine-based chemical absorption approach has achieved the highest level of maturity in post-combustion CO2 capture [4]. Amine-based carbon capture technology has successfully been employed to capture CO_2 from flue gases with a low CO_2 concentration [12]. This

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* Correspondence: nycogl@yahoo.fr

technology has been implemented in large-scale power plants, cement plants, and other sectors with significant carbon emissions [13]. Capture of carbon dioxide has taken center stage globally due to the increasing adverse effects of CO_2 emissions. These emissions are generated from anthropogenic activities using fossil fuels for electric power generation, transportation, and heating/cooling purposes in residential and office buildings [11, 14]. Based on total global emissions, coal, and crude oil emitted the most CO_2 compared to natural gas [15]. Also, through the use of these fossil fuels, the generation of electricity was the sector that generated the most CO_2 emissions. Due to the relatively cheap cost and global availability, coal will most likely be the preferred fossil fuel for electricity production in the coming decades, constituting the highest source of CO_2 emissions [16, 17]. Hence, capturing the CO_2 from fossil fuel power generation plants is imperative to limit its adverse effects. The process of combusting fossil fuels for electric power generation and removing CO_2 afterward before releasing the exhaust gas can be classified into post-combustion, pre-combustion, and oxyfuel combustion CO_2 capture [15, 18]. In post-combustion CO_2 capture, the flue gas is produced from combusting fossil fuels with air for power generation as seen in Equation (1) [19].

$$\mathrm{CxHy} + z(O_2 + 3.76N_2) \rightarrow aCO_2 + bH_2O + cO_2 + dN_2 \quad (1)$$

Where 'z' is the stoichiometric coefficient of air. The stoichiometric coefficients of the products (a, b, c, d) will depend on those of the reactants (x, y, z).

The combustion reaction in Equation (1) produces mainly nitrogen (N_2), CO_2, water (H_2O), and unreacted oxygen (O_2). The flue gas CO_2 concentration from this combustion process is usually between 10 and 15% for coal-fired power plants and 3-8% for natural gas-fired power plants [20]. However, other unwanted gases are produced (SOx, NOx, fly ash, metals, etc.) due to impurities in fossil fuels. These impurities and O_2 often lead to amine solvent degradation [21]. Therefore, these impurities must be removed to the lowest concentrations before capturing CO_2. The CO_2 capture efficiency is usually targeted at 90% [22]. The capture of CO_2 is also driven by advancements in CO_2 utilization routes like CO_2 to gaseous and liquid fuels, CO_2 to chemicals and polymers, and CO_2 for enhanced oil recovery [23, 24]. In all combustion processes that require CO_2 capture, the separation of CO_2 can be achieved by various techniques including absorption, adsorption, membrane, and cryogenic processes [25]. However, CO_2 absorption using amine-based chemical solvent has attracted the most attention due to its maturity, cost-effectiveness, and ability to handle large volumes of flue gas streams [26]. The increase in greenhouse gas emissions, especially CO_2, constitutes a global issue because of its dangerous effects on the climate and the environment. For this crucial CO_2 emissions reduction, a solution is to capture carbon dioxide by a post-combustion process using two coupled columns [27]. Numerous previous studies have indicated that air pollution is linked to a substantial number of premature deaths and respiratory, and cardiovascular diseases. So, a significant number of global deaths are associated with indoor and outdoor air pollution [28, 29]. In Togo, researchers have identified air pollution as a leading cause of premature death annually, surpassing unsafe drinking water and malnutrition. Unfortunately, air quality monitoring networks are largely absent. The primary sources of pollution in Togo are attributed to road traffic, industrial activities, domestic fires, other human-induced sources, and natural sources [30, 31]. While Togo is experiencing significant economic growth, most of these activities are concentrated in urban areas. The country possesses substantial deposits of high-quality limestone for clinker production, a crucial component in cement manufacturing. This intensive production poses significant risks and environmental damage, further exacerbated by large-scale mining permits for a limestone deposit in Tabligbo, Yoto Prefecture [32, 33]. The transformation of these deposits has major environmental drawbacks, impacting air, water, and soil and the well-being of workers, visitors, and local populations. Togo currently hosts four cement factories (CIMTOGO, FORTIA, DIAMOND CEMENT, and CIMCO), collectively producing 3 to 4 million tons of cement annually, with an estimated CO_2 production of approximately 814.136 Ggt in 2015 [34]. This is likely to increase with rising demand and the establishment of new industries. The cumulative industrial activities in Togo have significant implications for humans and the environment, with limited research in this area to address the associated damages [35, 36]. Various techniques have been identified for capturing CO_2 emissions from manufacturing and processing industries, such as post-combustion capture, which involves capturing CO_2 from furnace flows and exhaust gases after combustion [14, 37, 38]. Reference molecules like monoethanolamine (MEA), diethanolamine (DEA), N-methyl diethanolamine (MDEA), piperazine (PZ), and 2-amino-2-methylpropan-1-ol (AMP) are used as industrial solvents for CO_2 capture due to the solubility of CO_2 in aqueous amine solutions [39, 40]. CO_2 capture and storage remain effective strategies for mitigating the greenhouse effect by reducing cumulative CO_2 emissions [41, 42]. Aqueous diethanolamine (ADA) is the reference solvent for this technology, while biphasic solvents show promise in CO_2 capture due to their higher CO_2 capacity and a significant reduction in regeneration energy [43]. Some authors have simulated the post-combustion CO_2 capture with Aspen Hysys software in the same conditions (cement flue gases and MEA 30%) but considered other alternatives [44-46]. This work aims to simulate the capture of CO_2 released from the combustion kilns of the WACEM cement manufacturing plant in Tabligbo, southern Togo, using aqueous solutions of diethylamine (DEA), based on Aspen Hysys software (Hysys Process 2.2 version). To optimize the absorption of CO_2 from the flue gases by the DEA solution, data collected from WACEM's cement plants in Tabligbo (FORTIA and Diamonds Cement) have been utilized to design and simulate an absorption column for the CO_2 from cement plant fumes by the DEA using the HYSYS process 2.2 software.

2. Materials and Methods

2.1. Study Areas

The study was performed in the WACEM cement manufacturing industry in Tabligbo, Maritime region, south of Togo (latitude 6.582845, longitude 1.514997). The industry uses the combustion of clinker to manufacture cement. Figure 1 shows a photo of the cement plant and a Google map of the area. Tabligbo is a geologically rich area, primarily known for its sedimentary formations.

Figure 1.
Photo of the WACEM cement plant site

The region is dominated by limestone, clay, and marl sediments that have accumulated over time in a shallow marine environment. The limestones present are often exploited for cement production, which is one of the main industrial activities in the area. The marls and clays also play a key role in soil stability and are used in various industrial applications [47]. The region experiences a tropical climate with an average annual rainfall of about 1000 mm. The precipitation is distributed over two main seasons: March - June and September - November. This rainfall supports local agriculture, including the cultivation of maize, cassava, and other staple crops, while also impacting soil erosion, especially in areas where intensive agricultural practices are common [48]. The geology of Tabligbo, with its rich limestone deposits, has facilitated the establishment of the cement industry, a key sector for the local and national economy.

2.2. Presentation of the Simulation Software

Figure 2 presents the process flow diagram for the Hysys Process software. This model comprises a mixer designed to blend two inlet streams: a feed stream of water at 40°C and 20 bars with a flow rate of 1,000 kg/h, and a feed stream of DEA at the same temperature.

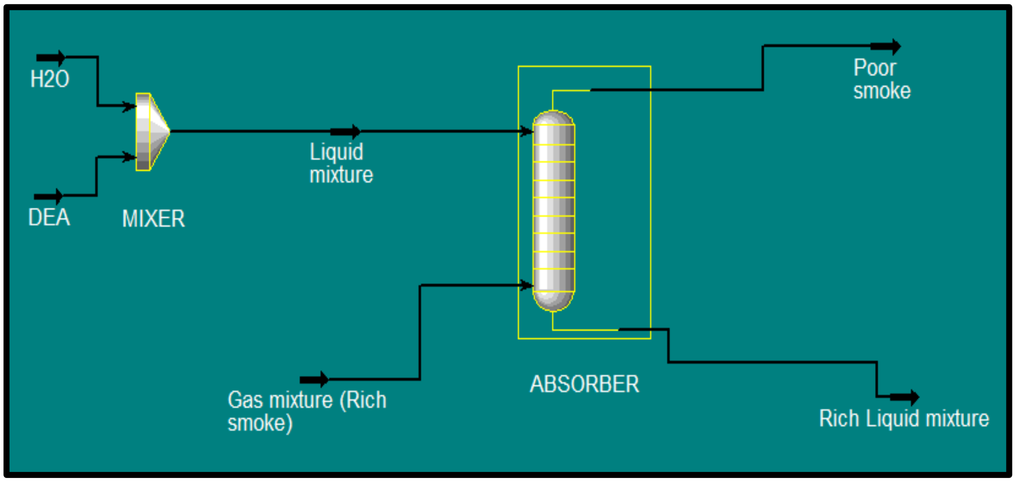


Figure 2.
HYSYS2.2 Process diagram flow.

The purpose is to produce a uniform mixture directed to the absorber column's head. Once the homogeneous liquid mixture with a constant temperature and mass flow rate is achieved, it is transferred to the top of the absorber column, which consists of ten (10) trays and features a pressure differential between the top and bottom of the column. The gaseous mixture (CO_2-rich smoke) is introduced at a constant temperature of 100°C and variable mass flow rates, allowing for a range of mass flow rates to be obtained for the smoke to be sent to the absorber to achieve maximum CO_2 absorption from the cement industry smoke. The Hysplit software was used with the geographical coordinates of WACEM to demonstrate the actual emission of particulate pollutants in the area.

2.3. Presentation of the Simulation Method

In this study, the Aspen Hysys software, (Hysys Process 2.2 version), is used to simulate the capture of CO_2 released from the clinker combustion kilns of the WACEM manufacturing plant. The simulation was based on the CO_2 capture using aqueous dimethylethanolamine (DEA) as the solvent [49]. Table 1 presents the various data on the rotary kilns at the WACEM cement plant in Tabligbo.

Table 1.
Data on WACEM rotary kilns at Tabligbo.

Data	Oven 1	Oven 2
Length (m)	86	85
Diameter (m)	5	5
Oven material inlet temperature (°C)	60	60
Oven material outlet temperature (°C)	1400	1400
Burner flame outlet temperature (°C)	2000	2000
Oven gas outlet temperature (°C)	450	450
The residence time of material in the cooler (min)	20	20
The residence time of material in the oven (h)	2	2
Oven rotation speed (rpm)	3,5 - 4,23	3,5 - 4,23
Material capacity (t/d)	6000 - 8000	6000 – 8000
Rate of inclination (furnace slope) %.	4	4
Main motor power (W)	800	800
Weight (t)	859-864	859-864

Data from the rotary kilns at the WACEM cement plant were used to calculate the different software parameters including the tray column height (12 m), the number of trays (10), the number of actual trays (17), the tray efficiency (0.58), the molar mass of the gas (29.15 g.mol^{-1}), the density of the gas and liquid mixture entering the column (4 kg.m^{-3}), the column diameter (1.5m), and the maximum gas velocity (0,12 m.s^{-1}), tray spacing (0.5 m), the density of the gas entering the column (1.13 kg.m^{-3}), the molar mass of the solvent entering the column (39.78 g.mol^{-1}).

2.4. Mixer

The mixer's role is to mix two flows at its inlet and send them to the absorber. A feed stream of water entering the mixer at 40°C and 20 bars is considered, with a flow rate of 1000 kg/h, and a feed stream of DEA entering at the same temperature. Table 2 illustrates the mixing conditions for the two streams, DEA and H_2O, and the composition of the resulting homogeneous liquid mixture, respectively. According to Table 2, the two streams mix, and the liquid mixture outlet stream passes through the absorber with a mass flow rate of 2000 Kg/h at a temperature of 39.98°C. This liquid mixture comprises approximately 80% H_2O and 20% DEA.

Table 2.
Composition of DEA, H_2O, and CO_2 absorption by the liquid mixture.

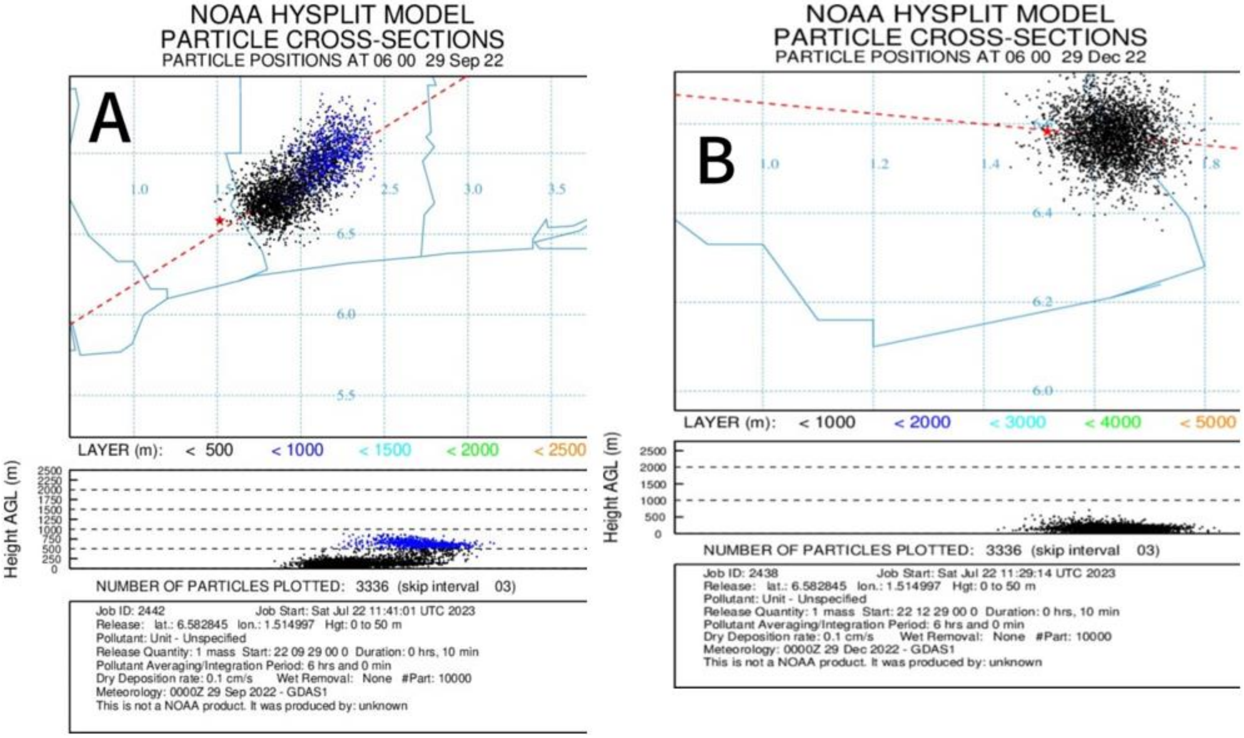
Name	DEA	H_2O	liquid mixture	Rich Smoke	Lean Smoke	Rich Mixture
Vapor	1.0000	1.0000	1.0000	0.9999	1.0000	0.0000
Temperature (F)	104.0	104.0	104.0	212.3	383.7	218.0
Pression (psia)	8.393	1.071	1.071	120.8	290.1	120.7
Molar flow (MMSCFD)	0.2745	1.114	1.389	0.1561	1.287	0.2578
Mass flow (Ib/hr)	2205	2205	4409	500	4399	510.2
LiqVol Flow (barrel/day)	213.5	151.3	364.8	41.44	371.2	35.01
Molar Enthalpy (10^4Btu/Ibmol)	-8.038	-10.33	-8.892	-36.07	-7.629	-11.99
Molar Entropy (Btu/Ibmol°F)	52.07	35.56	40.97	43.31	37.86	5.809
Heat flow (MMBtu/hr)	-0.9156	-12.65	-13.56	-0.6183	-10.78	-3.395

2.5. Absorber

In the absorption column, two flows meet in counter-current. The liquid mixture that exits from the mixer with a mass flow rate of 2000 kg/h at a temperature of around 40°C, is composed of DEA (20%) and H_2O (80%), and a gaseous mixture (rich smoke) composed essentially of CO_2 (14.45%), N_2 (71.93%), O_2 (1.45%), and H_2O (12.17%) entering the absorber at a temperature of 100°C at a lower flow rate than the liquid mixture. The absorption column consists of ten (10) trays, and there is a pressure difference between the top and bottom of the column.

3. Results and Discussion

Figure 3 presents the results of Hysplit simulations of pollutant emissions from the WACEM industry showing the vertical cross-sections of particle positions in the direction between September 29, 2022, and December 29, 2022. This emission manifests by a heap of polluting particles around the source, which spread through the zone. One can see the vertical dispersion of the particles at different times. The particles spread in high altitudes and far from the source in September. On the contrary, the particles stay around the source and are distributed within a layer of about 500 m above ground level in December, the winter period with dust clouds. Figure 4 presents the pressure, temperature, liquid, and vapor flow profiles through the trays.



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Release: lat.: 6.582845 lon.: 1.514997 Hgt: 0 to 50 m
Pollutant: Unit - Unspecified
Release Quantity: 1 mass Start: 22 09 29 00 0 Duration: 0 hrs, 10 min
Pollutant Averaging/Integration Period: 6 hrs and 0 min
Dry Deposition rate: 0.1 cm/s Wet Removal: None #Part: 10000
Meteorology: 0000Z 29 Sep 2022 - GDAS1
This is not a NOAA product. It was produced by: unknown

Job ID: 2438 Job Start: Sat Jul 22 11:29:14 UTC 2023
Release: lat.: 6.582845 lon.: 1.514997 Hgt: 0 to 50 m
Pollutant: Unit - Unspecified
Release Quantity: 1 mass Start: 22 12 29 00 0 Duration: 0 hrs, 10 min
Pollutant Averaging/Integration Period: 6 hrs and 0 min
Dry Deposition rate: 0.1 cm/s Wet Removal: None #Part: 10000
Meteorology: 0000Z 29 Dec 2022 - GDAS1
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Figure 3.
Position of emitted particles spotted using the Hysplit model corresponding to September and December 2022.

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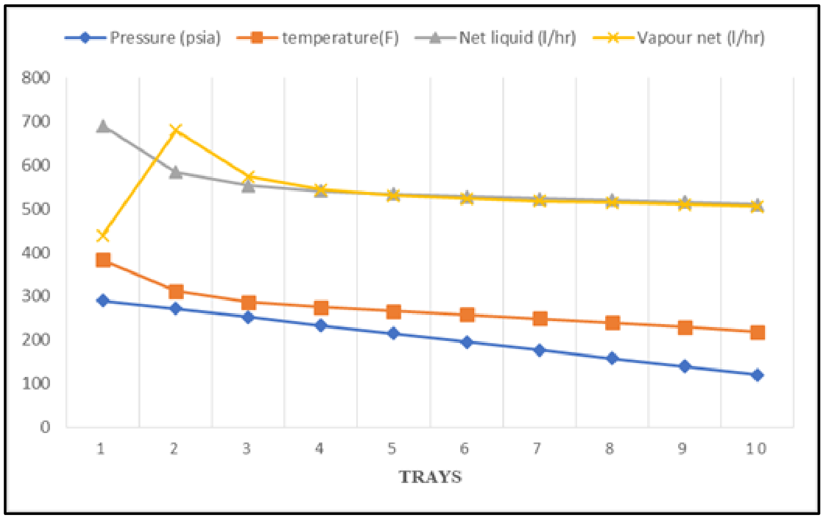

Figure 4.
Pressure and temperature profiles and liquid and vapor flow rates according to trays.

The pressure and the temperature profiles decrease as the number of trays is increased. This tendency means that increasing the number of trays led to their better cooling. Linear decreasing can be found for the pressure and temperature profile along the absorption column from the 3rd tray, which can be explained by the difference between the feed temperature and the tray temperatures [50]. Figure 4 shows also the profiles of vapor and liquid mass flow rates as a function of the number of trays. The evolution of these rates implies that beyond two plates, the quantities of gas and liquid are the same. The large slope of the gas mass flow rate indicates that for small numbers of racks, the flow rate of gas ejected into the environment is very high compared with the quantity of liquid flowing. The less condensed phase (gas) and the more condensed phase (liquid) flow identically for large numbers of racks proving the mixing of the two phases. The uniform and parallel evolution of liquid and vapor flow from the 3rd to the 10th trays may be due to the absence of a source of disturbance in the column.

Figure 5 illustrates the variation in the mole fraction of CO_2 emitted as a function of the variation in the trays. For small numbers of trays, the quantity of gas emitted is significant. CO_2 emissions decrease with the number of trays. The more there are trays, the less CO_2 is emitted in the WACEM cement plant in Togo.

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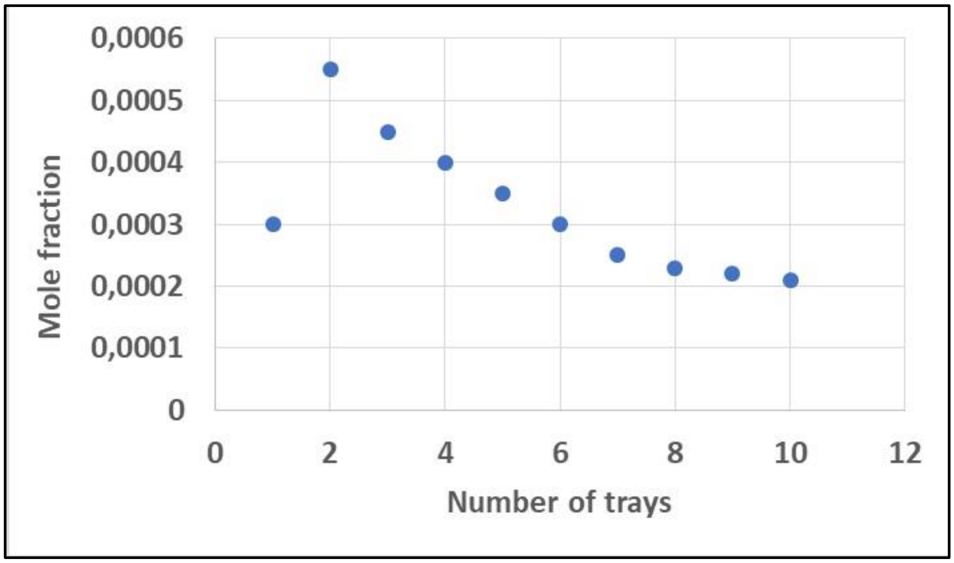

Figure 5.
Variation in mole fraction of CO_2 as a function of the number of trays.

An increase in the mole fraction of CO_2 absorbed by the DEA-containing liquid solution can be observed as the gas mixture rises at the beginning of the column before decreasing to a weak mole fraction near the 10th tray. This finding indicates that by increasing the number of trays, one can decarbonize the flue gases in the cement industry plants [51]. Figure 6 illustrates the CO_2 absorption rate as a function of the mass flow rate of the gas mixture. The finding shows that the absorption rate increases as the mass flow rate of the gas mixture decreases, and to approach total absorption, i.e. a zero-emission rate of CO_2, a mass flow rate in the range of 100 kg/h to 250 kg/h is required. It is possible to monitor the operating parameters of the plant's clinker combustion kilns, such as temperature, pressure, the DEA liquid produced, and the gas released into the kilns.

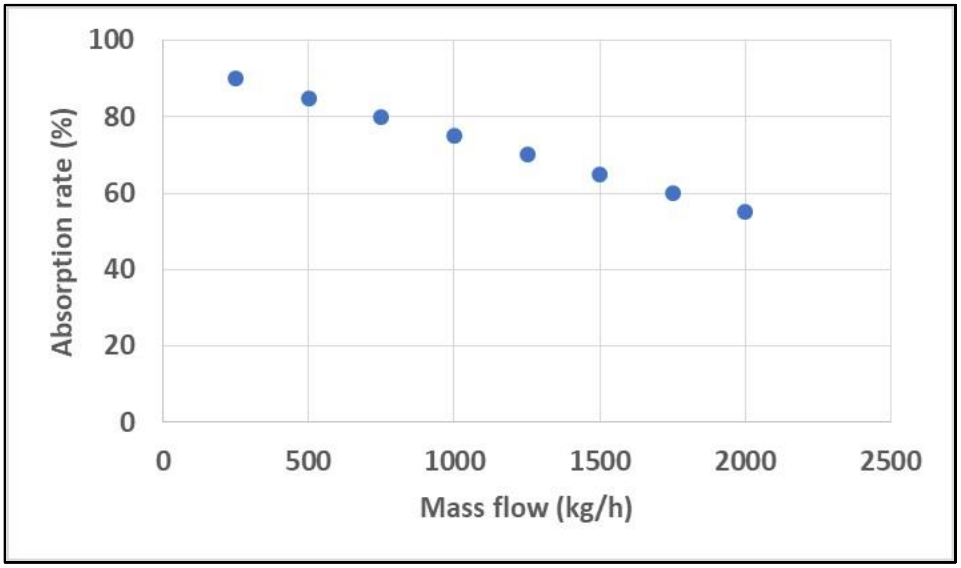

Figure 6.
CO_2 absorption rate as a function of gas mixture mass flow rate.

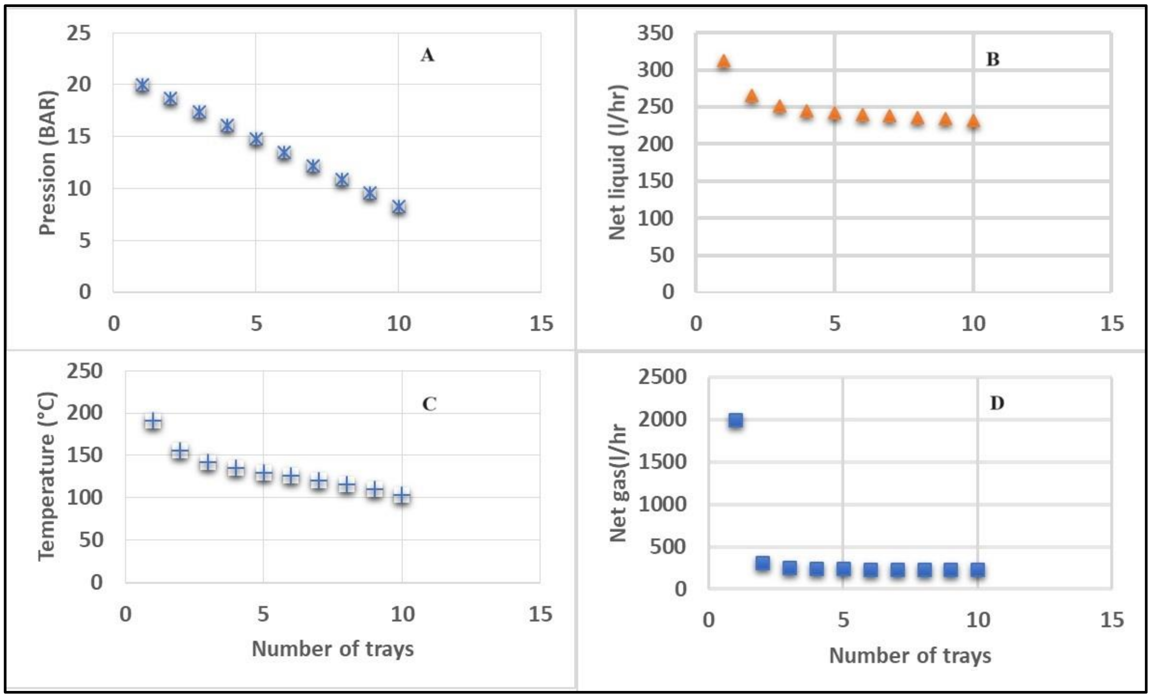

Figure 7.
Variation of pressure profile, temperature, net gas, and net liquid as a function of the number of trays.

To this end, a simulation was conducted to observe the progression of the DEA liquid within the system and the extent of gas absorption as a function of temperature and pressure. Figure 7 depicts the variation of pressure, temperature, net gas, and net liquid DEA profiles as a function of the number of trays; Figure 7a illustrates the increasing variation in furnace pressure, which ranges from a minimum of 8 bars to a maximum of approximately 20 bars for furnace temperatures between 100°C and 180°C and above. The data indicates that an increase in furnace temperature is correlated with an increase in pressure. Furthermore, the pressure within the rotary kiln demonstrates a progressive rise with temperature, reaching a point of stability above 180°C. This suggests that above 180°C, the emission of CO_2 reaches a constant state and the pressure decreases to overcome the constant value.

The evolution of the gas emitted as a function of the DEA liquid used was monitored as shown in Figure 8. The graph of the gas emitted as a function of the volume flow rate of DEA liquid introduced shows that absorption of the gas emitted in the furnace by the ejected DEA liquid is possible up to a value of 250l/h of DEA. Between 250l/h and 260l/h, the small amount of gas emissions in the furnace shows that the absorption of furnace gases by the DEA liquid can only take place at a maximum value of 250l/h of DEA liquid in the furnaces.

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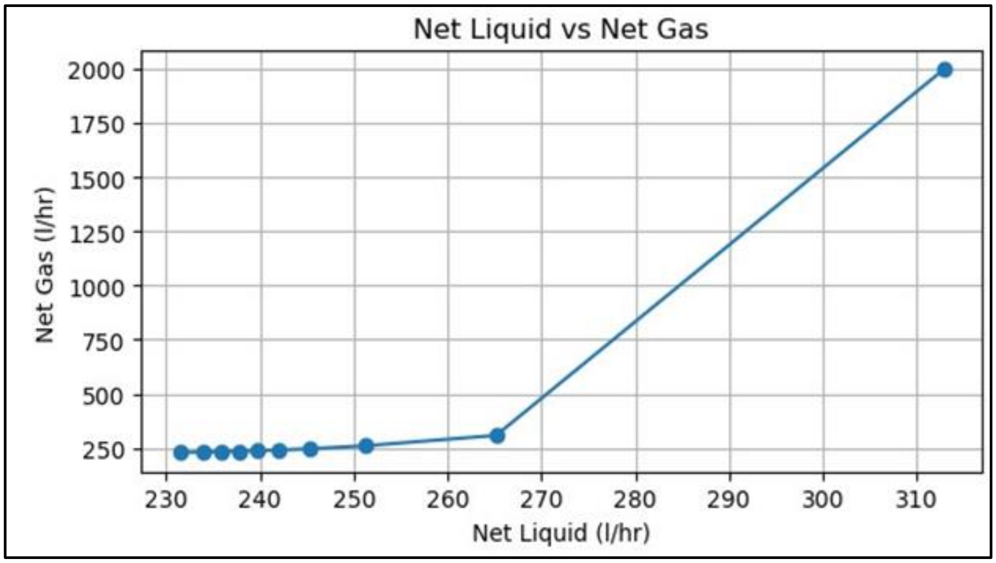

Figure 8.
Net gas profile as a function of the Net liquid DEA.

Above 270l/h of ejected DEA, the DEA becomes saturated and can no longer absorb the gas emitted in the furnace, in which case the volume of gas released increases rapidly. Similar research on CO_2 capture to reduce emissions from solvent amines was conducted by other researchers. Liu et al. studied the rate of CO_2 absorption in a biphasic solvent composed of aminoethyl ethanolamine and diethylethanolamine and demonstrated that amine scrubbing is currently the most promising technology for capturing CO_2 from gas turbines and coal flue gases[52]. The biphasic solvent, consisting of 25% aminoethylethanolamine (AEEA) and 50% diethylethanolamine (DEEA), could be a potential solution as it significantly reduces regeneration energy. The comprehensive solubility of N_2O and mass transfer studies on an effective reactive N, N-dimethylethanolamine (DMEA) solvent for post-combustion CO_2 capture involved investigating the physical solubility and mass transfer absorption performance of CO_2 in an aqueous DMEA solution under different operating conditions [53]. The results included the proposal and improvement of a predictive model of correlations, showing good agreement with experimental values with an error of 1.48%. Zhu et al. studied a compact and easy-to-use mass spectrometer for online monitoring of amines in the flue gas of a post-combustion carbon capture plant to demonstrate the instrument's analytical performance for various solvent amines and degradation amines [54]. The instrument was installed at the top of the absorption tower to provide real-time data on amine emissions to the plant's information management system. Other studies focused on the effect of temperature and gas flow rate on CO_2 capture by monoethanolamine (MEA) as a solvent. The absorption capacity was determined at different gas flow rates and temperatures, showing that absorption capacity increases as temperature decreases and flow rate increases [55, 56]. The ongoing pursuit of efficient CO_2 separation technologies is critical for addressing the climate crisis, and the current study on CO_2 capture using diethanolamine (DEA) aligns closely with recent advancements in carbon capture technology, particularly those involving multifaceted separation processes. The novel multi-sorbent process for CO_2/N_2 separation based on vacuum-swing adsorption (VSA), as described by Ward and Pini [57] provides insights into how improvements in separation efficiency can be achieved through innovative process configurations. The multisorbent configurations of layered-bed and mixed-bed processes offer noteworthy advancements over traditional single-adsorbent methods. These configurations enhance CO_2 recovery and purity, achieving a 5% increase in CO_2 recovery and significantly reducing energy usage by approximately 35% when adhering to stringent performance targets for post-combustion carbon capture [58]. This is particularly relevant to our study, which also emphasizes optimizing the absorption capacities of materials like DEA in rotary kilns to maximize CO_2 capture effectiveness. The research evidence indicates that at temperatures below 140°C, gas emissions remain low and stable due to the DEA liquid's capabilities to absorb gases [59]. Continuing with the development of TEPA-impregnated activated carbon, as discussed in previous articles, this new material showed significant CO_2 adsorption capacity and stability with up to 10 regeneration cycles, achieving optimal performance at 5% TEPA loading [60]. This result illustrates the complementarity between adsorption and absorption systems: while DEA provides a liquid solution with good absorption properties, AC-TEPA as a solid adsorbent can act synergistically, increasing the overall efficiency of CO_2 capture processes. This approach aligns with Zhu, et al. [54] who demonstrated the importance of real-time monitoring of amine emissions that allows for better management of capture and conversion processes while minimizing reagent losses. These studies corroborate the present work on CO_2 capture by amine solvents, particularly the diethyl ethanolamine used in this simulation [61]. This work will help the WACEM cement plant to mitigate CO_2 pollution, which poses environmental and health hazards.

4. Conclusion

This research demonstrates the feasibility of CO_2 capture from clinker combustion kilns at the WACEM Cement Industry using Aspen Hysys 2.2 and an aqueous diethylamine (DEA) solution. The simulation results show optimal CO_2 absorption when the flue gas mass flow rate does not exceed 12.5% of the liquid mixture at the column top, ensuring efficient gas-liquid interaction and minimizing CO_2 emissions. Additionally, using Hysplit software for emissions analysis provides valuable insights into particulate pollution in the surrounding environment. The study provides a feasible and adaptable solution for minimizing greenhouse gas emissions in cement production, contributing to more sustainable and environmentally responsible industrial practices by optimizing process conditions. Implementing this technology at an industrial level can significantly enhance environmental sustainability, supporting efforts to mitigate climate change. Future work should focus on experimental validation, economic analysis, and alternative absorbents to improve efficiency and cost-effectiveness.

Transparency:

The authors confirm that the manuscript is an honest, accurate, and transparent account of the study; that no vital features of the study have been omitted; and that any discrepancies from the study as planned have been explained. This study followed all ethical practices during writing.

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