

Original Research

**TEA of a Unique Two-Pathways Process for Post-Combustion CO<sub>2</sub> Capture**Rui Wang <sup>1</sup>, Husain E. Ashkanani <sup>2</sup>, Bingyun Li <sup>3</sup>, Badie I. Morsi <sup>1, \*</sup>

1. Department of Chemical and Petroleum Engineering, University of Pittsburgh, Pittsburgh, PA 15261, USA; E-Mails: [ruw28@pitt.edu](mailto:ruw28@pitt.edu); [morsi@pitt.edu](mailto:morsi@pitt.edu)
2. Department of Chemical Engineering, College of Engineering and Petroleum, Kuwait University, P.O. Box 5969, Safat 13060, Kuwait; E-Mail: [husain.ashkanani@ku.edu.kw](mailto:husain.ashkanani@ku.edu.kw)
3. Department of Orthopaedics, School of Medicine, West Virginia University, Morgantown, WV 26506, USA; E-Mail: [bili@hsc.wvu.edu](mailto:bili@hsc.wvu.edu)

\* **Correspondence:** Badie I. Morsi; E-Mail: [morsi@pitt.edu](mailto:morsi@pitt.edu)**Academic Editors:** Nagasree Garapati and Megan Smith**Special Issue:** [Carbon Dioxide Utilization: Strategies for an Evolving Climate](#)

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**Abstract**

A unique two-Pathways process using aqueous sodium glycinate for CO<sub>2</sub> capture from a split flue gas stream emitted from 600 MWe post-combustion coal power plant was developed in Aspen Plus v.10. The split gas flow rate used was 44.75 ton/h and contained 0.0023 mol% SO<sub>2</sub> and 13.33 mol% CO<sub>2</sub>. The process includes a washing unit, a CO<sub>2</sub> absorption unit, a reverse osmosis unit, and a solvent regeneration unit or an ultrafiltration unit. The washing unit uses deionized water to completely remove SO<sub>2</sub> and the CO<sub>2</sub> absorption unit uses SGS to capture at least 90 mol% of the CO<sub>2</sub> in the split flue gas stream. Upon CO<sub>2</sub> and SGS reactions, the resulting liquid products exhibit phase-separation into CO<sub>2</sub>-lean phase and CO<sub>2</sub>-rich phase, allow two distinct pathways. Pathway (i) is to regenerate mostly the CO<sub>2</sub>-rich phase, collect the released CO<sub>2</sub>, and compress it for sequestration purposes. Pathway (ii) is to send the liquid stream from the CO<sub>2</sub> absorption unit to the ultrafiltration unit to separate the solid nanomaterials. The hydraulics and mass transfer characteristics in the washing and CO<sub>2</sub> absorption units were obtained; and techno-economic analysis (TEA) for Pathways (i) and (ii),



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including Capital Expenditure (CAPEX), Operating Expenditure (OPEX), and Levelized Cost of CO<sub>2</sub> Captured (LCOC), were calculated and compared. The simulation results revealed that the CAPEX, OPEX, and LCOC for Pathway (i) were (\$12,039,251), (261 \$/h), and (54.01 \$/ton-CO<sub>2</sub> captured), respectively, and those for Pathway (ii) were (\$5,908,000), (237.2 \$/h), and (39.90 \$/ton-CO<sub>2</sub> captured), respectively. Moreover, in Pathway (ii), 8.19 ton/h of CO<sub>2</sub> were captured to produce 15.62 ton/h NaHCO<sub>3</sub> nanomaterials, which were sold to offset the overall process cost. The LCOC values indicate that Pathway (ii) is more cost-effective than Pathway (i) because LCOC values for Pathway (ii) are much lower than those for Pathway (i).

**Keywords**

CO<sub>2</sub> capture; sodium glycinate; packed-bed absorber; hydraulics; techno-economic analysis

**1. Introduction**

The atmospheric emissions of naturally occurring greenhouse gases (GHGs), including carbon dioxide (CO<sub>2</sub>), methane (CH<sub>4</sub>), nitrous oxide (N<sub>2</sub>O), and water vapor (H<sub>2</sub>O), and synthetically-made GHGs, such as chlorofluorocarbons (CFCs), are increasing the global average temperature, which has already reached 1°C above the pre-industrial level [1]. The increase of the global temperature due to atmospheric GHGs emissions has disastrous impacts on humans and the environment [2]. For instance, it will lead to: (1) melting of glaciers and ice in polar regions and rising of sea level, which could flood several worldwide coastal areas, leading to forced migration, hunger, and loss of human lives [3]; (2) warming and changing of the ocean chemistry, which could disrupt the ocean ecosystem and food web, resulting in dramatic consequences on marine species, and lifestyle of humans depending on them [4, 5]; and (3) increasing the intensity and frequency of cyclones, hurricanes, precipitation, and storm surge, leading to loss of human lives and infrastructure, dangerous floods, severe droughts and intolerable heatwaves [6].

The recent rise of the global temperature is due in part to the unmanageable global CO<sub>2</sub> emissions, which have increased from 22.8 Gt (Gigaton = 10<sup>9</sup> ton) in 1998 to 34.0 Gt in 2019 [7]. Table 1 shows CO<sub>2</sub> emissions from fuel combustion in 2019 [8, 9], and as can be seen the largest share of CO<sub>2</sub> emission comes from electricity and heat production plants at (41.2%), transportation (including aviation, navigation, cars, rails and pipelines transport) at (24.1%), small industries at (18.5%), buildings at (8.2%), and others at (8.0%), totaling 34.0 Gt.

**Table 1** CO<sub>2</sub> emissions from fuel combustion in 2019 [8, 9].

CO <sub>2</sub> emission source	Gigaton (Gt)	Share of total
Electricity and heat production plants	14.0	41.2%
Transportation, including aviation, navigation, cars, rail and pipeline transport	8.2	24.1%
Industry	6.3	18.5%
Buildings	2.8	8.2%
Others	2.7	8.0%

Total (Gt)	34.0	100%
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On the effort to mitigate global average temperature rise, the 2015-Paris Agreement on climate change aimed at holding the temperature rise to well below 2°C and to pursue endeavors to limit this rise to 1.5°C [10, 11]. In 2018, the Intergovernmental Panel on Climate Change (IPCC) projected that by 2030, the CO<sub>2</sub> emission has to decrease by 25% or 45% to limit the average global temperature to below 2°C or below 1.5°C, respectively [12]. To curb the CO<sub>2</sub> emissions into atmosphere, worldwide preventative and mitigative strategies have been implemented. Preventive strategies include programs, which are directly related to promoting the use of renewable energies and improving energy efficiency, while mitigative strategies refer to CO<sub>2</sub> emission controls from power plants and industrial facilities, which discharge huge amounts of CO<sub>2</sub> into the atmosphere. The European Union (EU) Strategic Energy Technology (SET) predicts that Carbon Capture and Sequestration (CCS) will contribute up to 30% of the total CO<sub>2</sub> emission reduction in EU by 2050 [13]. Ketzer, Iglesias [14] also estimated that the CCS may contribute to 20% of global CO<sub>2</sub> emission reduction by 2050 and to 55% by 2100.

Due to their different chemical properties, GHGs are removed from the atmosphere by various processes, e.g., CO<sub>2</sub> is absorbed by sinks, such as live plants, soil, and the ocean, however, CFCs are only destroyed by sunlight in the upper atmosphere. In 2020, the US-IEA reported that coal and natural gas were the major primary energy sources for the total global electricity generation at 38% and 23%, respectively [15]. To reduce the CO<sub>2</sub> emission from coal power plant, several CO<sub>2</sub> capture strategies are available, including pre-combustion, oxy-combustion, and post-combustion capture [16]. Despite the advantages of pre-combustion and oxy-fuel combustion captures, they are unlikely to replace post-combustion capture on a global scale [17] because the latter is capable of retrofitting with current gas turbine system [18]. Post-combustion capture technologies include using adsorbents, membranes, and chemical absorbents (solvents).

This paper is focusing on post-combustion CO<sub>2</sub> capture processes using only chemical solvents, while using membranes and solid adsorbents as post-combustion methods are beyond the scope of this study. Numerous chemical solvents have been used in post-combustion CO<sub>2</sub> capture processes, including aqueous ammonium hydroxide, potassium carbonate solutions, alkanolamines, and amino acids [19]. The Aqua-Ammonia process uses aqueous NH<sub>4</sub>OH for CO<sub>2</sub> capture from flue gas. Darde, Thomsen [20] simulated CO<sub>2</sub> capture from flue gas using chilled ammonia at temperature between 275 and 283 K. Liu, Wang [21] measured the overall mass transfer coefficients of CO<sub>2</sub> in 5% and 10% aqueous ammonia at 293 K and 313 K in a small wetted-wall column. Jiang, Yu [22] also simulated the CO<sub>2</sub> absorption by aqueous ammonia in Aspen Plus and optimized the process using inter-cooling system in the absorber to enhance CO<sub>2</sub> absorption and improve the overall process.

The Benfield process employs hot potassium carbonate (K<sub>2</sub>CO<sub>3</sub>) solutions to capture CO<sub>2</sub> and H<sub>2</sub>S from flue gas and natural gas [23]. This process was claimed to have several advantages, such as low regeneration energy, low volatility, multi-impurities capture (CO<sub>2</sub>, SO<sub>x</sub>, and NO<sub>x</sub>), beside producing fertilizer [24, 25]. Smith, Xiao [26] designed a laboratory-scale pilot plant to capture 4-10 kg/h CO<sub>2</sub> from an air/CO<sub>2</sub> feed gas rate of 30-55 kg/h and developed an Aspen Plus model, which was validated against the pilot plant data. In addition, The Cooperative Research Center for Greenhouse Gas Technologies (CO<sub>2</sub>CRC) developed the UNO MK3 process, which used K<sub>2</sub>CO<sub>3</sub> for capturing 90 % of the CO<sub>2</sub> emission in a large-scale power plant [26].

Aqueous monoethanolamine (MEA) is frequently used for CO<sub>2</sub> capture from flue gas and natural gas [19, 27-29]. The CO<sub>2</sub>-MEA reaction is fast, however, the production of stable carbamates [30], leads to an average CO<sub>2</sub> loading capacity and a high solvent regeneration energy requirement [31, 32]. Godini and Mowla [33] constructed a pilot-scale plant using MEA, obtained different experimental data in a wide range of operating conditions, and developed a mathematical model, which was validated against the pilot plant experimental data. Diethanolamine (DEA) and triethanolamine have relatively low heat of reaction with CO<sub>2</sub> and accordingly they require low regeneration duty [34, 35], nonetheless, their CO<sub>2</sub> absorption rates are too slow to use without the addition of a promoter [32]. Methyl-diethanolamine (MDEA) was used for CO<sub>2</sub> capture from flue gas [36]. Sterically hindered amine (e.g., 2-amino-2-methyl-1-propanol aka AMP) was reported to have a good CO<sub>2</sub> loading and a reasonable absorption rate [32, 37-49]. Law, Azudin [50] also developed a model in Aspen Plus v8.8 to optimize the CO<sub>2</sub> capture process using MEA and MDEA blends.

Sodium glycinate salt (SGS), as an amino acid (AA), was used as an absorbent for CO<sub>2</sub> capture due to its high CO<sub>2</sub> loading, non-volatile nature, and resistance to degradation in gas streams containing oxygen [51-54]. Lee, Choi [55] measured the density, viscosity, surface tension of SGS solutions at different concentrations at temperatures between 303 and 353 K. Park, Son [56] investigated the reaction kinetics between CO<sub>2</sub> and SGS at different temperatures between 298 K and 318 K using a stirred semi-batch vessel. Lee, Song [57] simulated the CO<sub>2</sub> absorption by SGS with different concentrations and temperatures using Pro-II and the physical properties of SGS solution predicted by Pro-II model were compared with actual experimental data. Also, Vaidya, Konduru [58] studied the CO<sub>2</sub> reaction kinetics with two aqueous amino acids, potassium glycinate and taurine using a stirred-cell reactor. More recently, Li, Wang [59] experimentally demonstrated the formation of bicarbonate nanoflowers and nanofibers when using amino acid (AA)-XOH solutions to capture CO<sub>2</sub>, wherein X stands for sodium (Na) or potassium (K) and AA are Glycine or Alanine.

The preceding introduction reveals the following: (1) numerous solvents have been used in post-combustion CO<sub>2</sub> capture processes at lab-scale and pilot-scale, however, the TEA of these processes are not available in the open literature; (2) it is impossible to compare the performance of the solvents used under similar operating conditions, including the same flue gas composition, which makes it difficult to select an effective solvent for CO<sub>2</sub> capture; and (3) even though AAs have been used for CO<sub>2</sub> capture, they were not used for producing high-value products, which could be sold to offset the cost of the CO<sub>2</sub> capture process.

The main objective of this study is to develop in Aspen Plus v.10 and perform techno-economic analysis (TEA) of a unique two-Pathways, post-combustion CO<sub>2</sub> capture process using SGS to remove CO<sub>2</sub> from a split stream from the flue gas emitted from the 600 MWe clean coal power plant, intended for use in the Wolverine Clean Energy Venture (WCEV) project. This process produces an aqueous CO<sub>2</sub>-rich stream, including solid sodium bicarbonate (NaHCO<sub>3</sub>) nanomaterials, which can be proceeded in two different Pathways (i) and (ii) as illustrated in Figure 1. In Pathway (i), the CO<sub>2</sub>-rich stream containing the solid nanomaterials is regenerated in a stripper and the released CO<sub>2</sub> is compressed in preparations for subsequent sequestration in geologic formation or for use in enhance oil recovery (EOR), considering the federal 45Q tax credits for CO<sub>2</sub> reduction are available at \$50/ton for sequestration and \$35/ton for EOR. In Pathway (ii), the NaHCO<sub>3</sub> solid nanomaterials, which have numerous uses [60-62], are separated from the CO<sub>2</sub>-rich stream and could be sold to offset the overall costs of the CO<sub>2</sub> capture process. In addition, this pathway could be a novel venue for producing NaHCO<sub>3</sub>, which is conventionally manufactured by the Solvay process or by mining

natron, a naturally occurring mineral consisting of sodium bicarbonate, sodium carbonate decahydrate ( $\text{Na}_2\text{CO}_3 \cdot 10\text{H}_2\text{O}$ ) and small amounts of sodium chloride and sodium sulfate [63].

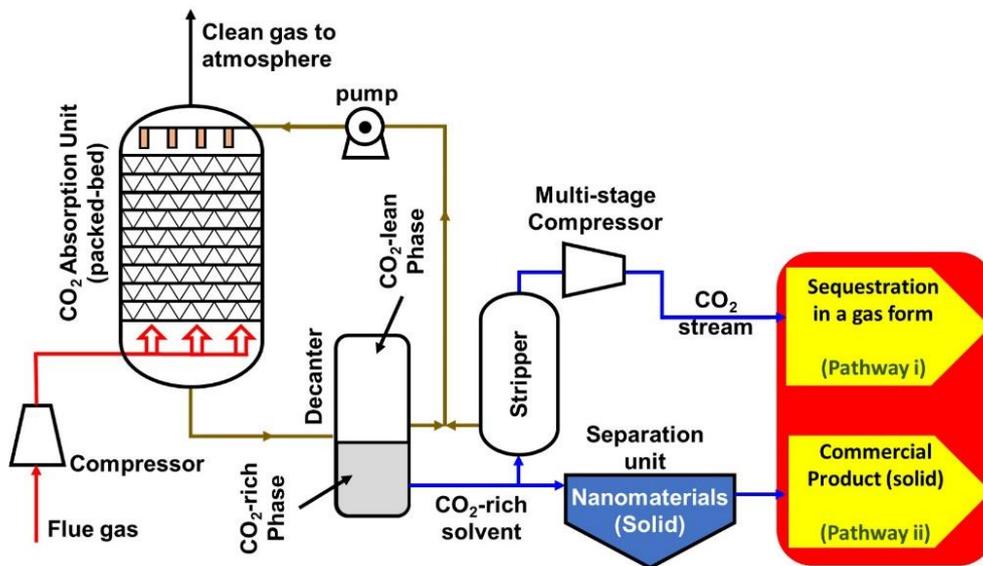


Figure 1 Process two Pathways.

It is important to mention that our two-Pathways process offers a great flexibility and a risk management strategy, however, market forces, including sequestration costs, product demand, and cost of electricity, would factor into the decision for selecting the desired pathway. Nonetheless, due to the huge daily atmospheric CO<sub>2</sub> emissions, Pathway (i) could be the main pathway to follow, while Pathway (ii) would be switched on when electricity is in high demand since CO<sub>2</sub> desorption in pathway (i) requires electricity.

## 2. Aspen Plus Simulation

Our chemical process simulation in Aspen Plus v.10 required, among other, precise knowledge of (1) The physico-chemical properties of the solute and solvent species, (2) The solute-solvent reaction kinetics, including reaction rate constants, and reaction orders with respect to the solute and the solvent; and (3) The design criteria of the unit operation to be used to carry out the chemical reactions, including flooding, gas-liquid mass transfer, two-phase pressure drop, liquid-phase holdup, and packing specific wetted area. It should be noted that the most common unit operation used in CO<sub>2</sub> capture is a counter-current packed-bed absorber. These will be discussed in the following.

### 2.1 Properties of the Flue Gas-Liquid Solvent Used

The composition of the flue gas from the Wolverine 600 MWe power plant used is given in Table 2; and the flow rate of the split flue gas stream used in this study was 12.43 kg/s (44.75 ton/h) at 353.15 K and 1.013 bar.

**Table 2** Flue gas composition from the Wolverine 600 MW power plant [64].

Components	mol %	wt %
CO <sub>2</sub>	13.33	20.18
H <sub>2</sub> O	12.31	7.63
N <sub>2</sub>	70.36	67.79
O <sub>2</sub>	4.00	4.400
SO <sub>2</sub>	2.35 × 10 <sup>-3</sup>	5.20 × 10 <sup>-3</sup>

Table 3 includes some properties of glycine and H<sub>2</sub>O, which are available in Aspen Plus v.10 database, and those of SGS, which were reported by Lee, Song [57]. The properties of H<sub>3</sub>O<sup>+</sup>, Na<sup>+</sup>, OH<sup>-</sup> and HCO<sub>3</sub><sup>-</sup> ions, which are present as result of the chemical reaction between CO<sub>2</sub> and SGS are also available in Aspen Plus database.

**Table 3** Properties of H<sub>2</sub>O, glycine and SGS [57].

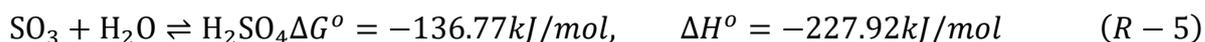
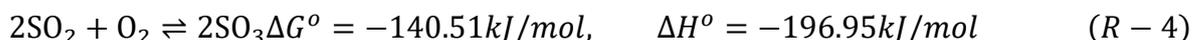
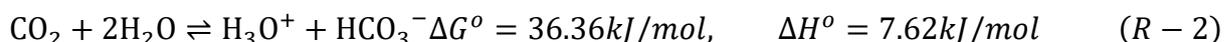
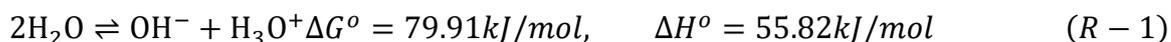
Properties	H <sub>2</sub> O	Glycine	SGS
Molecular weight, kg/kmol	18.01	75.07	97.05
Normal boiling point, K	373.20	699	541.54
Critical temperature, K	647.30	1,021	736.60
Critical pressure, MPa	22.05	6.74	4.27
Critical volume, m <sup>3</sup> /kmol	0.06	0.20	0.35
Acentric factor, -	0.34	0.69	0.95
Heat of formation, kJ/mol	-241.84	-390.37	-372.54
Gibbs free energy of formation, kJ/mol	-228.61	-299.90	-188.23

The constraints imposed on our CO<sub>2</sub> capture process were: (1) 100 mol% SO<sub>2</sub> removal from the split flue gas stream in the WU before CO<sub>2</sub> absorption, (2) at least 90 mol% CO<sub>2</sub> capture from the desulfurized flue gas in the CAU, (3) no flooding in the process to ensure smooth operation, (4) the water content in the CO<sub>2</sub> stream destined for sequestration must be less than 600 ppm to avoid the formation of CO<sub>2</sub> ice-like hydrates in the transportation lines, and (5) the packing height to diameter (H/D) ratio for the packed-beds used should be equal or greater than 6 to avoid channeling in both units as previously reported in the literature [65, 66]. The existence of preferential flow pathways, aka, channeling instead of homogeneous flow is often observed when (H/D) ratio is <5 [67]. The presence of channeling substantially reduces the gas-liquid interfacial area and mass transfer, which lower the absorption efficiency of the CAU [68]. To completely remove SO<sub>2</sub>, the raw flue gas is first washed in a WU using Deionized Water (DIW) in a counter-current packed-bed absorber wherein the reaction between SO<sub>2</sub> and H<sub>2</sub>O takes place.

## 2.2 SO<sub>2</sub> Reaction with H<sub>2</sub>O

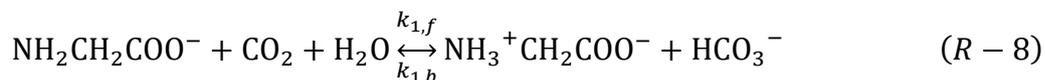
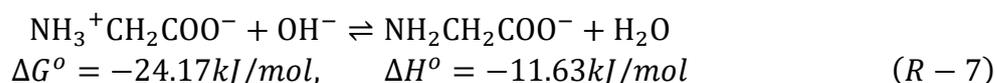
The Henry's Law constants for CO<sub>2</sub>, SO<sub>2</sub>, N<sub>2</sub> and O<sub>2</sub> absorption into DIW are available in the literature [69]. Also, the ELEC-NRTL model in Aspen Plus includes the following reactions: water dissociation (R-1), CO<sub>2</sub> and water reaction to form bicarbonate ions (R-2), bicarbonate dissociation in water to form carbonate ions (R-3), SO<sub>2</sub> reaction with oxygen to form SO<sub>3</sub> (R-4), and SO<sub>3</sub> reaction

with water to form H<sub>2</sub>SO<sub>4</sub> (R-5). All these are equilibrium reactions, and therefore their corresponding equilibrium rate constants can be calculated using the change of the standard Gibbs free energy ( $\Delta G^\circ$ ) [70].



### 2.3 CO<sub>2</sub> Reactions with Aqueous SGS

As the simplest amino acid, glycine (NH<sub>2</sub>CH<sub>2</sub>COOH) undergoes zwitterionic transformation to protonate the amino group according to reaction (R-6) to become zwitterion glycine (NH<sub>3</sub><sup>+</sup>CH<sub>2</sub>COO<sup>-</sup>), which does not react with CO<sub>2</sub> [71]. In the presence of NaOH, however, the amino group is deprotonated in this basic environment to form SGS (NH<sub>2</sub>CH<sub>2</sub>COONa), which is readily reactive with CO<sub>2</sub> as expressed in reaction (R-7). The CO<sub>2</sub> will then react with the SGS in water to form carbamate, which rapidly undergoes hydrolysis to produce glycine and bicarbonate ions according to reaction (R-8) [56]. The CO<sub>2</sub> will also react with H<sub>2</sub>O to produce bicarbonate and carbonate ions as expressed above is reactions (R-2) and (R-3), respectively. It should be noted that (R-6) and (R-7) are equilibrium reactions and their corresponding equilibrium constants can be obtained using the change of the standard Gibbs free energy ( $\Delta G^\circ$ ), however, (R-8) is a kinetic reaction whose forward and backward rate constants are required.



The forward rate constant ( $k_{1,f}$ ) for reaction (R-8) can be calculated by Equation (1) proposed by Lee, Song [72]. Also, based on the standard Gibbs free energy change ( $\Delta G^\circ$ ) and the equilibrium rate constant by Ziemer, Niederhauser [73], we propose Equation (2) to calculate the backward rate constant ( $k_{1,b}$ ) for reaction (R-8). Both  $k_{1,f}$  and  $k_{1,b}$  are in m<sup>3</sup>/kmol·s.

$$k_{1,f} = 1.95 \times 10^{13} \exp\left(\frac{-7,670}{T}\right) \quad (1)$$

$$k_{1,b} = 3.82 \times 10^{12} \exp\left(\frac{-9,508}{T}\right) \quad (2)$$

## **2.4 Washing Unit (WU) for Total SO<sub>2</sub> Removal from the Raw Flue Gas Stream**

Since the raw flue gas stream from the power plant contains SO<sub>2</sub> as given in Table 2, a WU was designed to capture all SO<sub>2</sub> from the raw flue gas stream using DIW to produce a desulfurized (sulfur-free) flue gas stream ready for CO<sub>2</sub> capture. The WU is a counter-current fixed-bed absorber packed with Mellapak 250Y. The flow rate of the split raw flue gas (12.43 kg/s) used in our process enters the bottom of the WU at 353.15 K and 1.10 bar and the DIW stream (23.75 kg/s) enters from the top of the WU at 313.15 K and 1.03 bar. A compressor is used to deliver the raw flue gas to the WU at certain superficial gas velocity to overcome the expected pressure drop associated with the resulting counter-current two-phase flow in the unit (fixed-bed reactor).

Following the reaction between SO<sub>2</sub> and DIW as discussed above, the liquid-phase stream coming from the bottom of the WU is sent to a Reverse Osmosis Unit (ROU) for removing the dissolved sulfate ions (SO<sub>4</sub><sup>2-</sup>) from the “used” DIW. The ROU consists of 60 BW30-400 reverse osmosis membranes [74] arranged in three sets of 30, 18, and 12 membranes, respectively. The permeate (cleaned DIW) from the ROU is then recycled back to the top of the WU for another cycle of SO<sub>2</sub> removal, while the reject from the ROU containing H<sub>2</sub>SO<sub>4</sub>, can be concentrated and used as an acid, or neutralized using KOH to produce K<sub>2</sub>SO<sub>4</sub>, which can be used as a fertilizer [75]. On the other hand, the desulfurized flue gas stream coming from the top of the WU is sent directly to the CO<sub>2</sub> CAU, which is also a fixed-bed packed with (Mellapak 250Y) to react with the aqueous SGS stream coming from the top of the CAU. In this study using Aspen Plus v.10, the WU was modelled as a rate-based model, and the ROU with its specifications [76] was represented by one separator. It should be emphasized that there will be no need for this WU, if the raw flue gas is completely sulfur-free, however, in this study, the WU is an integral part of the CO<sub>2</sub> capture process whether in Pathway (i) or Pathway (ii).

## **2.5 Process Flow Diagram (PFD) for Pathway (i)**

Figure 2 shows the PFD for Pathway (i), which also includes the WU. This Pathway is designated to capture at least 90 mol% of CO<sub>2</sub> in the split flue gas stream and preparation it for subsequent sequestration. In this figure, the solution stream coming from the bottom of the CAU is sent to a decanter in which, it exhibits a phase-separation into two phases (CO<sub>2</sub>-rich and CO<sub>2</sub>-lean). In this Pathway, only the CO<sub>2</sub>-rich phase is sent to the stripper for regeneration. It should be noted that the pure CO<sub>2</sub> stream leaving the separator in Figure 2 is further compressed to 152.7 bar using a 2-stage compressor with intercooling. As a result, the water content in this CO<sub>2</sub> stream was only 470 ppm, which is much lower than the limit of 600 ppm maximum set by one of the constraints imposed the process. Thus, this CO<sub>2</sub> stream is ready for subsequent sequestration. It should be noted that like the WU, the CAU was modelled in Aspen Plus v.10 using the rate-based model.

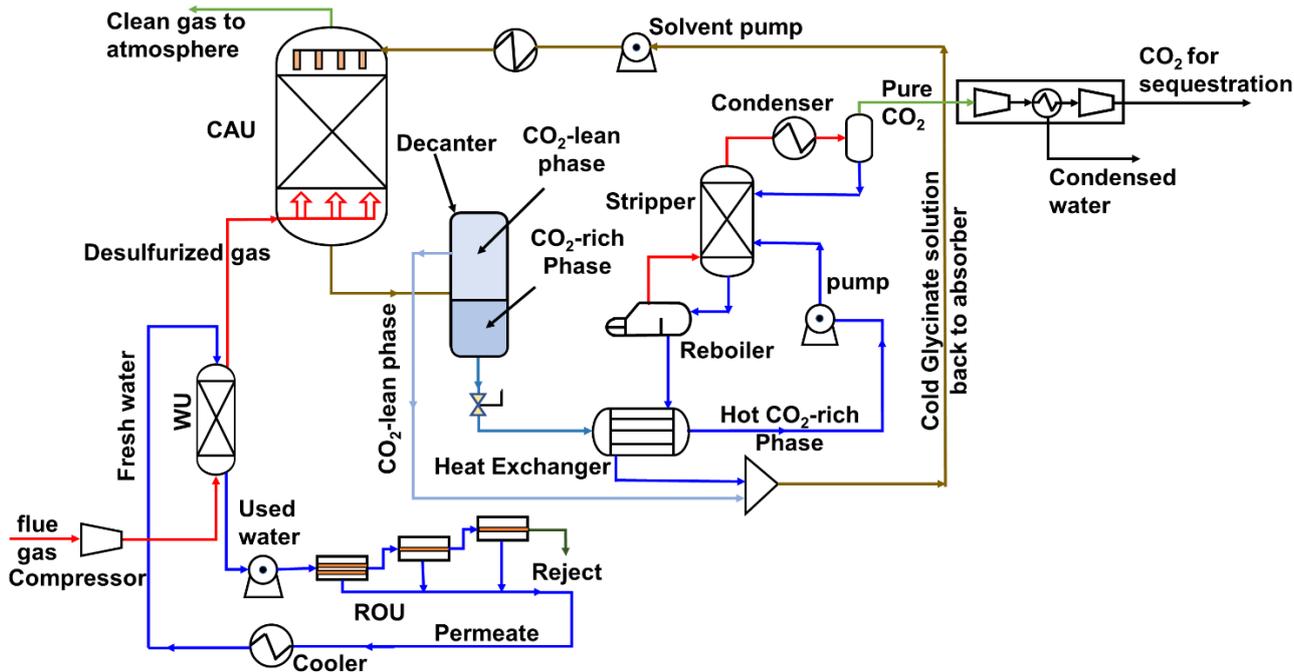


Figure 2 Process flow diagram for pathway (i).

### 2.6 Process Flow Diagram (PFD) for Pathway (ii)

Figure 3 shows the PFD for Pathway (ii), including the WU, which was discussed above. This Pathway is allotted to CO<sub>2</sub> capture for the production of NaHCO<sub>3</sub> nanoparticles. As can be seen in this figure, following the reactions between CO<sub>2</sub> and SGS in the CAU, the resulting solution stream coming from the bottom of the CAU is directly pumped into an Ultra Filtration Unit (UFU), which uses 49 SFP-2880 ultrafiltration membranes by DOW [77]. These membranes are arranged in series and used to separate the nanoparticles from the liquid-phase. In Aspen Plus simulator, like the ROU, the UFU with its specifications [77] was represented by one separator.

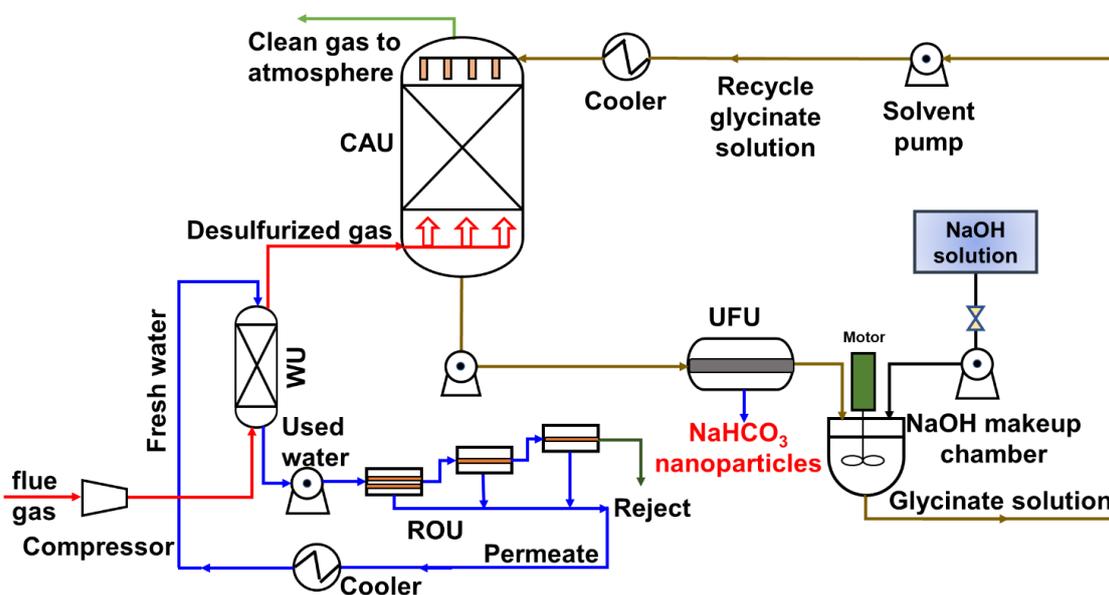


Figure 3 Process flow diagram for pathway (ii).

The filtrate from the UFU containing no nanomaterials is then mixed with NaOH in the makeup chamber, which is a continuously stirred tank reactor (CSTR) used to convert the glycine to SGS. The reaction between glycine and NaOH in the CSTR increased the temperature of the SGS produced to a maximum of 338.7 K. This SGS solution is then cooled to 298.15 K and recycled back to the CAU for another cycle of CO<sub>2</sub> capture. It should be underlined that maintaining the temperatures of the inlet DIW to the WU and the inlet SGS to the CO<sub>2</sub> absorber at 298.15 K, lowered the temperature throughout the process including the temperature of the SGS solution produced in the CSTR. This is important for our process since the thermal degradation temperature of SGS reported in the literature was 398 K [78, 79]. It is important to note that sodium glycinate solutions are stable at 220°C [78]. Also, the volatilities of the sodium glycinate solutions (SGS) were reported to be very low [80], resulting in a negligible solvent loss in our process.

### **3. Design Criteria of Countercurrent Packed-Beds Used in Aspen Plus v.10**

As stated above a counter-current packed-bed contactor is suitable for dealing with high flue gas and solvent flow rates due to their high voidage. A packed-bed absorber frequently used in CO<sub>2</sub> capture processes is a cylinder filled with packings of various types, shapes, and sizes whose main function is to increase the turbulence and to spread the liquid over their surfaces thereby increasing the contact area between the gas and liquid phases, which ultimately increases the gas-liquid mass transfer. The packing types are in general random or structured [81]. For using Aspen Plus in our process simulation in a counter-current packed-bed, a precise knowledge of the hydraulics (two-phase pressure drop, liquid holdup, and bed flooding) and mass transfer characteristics (Gas-side ( $k_G$ ) and liquid-side ( $k_L$ ) mass transfer coefficients, and specific packing wetted surface area ( $a_w$ )) for the gas-liquid systems used is required [82]. Extensive research was conducted to obtain the required available literature correlations for the hydraulics and mass transfer of the gas-liquid systems used in this study. These correlations are discussed in the following.

#### **3.1 Hydraulics Literature Correlations in Counter-Current Packed-Beds**

Literature correlations on two-phase pressure drop in packed-beds are mostly empirical and were primarily developed for an air-water system. Sticlmair's [83] and Billet and Schultes' correlations [84] can directly predict the irrigated two-phase pressure drop based on the packing properties, as well as density, viscosity and flow rates of the gas and liquid systems used. Leva's [85] and Kister's [86] correlations, however, need interpolation for pressure drop on pressure drop charts. The literature correlations for the liquid holdup ( $\beta_L$ ), representing the liquid retained in the packed-bed during the two-phase flow are usually a function of liquid velocity, liquid density and viscosity, and packing properties [83]. Also, flooding in counter-current packed-beds can be detected using Leva's correlation [85].

#### **3.2 Gas-Liquid Mass Transfer Literature Correlations in Counter-Current Packed-Beds**

Onda, Takeuchi [87], Bravo and Fair [88], and Billet and Schultes [89] developed empirical correlations to predict  $k_L$ ,  $k_G$  and  $a_w$ , for the gas-liquid systems used in this study using structured packing. These correlations covered wide ranges of gases and solvents with different physico-chemical properties, and the experiments used to develop them were carried under ambient

pressure and temperature, which make them valid for post-combustion CO<sub>2</sub> absorption at ambient conditions.

#### 4. Process Techno-Economic Analysis (TEA)

The TEA calculations include the capital expenditure (CAPEX), operating expenditure (OPEX) and the levelized cost of CO<sub>2</sub> capture (LCOC). The equations used for calculating the total CAPEX of our process equipment can be found elsewhere [90-92]. The installation factors for the equipment used is available in Towler and Sinnott [91]. Also, the Chemical Engineering Plant Cost Index (CEPI) by Lozowski [93] was used to adjust the capital cost to the 2020 USD. The plant was assumed to operate only for 300 days per year. Since LCOC is dependent on the capital and operating costs, the TEA assumptions were, the Plant lifetime (*N*), Annual discount rate (*i*), Capacity factor (*f<sub>c</sub>*), Operating and Maintenance (O&M) cost factor (*f<sub>O&M</sub>*) and the Capital recovery factor (*f<sub>CR</sub>*). The annual Operating and Maintenance (O&M) cost was assumed to be 4% of total CAPEX as reported in the literature [94, 95] and given in Equations (3) and (4). The recovery factor and the Levelized Cost of CO<sub>2</sub> capture (LCOC) were calculated using Equations (5) and (6), respectively.

$$CAPEX_{2020} = \sum_i^{items} (CAPEX)_{2020,i} \quad (3)$$

$$OPEX_{2020} = (37 \sum W) + (C_{NaOH}\dot{m}_{NaOH} - C_{NaHCO_3}\dot{m}_{NaHCO_3}) + 0.04(CAPEX_{2020})/7200 \quad (4)$$

$$f_{CR} = \frac{i(1+i)^N}{(1+i)^N - 1} \quad (5)$$

$$LCOC = \left(\frac{f_{CR}}{f_c}\right) \sum (CAPEX_{2020})/\dot{m}_{CO_2} + OPEX_{2020}/\dot{m}_{CO_2} \quad (6)$$

In this study, the assumed values of these parameters are given in Table 4.

**Table 4** Parameters used for LCOC calculation.

Parameter	Value
Cost of electricity [96]	\$37/MWh
<i>i</i>	10%/year
<i>N</i>	30 years
<i>f<sub>c</sub></i>	0.8
<i>f<sub>O&amp;M</sub></i>	4% of the Total CAPEX, \$/year
<i>f<sub>CR</sub></i>	0.10608
<i>C<sub>NaOH</sub></i>	Cost of NaOH, 450 \$/ton
<i>ṁ<sub>NaOH</sub></i>	NaOH mass flow rate, ton/h
<i>C<sub>NaHCO<sub>3</sub></sub></i>	Cost of NaHCO <sub>3</sub> , 200 \$/ton
<i>ṁ<sub>NaHCO<sub>3</sub></sub></i>	NaHCO <sub>3</sub> mass flow rate, ton/h

## 5. Results and Discussion

The discussion will follow this sequence: (1) WU and ROU; (2) CO<sub>2</sub> absorber for Pathways (i) and (ii); (3) the stripper in Pathway (i) and the UFU in Pathway (ii); and (4) Comparative TEA for the two Pathways.

### 5.1 Characteristics of the WU and ROU

The characteristics of the WU and the ROU are given in Table 5 with packing height to absorber internal diameter ratio (H/D) of 6. Table 6 gives the two inlet streams to and two exit streams from the WU. As can be noted, the inlet raw flue gas flow rate is 12.43 kg/s at (at 353.15 K and 1.104 bar), the inlet DIW flow rate is 23.75 kg/s (at 313.15 K and 1.034 bar), the outlet desulfurized flue gas flow rate is 12.10 kg/s (at 314.94 K and 1.034 bar), and the outlet DIW flow rate is 24.08 kg/s (at 326.1 K and 1.03 bar). The ROU consists of three stages, including 60 membranes (type BW30-400 by DOW [74]). The inlet used DIW flow rate to the ROU is 24.08 kg/s at (at 326.1 K and 1.03 bar), the permeate (clean DIW) flow rate is 24.75 kg/s (at 326.1 K and 14.5 bar), and the reject flow rate is 0.33 kg/s (at 326.1 K and 14.5 bar). The permeate is recycled back to the WU.

**Table 5** Characteristics of the WU and the ROU.

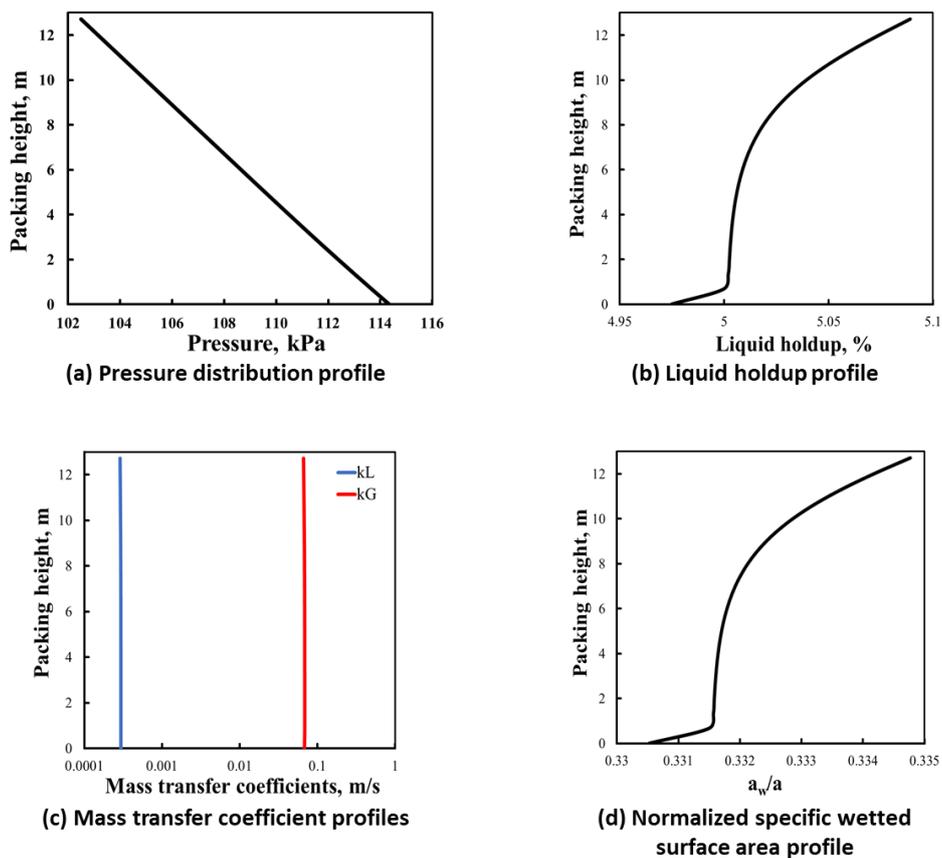
Inlet DIW flow, kg/s	23.75
Column diameter, m	2.12
Column height, m	15.23
Packing height, m	12.72
Packing type	Mellapak 250Y
ROU membrane type	BW30-400 membrane
Number of membranes	60
Inlet DIW flow, kg/s	23.75

**Table 6** Inlet and outlet streams of the WU.

	<b>Inlet flue gas</b>	<b>Desulfurized flue gas</b>	<b>Inlet DIW</b>	<b>Outlet DIW</b>
Pressure, bar	1.104	1.034	1.034	1.031
Temperature, K	353.15	314.94	313.15	326.1
Mass flow rate, kg/s	12.43	12.10	23.75	24.08

#### 5.1.1 Hydraulics and Mass Transfer in the WU

The two-phase pressure, liquid holdup, liquid-side and gas-side mass transfer coefficients, and the normalized specific packing wetted surface area are presented in Figure 4. As can be observed in this figure, the total pressure drop throughout the WU is only 0.08 bar (8 kPa), and there is a strong similarity between the liquid holdup and the normalized specific wetted surface area profiles. Also, the resistance to gas-liquid mass transfer is located in the liquid-side because the liquid-side resistance ( $1/k_L$ ) is greater than gas-side resistance ( $1/k_G$ ).



**Figure 4** Hydraulics and mass transfer in the WU.

5.1.2 Performance of the WU and ROU

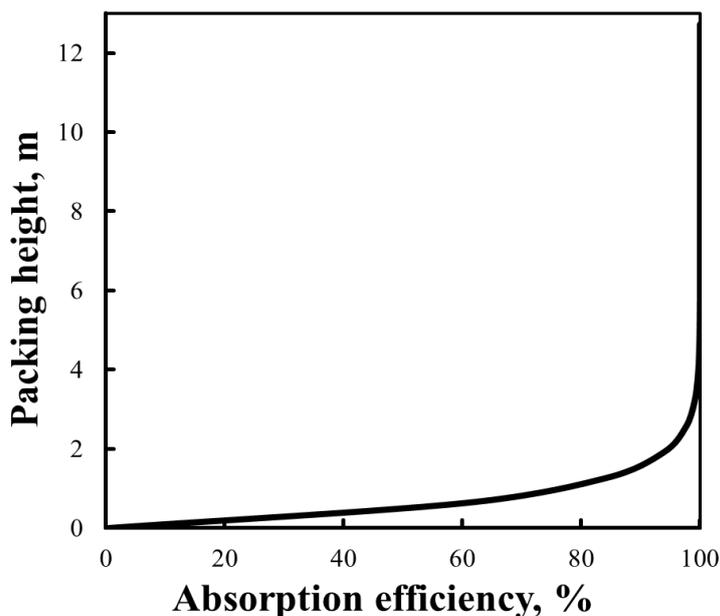
The performance of the WU in term of the composition of each component in the inlet and outlet streams of the WU is given in Table 7. As can be observed the desulfurized flue gas is completely sulfur-free, and the CO<sub>2</sub> content of this stream going to the absorber is 20.7 wt%.

**Table 7** Performance of the WU.

	Inlet flue gas	Desulfurized flue gas	Inlet DIW	Outlet DIW
Components	wt%	wt%	wt%	wt%
CO <sub>2</sub>	20.18	20.71	$5.34 \times 10^{-4}$	$1.05 \times 10^{-2}$
H <sub>2</sub> O	7.63	5.13	99.99	99.98
N <sub>2</sub>	67.79	69.64	$4.93 \times 10^{-5}$	$9.73 \times 10^{-4}$
O <sub>2</sub>	4.4	4.52	$5.87 \times 10^{-6}$	$1.16 \times 10^{-4}$
SO <sub>2</sub>	0.0052	0	0	0
H <sub>2</sub> SO <sub>4</sub>	0	0	$2.18 \times 10^{-4}$	$4.29 \times 10^{-3}$

The SO<sub>2</sub> absorption efficiency profile is shown in Figure 5, and as can be seen the WU is able to completely remove SO<sub>2</sub> from the flue gas split stream. Actually, a packing height of about 6.0 m is largely sufficient for the removal of 100 mol% of SO<sub>2</sub> from the split flue gas stream rather than 12.72 m. However, the packing height to internal diameter ratio (H/D) of a fixed-bed reactor was

constrained in this study to be greater than 6 to avoid fluid channeling. Therefore, the packing height of the WU was calculated as 12.72 m.



**Figure 5** SO<sub>2</sub> absorption efficiency profile in the WU.

Also, considering the flow rates of the used DIW and the permeate, the performance of the ROU in removing the dissolved SO<sub>2</sub> from the used DIW stream is almost 100 wt%.

### 5.1.3 Characteristics of the CAU in the Two Pathways

Table 8 shows the characteristics of the CAU used for Pathways (i) and (ii); and as can be observed more solvent flow rate and taller absorber are used in Pathway (i) than Pathway (ii) to have the same absorption efficiency. It is important to note that the flow rates of 53 L/s and 30 L/s are the minimum solvents flow rates of Pathways (i) and (ii) to meet the CAU absorption efficiency, which was constrained to at least 90 mol% CO<sub>2</sub> capture from the flue gas stream. The residence time of the desulfurized flue gas in the CAU was 137.1 s for pathway (i) and 104.1 s for pathway (ii). These residence times in the CAU lie within the reported residence time ranges (10-200 s) for post-combustion CO<sub>2</sub> absorption using ethanolamines in packed-beds [97, 98]. These residence times for SGS are longer than those in ethanolamines since the reaction kinetics of CO<sub>2</sub> and SGS are slower when compared with those of CO<sub>2</sub> and ethanolamines.

**Table 8** Characteristics of the CAU in the two pathways.

	Pathway (i)	Pathway (ii)
Pressure, bar	1.013	1.013
Temperature, K	313.15	298.15
Mass flow rate of 3M SGS solution, kg/s	58	33
Solvent flow rate, L/s	53	30
Column diameter, m	6	6
Column height, m	63.42	39.42

Packing height, m	60	36
CO <sub>2</sub> capture efficiency, mol%	91	91
Packing type	Mellapak 250Y	

#### 5.1.4 Hydraulics and Mass Transfer in the CAU

Table 9 shows that the absorber hydraulics and mass transfer characteristics in Pathways (i) and (ii), expressed in terms of pressure drop, liquid holdup, and normalized specific packing wetted surface area, are nearly the same. The hydraulics and mass transfer were sensitive to the solvent flow rate, and physico-chemical properties of the liquid and gas phases. Faster the solvent flow rate, higher the liquid hold-up, and larger the mass transfer coefficients and the normalized specific packing wetted area.

**Table 9** Hydraulics and mass transfer in the CAU.

	Pathway (i)	Pathway (ii)
Pressure drop, bar	0.006	0.004
Liquid holdup, vol%	5.17-5.50	4.27-4.83
Normalized specific packing wetted area, -	0.243-0.246	0.193-0.200
Liquid-side mass transfer coefficients ( $k_L$ ), m/s	$1.61 \times 10^{-4}$ - $2.26 \times 10^{-4}$	$8.22 \times 10^{-5}$ - $1.21 \times 10^{-4}$
Gas-side mass transfer coefficient ( $k_G$ ), m/s	$2.49 \times 10^{-2}$ - $2.83 \times 10^{-2}$	$1.25 \times 10^{-2}$ - $1.69 \times 10^{-2}$
Packing type	Mellapak 250Y	

#### 5.1.5 Performance of the CAU in Pathways (i) and (ii)

Considering the characteristics of the CAU given in Table 8, the CO<sub>2</sub> removal efficiency in Pathway (i) is 91 mol%, which is very close to that (90.7 mol%) in Pathway (ii) even though the solvent flow rate and packing height in Pathway (i) are respectively 77% and 67% greater than those in Pathway (ii). This behavior can be related to the composition of the recycled solvent in the case of Pathway (i). Different from Pathway (ii), the recycled liquid solvent in Pathway (i) is not 100% SGS, which appeared to dramatically affect the SGS chemical reaction kinetics with CO<sub>2</sub> in the flue gas stream. Actually, only 62 mol% of NaHCO<sub>3</sub> is in CO<sub>2</sub>-rich phase, which is sent to the stripper for regeneration and further recycling to the absorber.

In Pathway (i), the CO<sub>2</sub>-rich stream coming from the absorber is sent directly to decanter, in which the solution exhibits a phase separation into CO<sub>2</sub>-rich and CO<sub>2</sub>-lean phases. The CO<sub>2</sub>-rich phase represents 25 vol% of the solution and contains 62 mol% of NaHCO<sub>3</sub> nanomaterials. In this study, only the CO<sub>2</sub>-rich phase is sent for regeneration in a stripper. The characteristics of the stripper used in Pathway (i) are given in Table 10.

**Table 10** Characteristics of the stripper used in Pathway (i).

Stripper diameter, m	3
Stripper height, m	20.78
Packing height, m	18
Packing type	Mellapak 250Y

Reboiler duty, MW	4.04
Reboiler temperature, K	396.21
Condenser duty, MW	-0.82
Condenser temperature, K	293.15

In Pathway (ii), the liquid stream coming from the absorber (CAU) is sent directly to the UFU, which uses 49 SFP-2880 ultrafiltration membrane by DOW [77] to separate the NaHCO<sub>3</sub> nanomaterials. The separation efficiency of this membrane was estimated at 99.95%. Ultimately, the production of 15.62 ton/h of NaHCO<sub>3</sub> nanomaterials in Pathway (ii) requires the use of SFP-2880 ultrafiltration membrane in the UFU and an NaOH make-up rate of 7.43 ton/h. It should be noted that the CO<sub>2</sub> released upon regeneration is sent to a two-stage compressor to boost its pressure to 152.7 bar, which makes it ready for subsequent sequestration.

The mass flow rate and composition of the stream in the decanter and stripper are given in Table 11. At steady state, the total flow rate of the water and NaHCO<sub>3</sub> in the solution coming from bottom of the CAU were 150.7 ton/h, and 25.3 ton/h, respectively. Due to phase separation, the water and NaHCO<sub>3</sub> flow rates in the upper phase were 113.0 ton/h and 9.6 ton/h, while the water and NaHCO<sub>3</sub> flow rates in the lower phase were 37.7 ton/h and 15.7 ton/h, respectively. This means that the amount of water in the lower phase represents only 25 wt% of the total water in the solution. In pathway (i), only the lower phase was sent to the stripper for regeneration and since the amount of water in this stream was small, the stripper reboiler duty required to remove all CO<sub>2</sub> from this phase was only 4.04 MW. The regenerated stream from the reboiler (44.6 ton/h) consisted of 16.73 wt% NaOH, and 83.27 wt% H<sub>2</sub>O. Also, in pathway (i), the upper phase (171.6 ton/h) composed of 15.36 wt% SGS, 13.16 wt% glycine, and 5.6 wt% of NaHCO<sub>3</sub> and 65.88 wt% H<sub>2</sub>O. This upper phase was not regenerated, rather it was mixed with regenerated stream from the reboiler and recycled back to the CAU. Upon the reaction between NaOH and glycine, the recycled stream to the CAU consisted of 20.42 wt% SGS, 4.08 wt% Glycine, 4.5 wt % NaHCO<sub>3</sub>, and 71.0 wt% H<sub>2</sub>O. Thus, the recycled liquid stream to the CAU was not pure SGS solution.

**Table 11** Decanter and stripper stream results.

	CO <sub>2</sub> -rich out (CAU)	Upper phase (decanter)	Bottom-phase (decanter)	CO <sub>2</sub> -Lean out (stripper)	Recycled to (CAU)
Mass flow rate, ton/h	224.96	171.60	53.36	44.59	216.19
Mass percentage (wt%)					
SGS	11.72	15.36	-	-	20.42
H <sub>2</sub> O	67.01	65.88	70.63	83.27	71.00
Glycine	10.04	13.16	-	-	4.08
NaHCO <sub>3</sub>	11.23	5.60	29.47	-	4.50
NaOH	-	-	-	16.73	-
CO <sub>2</sub>	-	-	-	-	-

### 5.2 CAPEX and OPEX of Pathways (i) and (ii)

Table 12 shows the calculated CAPEX (\$12,039,251) and OPEX (261 \$/h) of Pathways (i) are much greater than the CAPEX (\$5,908,000) and OPEX (237.24 \$/h) for Pathway (ii). This behavior could be attributed to the cost of the stripper, initial solvent cost, heat exchangers, rotating equipment, as well as a more expensive larger CO<sub>2</sub> absorber in Pathway (i).

**Table 12** CAPEX and OPEX of Pathways (i) and (ii).

	Pathway (i)	Pathway (ii)
<b>CAPEX, USD 2020</b>		
Heat Exchangers	\$5,501,400	\$1,872,719
Packing Cost	\$324,618	\$181,182
Absorber	\$2,886,734	\$2,319,957
Initial solvent cost	\$862,826	\$202,437
Washing unit	\$253,682	\$253,682
Rotating equipment	\$1,481,531	\$349,999
ROU	\$153,250	\$153,250
UFU	--	\$127,020
Reaction Chamber	--	\$447,755
Stripper	\$575,211	--
<b>Total CAPEX, 2020 USD</b>	<b>\$12,039,251</b>	<b>\$5,908,000</b>
<b>OPEX, USD 2020/h</b>		
NaOH makeup	--	3,345.7
NaHCO <sub>3</sub> production	--	-3,169.9
O&M	66.88	32.82
Total electricity cost	194.23	28.71
<b>Total OPEX, 2020 USD/h</b>	<b>261.11</b>	<b>237.24</b>

Table 13 gives the LCOC values for both Pathways, and as can be seen, the LCOC (54.01 2020-USD/ton-CO<sub>2</sub> captured) of Pathway (i) is greater than the LCOC (39.90 2020-USD/ton-CO<sub>2</sub> captured) of Pathway (ii). This is not surprising considering the higher CAPEX and OPEX associated with Pathway (i) than Pathway (ii).

In addition, Pathway (ii) can capture 8.19 ton/h of CO<sub>2</sub> and produce 15.62 ton/h of saleable NaHCO<sub>3</sub> solid nanomaterials, and Pathway (i) can capture 8.21 ton/h of CO<sub>2</sub> with 99.95 mol% purity, which can be used or sent for sequestration.

**Table 13** LCOC of Pathways (i) and (ii).

	Pathway (i)	Pathway (ii)
Capacity factor, $f_c$		0.8
Discount rate $i$ , 1/year		10%
Lifetime $N$ , year		30
Capital recovery factor $f_{CR}$ , 1/year		0.106079
$\dot{m}_{CO_2}$ , ton/h	8.21	8.16

NaHCO <sub>3</sub> produced, ton/h	-	15.62
CAPEX to LCOC, \$/ton-CO <sub>2</sub>	22.20	10.92
OPEX to LCOC, \$/ton-CO <sub>2</sub>	31.81	28.98
<b>Total LCOC, \$/ton-CO<sub>2</sub></b>	<b>54.01</b>	<b>39.90</b>

### 5.3 Sensitivity Analysis for Pathway (ii)

We have conducted sensitivity analysis for pathway (ii). The CAU unit was 6 m diameter packed with Mellapak 250Y to a height of 36 m. The power plant capacity, SGS molarity, and SGS flow rate were varied to study their effects on the TEA (CAPEX, OPEX, and LCOC), hydraulics (two-phase pressure drop and liquid holdup), and the normalized specific wetted area of the packing. These results are given in Table 14, Table 15, and Table 16, where all TEA costs are in 2020-USD. Table 14 shows that increasing the power plant capacity at 3 M (mol/L) of SGS and about 90 mol% CO<sub>2</sub> capture, increases the CAPEX, OPEX and liquid holdup, CAU pressure drop and normalized packing specific wetted area, but decreases the LCOC of the process. These data are in agreement with previous finding for pre-combustion CO<sub>2</sub> capture applications presented by [92], Ashkanani, Wang [94]. Table 15 shows that increasing the SGS molarity in mol/L increases the CO<sub>2</sub> capture efficiency, CAPEX, OPEX, liquid holdup, and normalized packing specific wetted area, but decreases the LCOC of the process. Table 16 shows that increasing the SGS flow rate in L/s increases the CO<sub>2</sub> capture efficiency, CAPEX, OPEX, liquid holdup, and the normalized packing specific wetted area, but decreases the LCOC of the process.

**Table 14** Effect of power plant capacity on the process TEA, hydraulics, and the normalized packing specific wetted area (SGS = 3M).

Plant Capacity (MWe)	CO <sub>2</sub> absorption efficiency (%)	CAPEX (Million USD)	OPEX (USD/h)	LCOC (USD/ton.CO <sub>2</sub> )	Liquid holdup (%)	CAU pressure drop (Pa)	a <sub>w</sub> /a (%)
6	90.29	5.01	75.83	46.03	3.61-4.04	161	15.7-16.2
10	90.73	5.91	89.40	39.90	4.27-4.83	396	19.3-20.0
30	90.08	9.51	143.93	33.61	6.10-7.04	2,989	30.0-31.1
60	90.07	14.45	218.69	32.06	7.01-8.18	7,568	35.6-37.1
120	90.86	23.99	363.07	31.66	7.66-8.85	16,079	39.5-41.1

**Table 15** Effect of SGS molarity on the process CO<sub>2</sub> absorption efficiency, TEA, hydraulics and the normalized packing specific wetted area.

SGS molarity, (mol/L)	CO <sub>2</sub> absorption efficiency (%)	CAPEX (Million USD)	OPEX (USD/h)	LCOC (USD/ton.CO <sub>2</sub> )	Liquid Holdup (%)	CAU pressure drop, (Pa)	a <sub>w</sub> /a (%)
1.0	44.38	5.72	86.51	55.88	4.09-4.26	353	17.1-17.34
1.5	59.89	5.80	87.74	47.87	4.11-4.36	362	17.5-17.9
2.0	72.18	5.84	88.47	44.01	4.15-4.49	372	18.1-18.5
2.5	82.22	5.88	89.04	41.51	4.21-4.65	384	18.6-19.2
3.0	90.73	5.91	89.40	39.90	4.27-4.83	396	19.3-20.0
3.5	97.99	5.92	89.55	38.70	4.37-5.05	407	20.0-20.8

**Table 16** Effect of SGS solvent flow rate on the process CO<sub>2</sub> absorption efficiency, TEA, hydraulics, and the normalized packing specific wetted area (SGS = 3 M).

SGS flow rate (L/s)	CO <sub>2</sub> absorption efficiency (%)	CAPEX (Million USD)	OPEX (USD/h)	LCOC (USD/ton.CO <sub>2</sub> )	Liquid holdup (%)	CAU pressure drop (Pa)	a <sub>w</sub> /a (%)
20	63.65	5.60	84.76	45.28	3.86-4.31	387.85	16.5-17.2
22	69.30	5.66	85.72	43.78	3.96-4.41	389.84	17.1-17.8
24	74.83	5.72	86.65	42.54	4.05-4.50	391.76	17.7-18.3
26	80.26	5.79	87.57	41.50	4.13-4.58	393.51	18.3-18.9
28	85.56	5.85	88.50	40.63	4.20-4.66	394.98	18.8-19.4
30	90.73	5.91	89.40	39.90	4.27-4.83	395.91	19.3-20.0

## 6. Concluding Remarks

In this study, a unique process using aqueous SGS for CO<sub>2</sub> capture from a split stream of actual flue gas containing 0.0023 mol% SO<sub>2</sub> and 13.33 mol% CO<sub>2</sub> was developed in Aspen Plus v.10. The flow rate of the split stream used was maintained at 12.43 kg/s and the SGS flow rate was varied to

capture more than 90 mol% CO<sub>2</sub> from the split flue gas stream. The process has two Pathways (i) to capture and prepare CO<sub>2</sub> for sequestration and (ii) to produce NaHCO<sub>3</sub> nanomaterials. Since the raw flue gas contained SO<sub>2</sub>, a WU was designed to remove all SO<sub>2</sub> before CO<sub>2</sub> capture in both Pathways.

The physico-chemical properties of the aqueous SGS solutions were obtained from the literature, regressed, and implemented in Aspen Plus v.10. The CO<sub>2</sub>-SGS reaction kinetics were also obtained and the system hydraulics (two-phase pressure drop and liquid holdup), and mass transfer characteristics (gas-side and liquid-side mass transfer coefficients and the normalized specific wetted surface area of the packing) were calculated. The total CAPEX, OPEX, and LCOC of the two Pathways were also calculated. The WU and CAU are counter-current fixed-bed absorbers packed with Mellapak 250Y.

Under the operating conditions used, no flooding was observed in WU and CAU. The two-phase pressure, liquid holdup, liquid-side and gas-side mass transfer coefficients and the normalized specific wetted area of packing in the WU and CAU were obtained and presented as a function of the packing height. The data in both units indicated that the gas-side mass transfer coefficients were much greater than the liquid-side mass transfer coefficients and the behavior of the liquid holdup and the normalized specific wetted area of packing were similar in both units. Our hydraulics predictions, however, need to be validated against actual data from an industrial post-combustion power plant.

Aspen Plus TEA calculations showed that considering a plant lifetime of 30 years, an annual discount rate of 10%, a capacity factor of 0.8, an annual O&M cost factor of 4% of the total CAPEX in 2020-USD, and a capital recovery factor of 0.106, the CAPEX, OPEX and LCOC of the Pathway (i) were (\$12,039,251), (261 \$/h), and (54.01 \$/ton of CO<sub>2</sub> captured), respectively. Also, the CAPEX, OPEX and LCOC of the Pathway (ii) were (\$5,908,000), (237 \$/h), and (39.90 \$/ton of CO<sub>2</sub> captured), respectively. In addition, Pathway (i) can capture 8.21 ton/h of CO<sub>2</sub> with 99.95 mol% purity, which are ready for subsequent sequestration, however, Pathway (ii) can capture 8.16 ton/h of CO<sub>2</sub> and produce 15.62 ton/h of saleable NaHCO<sub>3</sub> solid nanomaterials.

## 7. Future Prospects and Challenges

Among the 20 known amino acids, Gly and Alanine (ALA) react with CO<sub>2</sub> to produce nanomaterials, which exhibit a phase separation (CO<sub>2</sub>-rich and CO<sub>2</sub>-lean phases) and subsequently there are potential opportunities for using aqueous amino acid salts for CO<sub>2</sub> capture and producing high-value nanomaterials, which can be sold to offset the overall cost of the capture process. The selection of the base (NaOH, KOH, etc.) to react with the amino acid could open more venues in CO<sub>2</sub> capture, while producing vital products with many diverse applications. For instance, sodium bicarbonate could be used in baking and personal hygiene, as food additives, a disinfectant, a fire extinguisher, and as a promising agent for cancer therapy [3]. Also, the potassium bicarbonate could be used for food and drink, fire extinguisher, and agriculture, especially for acidic soil neutralization and organic farming [99, 100]. The choice of the base could also enhance its reaction kinetics with CO<sub>2</sub> so that amino acid salts could be used to capture small concentrations of CO<sub>2</sub> whether from Natural Gas Combined Cycles (NGCC) or direct air capture (DAC). The challenges in using amino acid salts for CO<sub>2</sub> capture reside in maintaining bicarbonates in the presence of side-products and separating the nanomaterials from the liquor in a continuous process. Also, the availability of ample capacity of NaOH could be a challenge, however, our estimates showed that NaOH available in the

market was more than enough for our process. Another challenge is the ability of amino acids to achieve a net negative CO<sub>2</sub> emission, which could be assessed by performing Life Cycle Analysis (LCA) along with the TEA of the process.

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### **Author Contributions**

Mr. Rui Wang collected the data and performed the TEA; and Drs. Husain Ashkanani, Bingyun Li, and Badie Morsi revised and polished the manuscript.

### **Competing Interests**

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