Process intensification of CO_2 desorption

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Highlights

- Solvent regeneration is the major energy drawback of post-combustion CO_2 capture.

- PI technology can reduce the energy penalty of CO_2 desorption by 65 %.

- Performance metrics are proposed to evaluate the PI technologies in CO_2 desorption.

Abstract

Anthropogenic climate change due to, amongst other, unhindered CO_2 emissions is a major worldwide concern. The post-combustion capture (PCC) process using a solvent, known as chemical absorption, is the most effective way today to reduce CO_2 emissions from large point sources. However, high capital investment costs when using the conventional packed bed absorber/desorber technology, and high energy requirements during solvent regeneration are the primary obstacles for its large-scale implementation. Different process intensification (PI) technologies to desorb CO_2 from the solvent have been introduced to mitigate the energy consumption compared to the conventional packed bed technology. This article reviews different technologies that can be used for CO_2 desorption intensification. In this context, rotating packed beds, microreactors and membrane contactors have been explored as potential alternatives to intensify the desorption due to their superior mass and heat transfer. Alternative energy sources like ultrasound and microwave have also been used to improve the desorption performance of conventional equipments. PI can also be realized by using novel solvents with improved desorption kinetics in combination with intensified equipment. Thus, in this article, a comprehensive assessment of different existing PI technologies based on regeneration energies and regeneration efficiencies relative to conventional technology is presented. The intensification of mass transfer for the different technologies is compared and a new parameter, named the *Regeneration Factor* is proposed to evaluate the performance of PI equipment. This study outlines the advancements in process intensification of CO_2 desorption technologies to date and presents an overview of the merits and limits of all technologies.

Keywords: CO_2 capture; Process Intensification (PI); Solvent regeneration technologies; Energy requirements; Performance metrics.

1. Introduction

Global warming and associated climate change are a major worldwide concern. Although COP21 set forth an ambitious target of restricting temperature rise to below 2 °C by 2100, the COP26 held in Glasgow in 2021 has now set a tougher target.[1] The objectives defined should result in net-zero emissions by 2050 to restrict the average rise to 1.5 °C. In the short term, the goal is now phasing out of coal-derived power, preventing deforestation, promoting electric vehicles, and renewables by 2030. Uninhibited emissions of greenhouse gases (GHGs) like Carbon Dioxide (CO_2), Methane (CH_4), Nitrous oxide (N_2O), and water vapour (H_2O) are the major contributors. Among them, carbon dioxide (CO_2) is the largest contributor. Its emissions have strongly increased over the last decades mainly due to the increased burning of fossil fuels.[2-4] Although fossil fuels such as coal, natural gas, and oil have been the primary energy sources (>80% of consumed) for several decades, the excessive combustion of fossil fuels has resulted in an increase of the CO_2 concentrations in the atmosphere from 339 ppm in 1980 to 412 ppm in 2020.[4-8] Such high CO_2 concentrations in the air (7-10%) endanger human life and health.[9] Thus, this excessive amount of CO_2, being two-thirds of all GHGs emissions, needs to be reduced from industrial and human activities.[10,11]

Among the GHGs, CO_2 is a very stable molecule ($\Delta G^{o}_{f,\ 298K} = -394.4$ kJ/mol), with a very long half-life of 120 years.[2] Thus, technologies such as Carbon dioxide Capture and Storage (CCS) and Carbon dioxide Capture and Utilization (CCU) have been developed.[12,13] In the CCS process, the captured CO_2 at the point source (such as power plants, steel, and cement industry) is transported and stored in onshore or offshore geological formations, oceans, and mineral carbonates.[9] CCS must be considered as a rapid solution to CO_2 mitigation.[14] In CCU, the captured CO_2 is converted into value-added products, thus reducing the net cost of CO_2 removal.[12,15] CCU needs to become economically viable by reducing the high electricity/energy demands and other major investment costs associated with the conversion of this thermodynamically very stable CO_2.[14] Carbon capture technologies can be further classified into pre-combustion, oxy-combustion, and post-combustion capture. In pre-combustion, the carbon-rich solid fuel undergoes gasification to produce syngas ($CO+H_2O$) in the first stage. Subsequently, the syngas is fed into a water-gas-shift reactor, wherein the product CO_2 is efficiently captured, whereas H_2 is utilized as a source of energy. However, being a two-stage process, such technologies involve significant capital costs.[16,17] Oxy-combustion technology uses pure oxygen instead of air to burn fossil fuels and aims to remove CO_2 at low temperatures.[16] Post-combustion capture (PCC) aims to trap CO_2 after the combustion of fossil fuel. It is the most efficient strategy to curb CO_2 emissions as existing coal or natural-gas-fired power plants do not require significant retrofitting.[9,18]

Among PCC technologies such as adsorption, absorption, and membrane separation, the use of amine-based solvents for the chemical absorption process is the most mature and the most used one in an industrial environment (Figure 1).[8,19,20] Mass transfer rates, stability in a high-temperature environment in the presence of O_2, solids generation, volatility, toxicity, biodegradability, and cost of solvent are to be considered when designing new CO_2 absorbents.[21,22] Because of its low cost and high reactivity with CO_2, typically primary amines like Monoethanolamine (MEA) are employed as a benchmark solvent.[23] For instance, MEA offers superior CO_2 mass transfer rates, is low-cost, and is biodegradable. However, it shows moderate oxidative and thermal degradation, as well as moderate toxicity. When used in larger amounts, it can also be corrosive. However, the high heat of reaction with CO_2 (85.6 kJ/mol CO_2) is a significant disadvantage, leading to increased energy requirements during desorption.[22]

Various technologies developed for the CO_2 absorption process have been widely studied and reviewed in the past.[24-29] Among them, packed bed columns are the most common equipment used for chemical absorption.[23,30,31] They are capable of handling large gas volumes and can be adapted to any existing plant. However, the relatively low mass transfer rates in packed bed columns result in the need for large absorber and desorber columns, thus accounting for 70% of the total capital expenditure (CAPEX).[6,32] A considerable flow of steam is also needed for solvent regeneration in the desorber column. The latter is a major energy drawback accounting for 70-80% of the total operational expenditure (OPEX).[18,33] The energy required to regenerate a solvent flow containing 30wt.% MEA can be up to 3.2-4.5 MJ/kg CO_2 which is far more than the theoretical value reported to be 0.436 MJ/kg CO_2.[34,35] The highly stable nature of MEA is attributed to the hydrogen in the alkyl group that results in a difficult breakdown of the bonds between CO_2 and MEA. It causes slower desorption rates and hence higher energy consumption. Additionally, the solvent regeneration temperature is high, 373-413K, resulting in solvent degradation. Thus, to compensate the solvent loss and corrosion caused by the formation of harmful degradation by-products increases the OPEX.[36-38] Although chemical absorption methods can achieve a high capture efficiency, the considerable amount of energy required for solvent regeneration is the main drawback of this technology.[39]

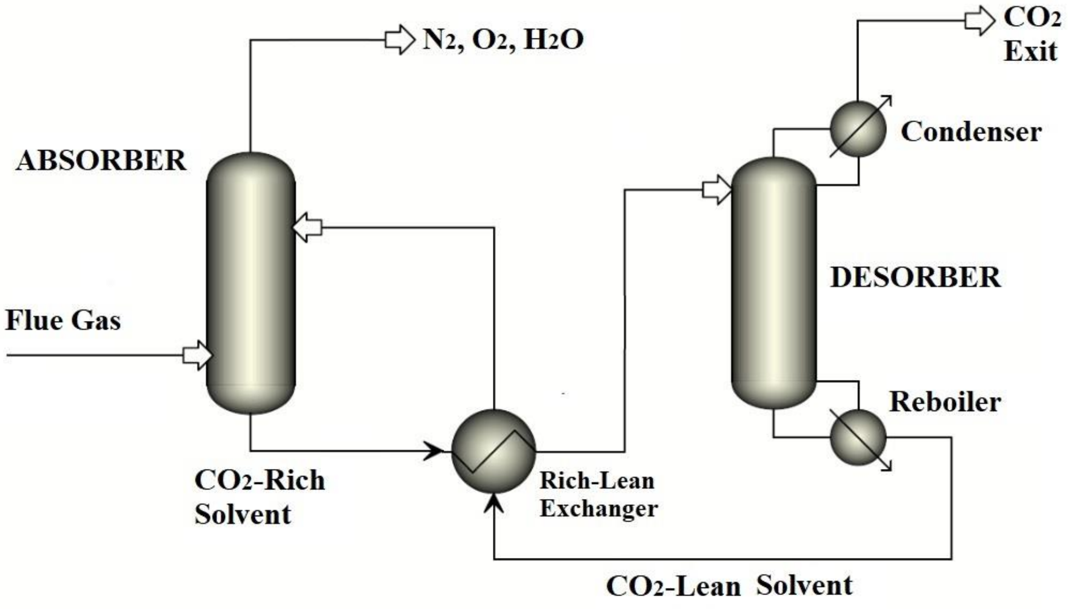


Figure 1. Simplified scheme of chemical CO_2 absorption and desorption.

Approximately one-third of a typical coal-fired power plant's net electricity output efficiency is wasted due to the high energy consumption of the desorption process.[40] When CCS is implemented on a power plant, the generation of energy reduces by 20-30%.[41,42] Since CO_2 capture and further compression of CO_2 require both thermal energy and electricity, power plant output would decrease. The "efficiency penalty" refers to the difference in performance between power plants with and without CCS. When the desorption process is carried out at high pressures, the pressure difference at compression is reduced. This results in a lower efficiency penalty. At low operating pressures, however, regeneration temperature is lower than desired. This reduces the regeneration performance of the rich solvent and increases the regeneration energy, resulting in a larger efficiency penalty.[43] To estimate the efficiency of a power plant, it is recommended to calculate total equivalent work including pump and compression work and heat duty work. This should be done on an electricity basis. Pumps and compressors demand energy directly from the power plant. Reboiler utilizes steam which may create electricity in the plant turbines.[44,45] In general, 40% thermal efficiency is used for the conversion of thermal heat into electricity since it is a reasonable assumption for the various types of power plants with CCS.[46]

To resolve existing problems, optimization strategies for the desorption, or solvent regeneration step, strategies like advanced heat integration, inter-cooling, rich solvent splitting, multi-pressure stripper can be used. Although the operating costs decrease, the technology still suffers from technical and economic challenges resulting in difficult operation and control of an industrial plant.[31,34,47,48] On the other hand, process intensification (PI) techniques could significantly reduce the size of the desorber and hence energy requirement. PI is a strategy that aims to significantly enhance chemical processes by reducing the equipment size/capacity ratio, energy consumption or waste output.[49] It is also more profitable, environmentally benign, and safer technologies. One of the main goals of PI is the reduction of the volume of process equipment, which requires improved mixing and heat/mass transfer characteristics.[50] The improved gas-liquid interactions in PI technologies make them promising solutions for the high energy penalty of solvent regeneration, without affecting the production capacity. This ultimately leads to lower CAPEX and OPEX for solvent regeneration.[34,51]

Given the fact that the regeneration of the solvent is capital intensive, the limited focus on the desorption step in the past can only be considered surprising. But it can probably be ascribed to the complex hydrodynamics, linked to the reversible reactions occurring in the desorber.[52] As a result, the number of review papers considering only the desorption process is limited.[34, 53-56] However, a better understanding of the desorption process could result in a significant improvement of the combined absorption and desorption process wherein a reduction of the overall energy efficiency of the process is a major goal. This article aims to provide a comprehensive state-of-art review of the CO_2 desorption process focusing on process intensification technologies. In the first part, the state of that of the conventional packed bed technology is presented. Following this, the performances of process intensification technologies compared to the conventional packed bed technology are discussed. To compare all process intensification technologies, a new parameter, the regeneration factor, which includes regeneration energy, efficiency, and desorber volume, is introduced. It has been assessed for different PI equipment.

2. Conventional Packed Bed Technology

The conventional packed bed technology is the most widely used technology for solvent regeneration (see Figure 1). A CO_2 enriched solvent (the ‘rich’ solvent) stream coming from the absorption section is preheated to the desired operating temperature. Subsequently, it is brought in countercurrent contact with steam generated in a reboiler in a desorber column. The countercurrent contact between these two streams assures a continuous driving force for mass and heat transfer over the height of the column.[57-59] The carrier steam supplies the energy needed to strip the CO_2 that is chemically bound to the solvent.[9] The CO_2 leaving over the top of the desorber is then compressed and transferred for storage (CCS) or utilization (CCU), while the ‘lean’ solvent is recycled to the absorber column.[34]

Mass transfer characteristics significantly influence the capital cost of the CO_2 desorption column and subsequent the solvent regeneration costs.[60] The CO_2 absorption process is exothermic. Whereas, the desorption step, in which CO_2 is transferred from the liquid (CO_2-rich solvent) to the gas phase (CO_2 and H_2O), is an endothermic process. In most desorption studies this interphase mass transfer is described using the two-film model wherein the mass transfer is assumed to be linear in the interface. Further, the CO_2 concentration in gas and liquid phases in the two-film interphase are assumed to be in equilibrium.[60,61] However, film thicknesses and interfacial area are difficult to measure.[61] Thus, the mass transfer rate is calculated based on gas and liquid bulk concentrations, using an estimated overall volumetric liquid mass transfer coefficient K_La_V, based on the overall liquid mass transfer coefficient, K_L and the interfacial area, a_V. Weiland et al. studied CO_2 desorption mass transfer rates when using MEA solvents in a packed bed column.[62] The volumetric overall liquid mass transfer coefficient K_La_V is found to depend linearly on the liquid flow rate, as a consequence of an increase in both the liquid overall mass transfer coefficient, K_L and the interfacial area, a_V.[62] Mass transfer correlations for packed bed columns and PI technologies are given in Supporting Information (Table S1). The liquid-only mass transfer (k_La_V) correlation for a packed bed desorber accounts for simultaneous mass, heat, and momentum transport.[63] Further, k_La_V increases with temperature and liquid flow rate but decreases with amine concentration.[63] Sakwattanapong et al. employed MEA as the base solvent in a lab-scale packed bed desorber.[64] Following these, Tobiesen et al. developed a numerical mass transfer model for a CO_2 desorption column that takes into account desorber packings, reboiler, and condenser.[65]

Moreover, MEA was used as a reference solvent in several pilot plant studies.[66-69] Kwak et al. investigated the CO_2 absorption/desorption for MEA in a pilot plant and studied the effect of operating conditions such as temperature, pressure, and flow rate on the regeneration energy.[68] The required regeneration energy was found to increase with increasing desorber pressure. It was highly recommended to design the bottom of the desorber tower with corrosion-resistant materials due to higher temperatures in this section.[68] Lemaire et al. used an advanced MEA process with the addition of high-performance inhibitors to be able to use MEA at concentrations higher than 30wt.% without observing degradation or corrosion. By doing this, the reboiler heat duty was reduced to 3.1-3.3 MJ/kg CO_2.[70] A recently published review study covering a wide variety of pilot plant studies with MEA is highly recommended to readers for detailed information on the CO_2 desorption process.[56] Conventional packed bed technology when using MEA for CO_2 desorption is considered as the reference technology. In the following section, several PI equipment and methods are evaluated with respect to the existing packed bed technology.

3. Process Intensification Technologies

Process intensification (PI) for CO_2 desorption broadly includes the application of technologies that minimize the size of the equipment by employing an intensified field of centrifugal, electrical and microwave. Usually, it employs a combination of processes to intensify gas-liquid mass and heat transfer in a single high-performance equipment to simplify processes. This not only results in reducing the physical scale but also energy consumption of the equipment.[71] To date, three PI approaches have been widely applied to intensify CO_2 desorption processes: i) use of process-intensifying equipment with enhanced mass and heat transfer characteristics ii) use of process-intensifying methods such as ultrasound and microwave iii) use of process intensifying materials including novel solvents, blended amines and catalysts requiring lower regeneration energy,[29,49,72] as illustrated in Figure 2. An overview of process intensification technologies focusing on their advantages and disadvantages is summarized in Table 1. An overview of their operating conditions is given in Table 2. A summary of patents provided in Supporting Information (Table S2).

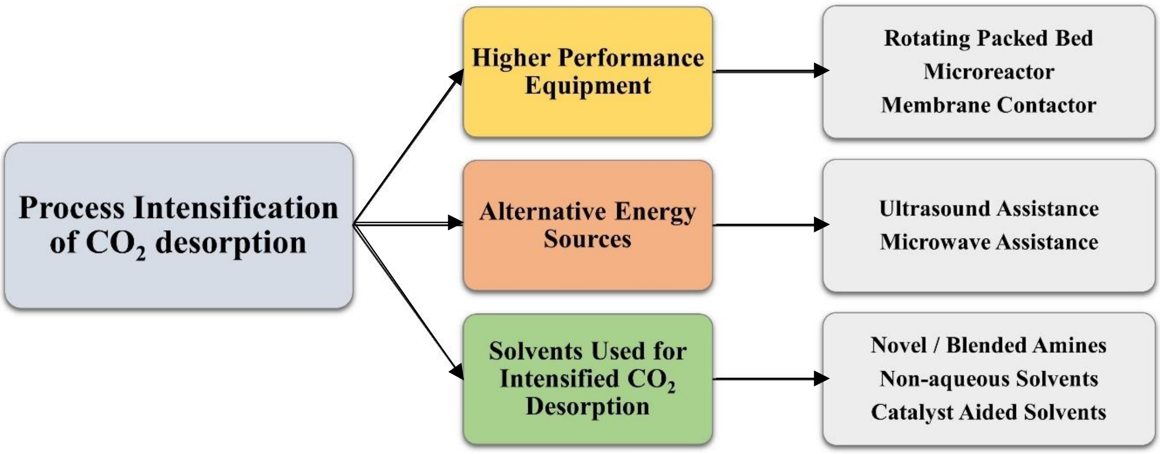


Figure 2. Process Intensification of CO_2 desorption.

Prior to a more detailed discussion of different PI technologies, an overview of their regeneration performance is presented in Figure 3, which depicts a range of regeneration efficiency values with respect to regeneration energy. It should be noted that each of the shaded regions in Figure 3 compiles results found in literature for conventional packed bed technology and different PI technologies. Novel PI equipment is assessed in comparison to the conventional packed bed technology. The relationship between its regeneration energy required to desorb one kg of CO_2 from the liquid and its efficiency (percentage of CO_2 present in a regenerated solvent) is shown. As the total energy for solvent regeneration is supplied by hot steam passing through a reboiler at the bottom of a desorber column, the energy consumption for regeneration is also referred to as reboiler heat duty.[54,64] Regeneration energy comprises contributions from the heat of vaporization (~23%), the sensible heat of the solvent (~26%), and the heat of desorption (~51%). The heat required to generate volatile components (generally water) and realize the operating CO_2 partial pressure needed to strip CO_2 is known as the heat of vaporization. The sensible heat is the heat necessary to bring the CO_2-rich solvent to its boiling point. The heat of the desorption, which is mainly determined by the solvent used, is the energy necessary to break the chemical bonds between CO_2 and the absorbing solvent.[64,73,74]

To gauge the regeneration efficiency with respect to the regeneration energy required, a detailed review of PI desorption technologies was performed. In what follows an overview of the literature found for PI technologies, with an approach summarized in Figure 2, is made. Based on this overview, these PI technologies can then be classified as shown in Figure 3. The conventional packed bed technology, as discussed in Section 2, is selected as the reference case in Figure 3. Regeneration energies in the range of 3.7-9 MJ/kg CO_2 and regeneration efficiencies in the range of 34-60.9% were reported for the conventional packed beds employing MEA.[64,65,75]

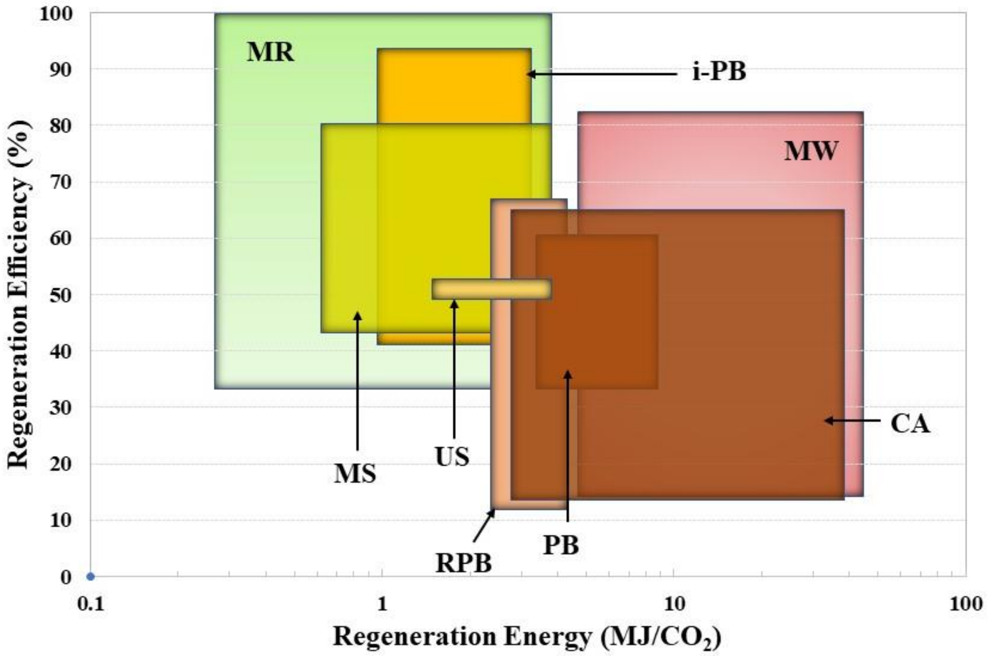


Figure 3. Regeneration performance of technologies based on regeneration energy and the efficiency.

PB=Packed Bed, i-PB=Intensified Packed Bed, RPB=Rotating Packed Bed, MW=Microwave, US=Ultrasound-Assisted Regeneration, MR=Microreactor, MS=Membrane Stripping, CA=Catalyst-Aided Solvent Regeneration.

3.1 High-Performance Equipment

3.1.1 Rotating Packed Bed Technology

In rotating packed bed technology (RPB), which is a high gravity (HIGEE) reactor technology, the gravity field in the packed bed columns is replaced by a centrifugal field resulting in intensified equipment for CO_2 desorption.[76] Besides, the fact that solvent is forced through a packed bed, the solvent is stretched and disintegrated into small droplets and thin liquid films. Consequently, there is a large contact area between the gas and liquid phases with a faster renewal of the gas-liquid interphase, resulting in high mass transfer rates. It also results in an increased heat transfer rate, reducing the heat losses compared to a conventional packed column. RPB is thus expected to reduce the regeneration energy consumption as well as the required unit volume for desorption due to its superior heat and mass transfer. Jassim et al. were the first to study the use of a RPB unit for CO_2 desorption using the MEA solvent. The height and diameter of the packed bed were observed to reduce by a factor of 8.4 and 11.3, compared to a conventional packed bed operated under identical desorption conditions.[76] Cheng et al. used a back pressure regulator at the RPB outlet to operate desorption at varying pressures (Figure 4a).[77] More specifically, the effects of the rotational speed, liquid flow rate, reboiler temperature, and pressure on regeneration efficiency, regeneration energy, and the volume of the desorber were analyzed. The volumetric liquid-only mass transfer coefficients in RPBs, using a 30wt.% MEA solvent was reported to be 3 times higher than in a conventional packed bed desorber.[62,77] The higher mass transfer coefficient was found to lower the heat losses compared to the packed bed technology. The latter was attributed to a reduced desorber volume and external surface area.[77] The reported RPB volume decreased by a factor of ten compared to conventional packed bed technology. As the liquid flow rate increased, the contact time between gas and liquid decreased, resulting in a decreased regeneration efficiency and increased regeneration energy.[77] The mechanical energy needed for rotation however was found to be much lower than the heat losses and regeneration energy and can thus be neglected when analyzing the total energy required for the RPB desorption process. Regeneration energy was found to be 4.51 MJ/kg CO_2 for MEA, whereas for a blended solvent of piperazine (PZ) and diethylenetriamine (DETA) only 3.40 MJ/kg CO_2 is consumed. Furthermore, not only the heat of reaction and heat capacity, but also the heat of vaporization when using PZ and DETA are lower than the corresponding values for MEA, when operating at higher pressures.[77] The effect of varying operating pressure (2-3.5 atm) on regeneration energy and efficiency of PZ-DETA solvent was also evaluated.[4] It was found that increasing the operating pressure results in an increased ratio of the sensible heat to reaction heat and a reduced ratio of the vaporization heat to reaction heat. The latter resulted in a reduced regeneration efficiency with lower amount of desorbed CO_2. The lowest regeneration energy (3.2 MJ/kg CO_2) was reported at 2.5 atm, at which the effect of operating pressure on the sensible heat and heat of vaporization was similar. Significantly, the regeneration energy consumption is 54.8% lower than the value for MEA (7 MJ/kg CO_2).[4]

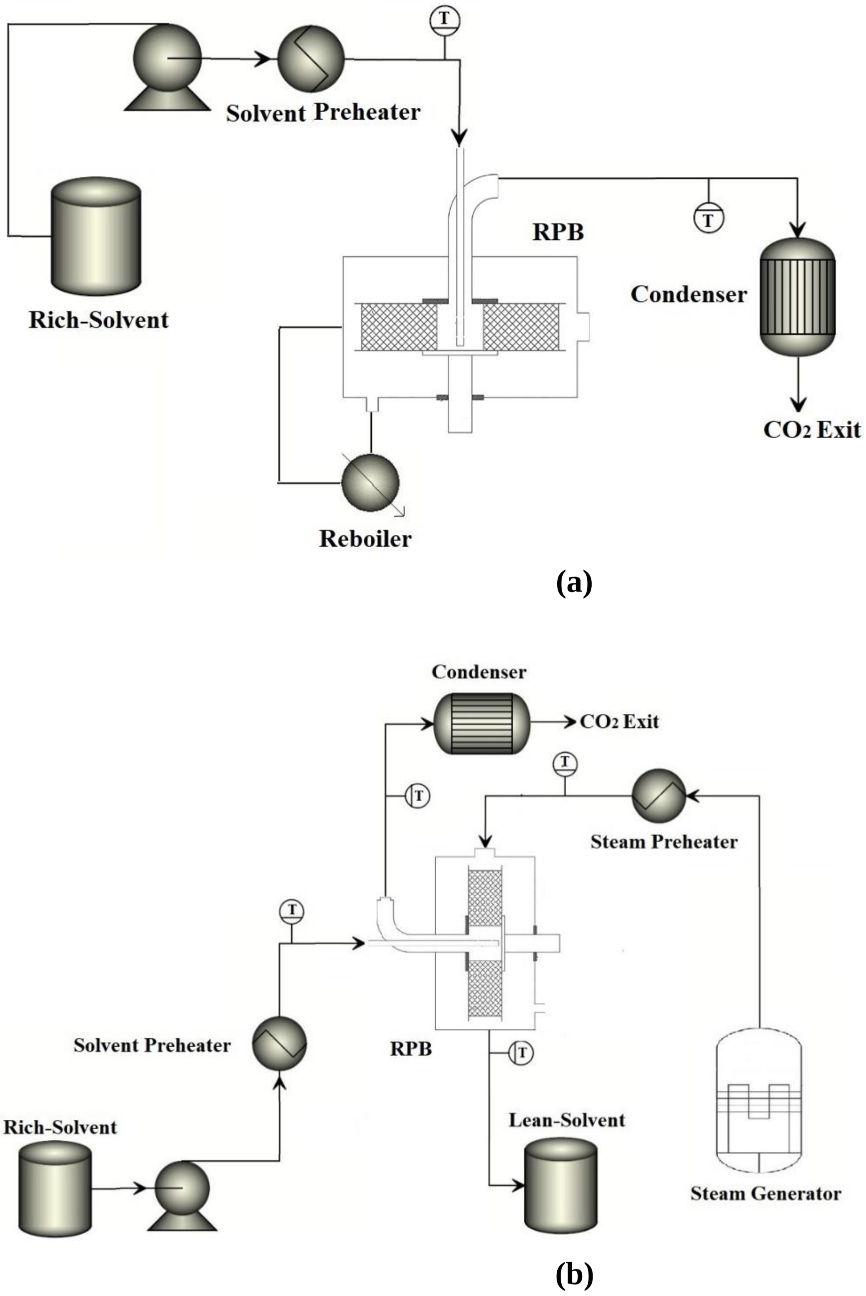


Figure 4. Schematic flow diagram for CO_2 desorption in a) RPB b) RPB+DSS.

Recently, Wang et al. studied CO_2 desorption in an RPB with Direct Steam Stripping (DSS). A typical schematic flow diagram of RPB with DSS is presented in Figure 4b.[5] It was reported that the use of DSS increased the driving force for mass transfer and decreased the energy consumption by 20–30% compared to the Conventional Reboiler Regeneration (CRR) process. In the latter study, CO_2 desorption was intensified in two ways. A novel tri-blended solvent, i.e.

the 2-amino-2-methyl-1-propanol (AMP), 2-(2-aminoethylamino) ethanol (AEEA), N-methyl pyrrolidone (NMP), was used. Two high-performance mass and heat transfer devices, i.e. an RPB and DSS were tested. The tri-blended solvent was found to be beneficial for solvent regeneration, which was attributed to the formation of unstable carbamates, and steric hindrance of AMP, resulting in an easier breakdown of the bonds between CO_2 and the solvent. NMP, added in the tri-blended solvent, has a high boiling point and a low heat capacity, resulting in less solvent evaporation and heat requirement to increase the temperature of the absorbent, respectively. Overall, the regeneration energy is lower compared to the traditional aqueous amine solvents. The use of the novel tri-blended solvent and the introduction of DSS were both observed to have a significantly favorable effect on the regeneration energy, – a reduction of 36.6% for MEA in conventional packed bed and 17.4% for MEA in RPB combined with DSS.[5] The Carbon Clean company uses the RPB technology commercially for CO_2 absorption and desorption when utilizing a commercial amine buffer solution. The employment of the advanced solvent and RPB technology together is reported to reduce the overall cost by 50%, while maintaining high performance.[78] In conclusion, the use of an RPB for CO_2 desorption is promising, but the number of studies is still limited compared to the use of RPB for CO_2 absorption.[56] The consumption of electricity to drive the rotor of the RPB, in addition to the steam consumption for CO_2 desorption, may result in increased OPEX. The challenges get compounded due to the high flow rates in an RPB, the short gas-liquid contact times cause the low regeneration efficiency, which for sure needs further research and development.[55]

3.1.2 Microreactor Technology

The possibility to use microreactor technology for PI of CO_2 desorption was studied, given its several benefits like- increased separation efficiency, reduced equipment size, and lower energy consumption.[79-82] Due to an enhanced mass transfer rate between phases and a high heat transfer capability, the microreactor technology is considered a technology fit for CO_2 desorption wherein the requirement for interfacial contact of gas and liquid is also high.[54] Studies for CO_2 desorption in microreactor technology were largely spearheaded by two research groups (Tables 1 and 2).[71,83] Figure 5 shows a typical experimental setup where a CO_2-loaded amine solvent is introduced in a microreactor with a syringe pump. The rich solvent inside the microreactor separates into two phases (CO_2 and lean solvent) at operating desorption temperature (50-100 °C). The desired desorption temperature is maintained by heating with an electric coil wrapped around the microchannel. CO_2 and the lean solvent are discharged over the top and the bottom of a phase separator at the microreactor outlet.[61] A parametric study investigating the effect of varying the methyldiethanolamine (MDEA) concentration, the rich solvent flow rate, the CO_2 loading, and the desorption temperature was performed by Liu et al.[71] They also studied gas-liquid hydrodynamics in a microreactor. Although superior performances in terms of high mass transfer were determined in the microchannel geometry, the desorption efficiency was limited to 35%. In yet another study by Liu et al., heat transfer performance and energy consumption were investigated.[6] The rich solvent flow rate was found to have profound influence on heat transfer characteristics. It was attributed to the decreasing thickness of the thermal boundary layer with increasing flow rate. Although heat transfer coefficients in the nucleate boiling region increased with increasing temperatures and flow rates, the lowest energy consumption was 3.7 MJ/kg CO_2 which is higher than the minimum value of the base case (3.2-4 MJ/kg CO_2).[6,65] More recently, Aghel et al. examined the desorption of CO_2 from various aqueous and non-aqueous solvents (monoethanolamine, diethanolamine, methyldiethanolamine) in a microtubular geometry.[61,83,84] The energy consumption was found to vary linearly with temperature and solvent flow rate, but to be inversely affected by the amine concentration.[83] It was also reported that non-aqueous solvent solutions reduced the energy consumption by 73% compared to the aqueous solvent solutions. Further, the obtained values of 0.30 to 0.41 MJ/kg CO_2 for non-aqueous solvents are very close to the reported theoretical limit.[35,61,84] Although the microreactor technology offers higher regeneration efficiency with lower regeneration energies required, the actual implementation remains low. The latter must be attributed to the low capacity of the microreactor technology. For an economically viable throughput, several units operated in parallel would be necessary for industrial applications. However, a uniform flow distribution needs to be ensured in the distribution channels.[55] Thus, despite favorable characteristics of the microreactor for desorption, scale-up issues pose serious restrictions on large-scale application.

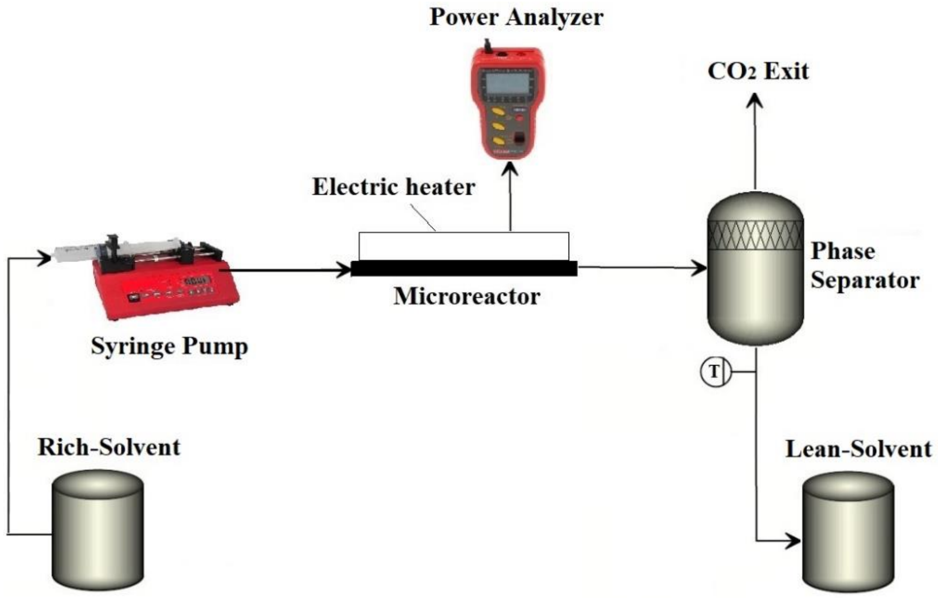


Figure 5. Schematic flow diagram for CO_2 desorption in a microreactor.

3.1.3 Membrane Stripping Technology

The membrane stripping process is considered a valuable technology for CO_2 desorption. The energy consumption is relatively low, and the regeneration efficiency is relatively high compared to other technologies. In membrane stripping, membrane contactors that allow CO_2 to permeate are used.[85] Figure 6 shows an experimental setup where a CO_2-loaded solvent is introduced in a membrane contactor.[86] For the membrane stripping technology, the overall mass transfer coefficient is found to depend on membrane resistance as well as gas and liquid film resistances.[87] Notably, the mass transfer resistance of the membrane itself contributes to the overall mass transfer coefficient by no more than 5%.[87-89] When using a membrane, the interfacial area is also estimated more easily as the membrane geometry itself determines the interfacial area. The interfacial mass transfer area of the membrane selected for research varies from 1500 to 3000 m^2/m^3. This is considerably higher than the interfacial area realized in conventional packed bed columns (100-800 m^2/m^3).[85] In membrane technology, the selected membrane is non-dispersive for the gas (except CO_2) and liquid phases. There is no entrainment, channeling, flooding, or foaming possible, which makes membrane technology advantageous compared to conventional columns.[90] Typically membranes for desorption have a high permeate flux and selectivity for CO_2, chemical and thermal stability, and enhanced hydrophobicity to prevent membrane wetting.[3] Among these characteristics, the most important factor is wettability because mass transfer decreases when the membrane pores become filled with liquid.[20]

Membrane vacuum regeneration technology with steam sweeping to remove the CO_2 that has been permeated through the membrane was studied by Fang et al. They used MEA as the solvent.[3] The regeneration performance of a cylindrical polypropylene (PP) hollow fiber membrane was determined.[3] Two modes of liquid flow, tube side and shell side, were investigated. Tube side flow was found to have a higher regeneration efficiency due to a lower mass transfer resistance. Optimizing the liquid flow rate is important as higher flow rates (50 ml/min) imply shorter residence and contact times resulting in a lower regeneration efficiency. The use of a longer membrane module to handle higher flow rates is an option but it also increases investment costs. Overall, the energy requirement for vacuum regeneration was reported to be 35.6% lower than the thermal regeneration energy need of a packed bed column.[3] Wang et al. developed an experimentally validated mathematical model to calculate the regeneration performance of CO_2 desorption for PP and polyvinylidene fluoride (PVDF) membranes to regenerate MEA.[91,92] The PP membrane was found to have longer stability and higher efficiency than the PVDF membrane. The energy consumption was also reported to be reduced by 28% compared to a conventional packed bed column.[92] A hydrophobic surface modifying macromolecule (SMM) membrane was used by Rahbari-Sisakht et al. Due to the hydrophobicity of the membrane, a high CO_2 desorption flux with 80% regeneration efficiency, even at high liquid flow rates was noted.[93] Solvents like potassium glycinate (PG), MEA, diethanolamine (DEA), and AMP can be used in combination with a PVDF membrane.[86] The CO_2 stripping flux and efficiency were found to increase with liquid temperature, pressure, and initial CO_2 loading, independent of the selected solvent.[86] Chan et al. used a polytetrafluoroethylene (PTFE) hollow fiber membrane combined with the high-temperature pressure swing method to remove the permeated CO_2.[94] It was reported to be the most energy-efficient method, with regeneration energies lower than 1 MJ/kg CO_2. The co-current flow of the liquid phase and gas phase in the membrane contactor resulted in an increased regeneration performance, leading to the higher driving force.[94] Moreover, ceramic hollow fiber membrane contactors were found to be applicable for gas-liquid stripping at high temperatures due to their high interfacial area per volume, their phase separation efficiency, and their excellent chemical and thermal stabilities. With increasing liquid flow rate, higher operating temperatures promote molecular diffusion and chemical kinetics resulting in high mass transfer efficiencies in a membrane contactor.[95] Readers are referred to recent reviews on membrane contactors for CO_2 separation wherein a detailed overview on membrane contactors for CO_2 capture is presented.[85,96] Even though CO_2 desorption when using membrane contactors provides lower regeneration energy consumption and higher regeneration efficiency compared to conventional packed bed technology, fouling, and wetting of the membrane pores could reduce the mass transfer, resulting in instability. An increased membrane module length is necessary to ensure high contact areas, which increases CAPEX.[3,20]

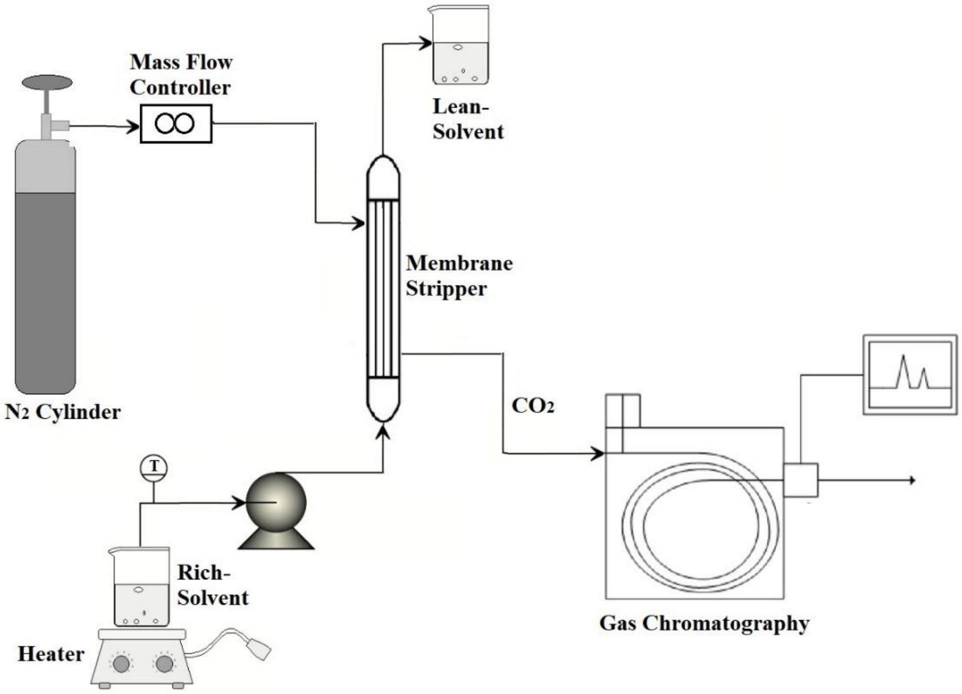


Figure 6. Schematic flow diagram for CO_2 desorption in a membrane contactor.

3.2 Alternative Energy Resources

3.2.1 Ultrasound-Assisted Stripping

Ultrasound-assisted stripping is one of the most promising process intensification technologies for CO_2 desorption. It uses cavitation, acoustic streaming, and heating effects instead of conventional heating in packed bed columns to strip CO_2 from the rich solvent.[97] These effects enable formation of millions of cavitation bubbles in the liquid phase which increase the interfacial area between gas and liquid. Cavitation bubbles can easily grow when free CO_2 diffuses to them. With the use of ultrasound, CO_2 is transferred from the liquid phase to the gas phase in the form of bubbles. Because these cavitation bubbles are initially in a vacuum condition, the driving force to transfer CO_2 to bubbles is considerable. As a result, the number of bubbles and the partial pressure of CO_2 inside the bubbles are critical factors for CO_2 stripping. These bubbles can also be unstable and create liquid micro-jets, which improves mass transfer during regeneration process.[98,99,100] It is known that there is a significant number of bubbles already in the reboiler section of conventional desorption column. Thus, using ultrasound in the reboiler section is intended to enhance the number of bubbles and promote the release of CO_2 from the liquid phase (Figure 7).[99] With ultrasound, desorption can proceed at low temperatures which reduces the energy consumption and CO_2 capture cost. Additionally, the CO_2 loading of the lean solvent is anticipated to decrease which does not alter the energy input in the reboiler section. This results in the reduction of the equipment size. Furthermore, this method offers advantages over current solvent regeneration techniques in terms of size, compact design, solvent degradation, and mass transfer improvement.[97]

The regeneration energy needed (1.5-3.6 MJ/kg CO_2) is lower than conventional packed bed columns (Figure 3). Regeneration efficiencies are also comparable to packed bed technology but remain lower than existing intensified technologies. However, a significant reduction of the CO_2 loading (0.2 mol CO_2/ mol amine) in the lean solvent can be expected when using this technology.[98] Abundance of cavitation bubbles in the liquid, due to the ultrasound treatment, induces a high interfacial area between gas and liquid which in turn leads to high mass transfer rates. Desorption can proceed at lower temperatures ($\leq$100 °C), resulting in less solvent degradation, and thus a reduced formation of solvent byproducts and less equipment corrosion.[98,101] Despite these merits, ultrasound technology has only been tested for CO_2 desorption by a few research groups so far. Ying et al. combined ultrasound technology with MEA solvents of different concentrations (30 and 70wt.%).[98] It was stated that a lower concentration of MEA resulted in higher CO_2 desorption rates as the diffusion of bubbles goes faster in a low viscosity solvent.[98] In another study of this group, the effects of operating variables – pressure, liquid flow rate, CO_2 loading, ultrasound intensity, frequency, and on-stream time - on CO_2 regeneration energy and efficiency were investigated. Among them, CO_2 loading was found to have a dominant influence on the reduction of regeneration energy consumption. In turn, the pressure was found to reduce the desorption rate and increase energy consumption. It was claimed that the ultrasound-assisted desorption reduced the regeneration energy by 35% compared to thermal heating, while the desorption rate was enhanced by a factor of 4.[99] In the latest study of Ying et al., ultrasound was applied in the reboiler section of a MEA-based CO_2 capture plant where the rich solvent is heated with steam to drive the desorption reactions releasing the CO_2 from the rich solvent. A cost estimation study showed 19% savings of operating costs for ultrasound-driven desorption technology compared to existing technologies.[100] In addition to ultrasound-assisted regeneration, megasonics-assisted (1MHz) regeneration was also tested. CO_2 desorption was performed at regeneration temperatures of 50 °C. The latter resulted in less solvent degradation. The solvent loss was only 3.15%, whereas it can rise to 11.65% for conventional technologies.[102] Liu et al. recently used ultrasound irradiation to regenerate the MDEA solvent in a microreactor system, referring to the method as an ultrasonic microreactor.[103] Adding ultrasound assistance enhanced the mass transfer performance at low temperatures and high values of microchannel diameters. The improved regeneration performance of the ultrasonic microreactor was ascribed to the higher bubble growth rate in the microreactor, which was caused by improved diffusion and increased bubble coalescence.[103] Reference is made to Figure 3 wherein the ultrasound technology performance is compared to that of other technologies. For a further detailed overview on ultrasound technology for desorption, the readers are referred to a recent review by Balraj et al.[97]

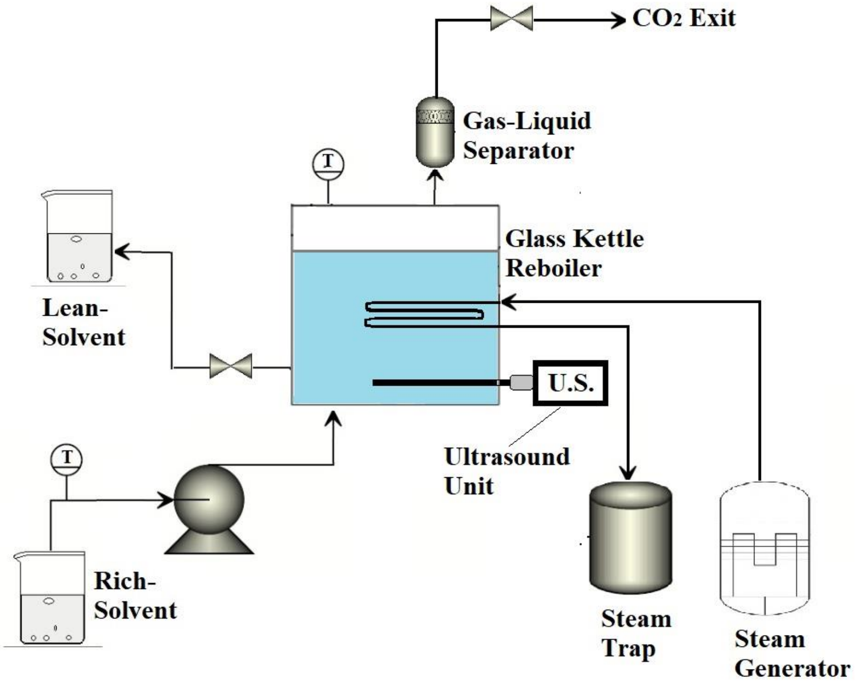


Figure 7. Schematic flow diagram for ultrasound assisted CO_2 desorption.

3.2.2 Microwave Heating

When the solvent to be regenerated is heated with microwaves, the required energy input is found to be larger than in the conventional packed bed technology for desorption (Figure 3).[104] However, regeneration energy for the microwave heating process was measured only for smaller volumes. This can significantly increase the energy duty, given the high surface-to-volume ratio of the unit.[104,105] Therefore, relative regeneration energies, that is the energy per mole of desorbed CO_2, are mostly reported. In microwave heating, molecules with a dipole moment absorb the microwave energy and convert it into heat. The energy transfer is thus more directly compared to thermal heating. Few studies however investigated the use of microwave irradiation for the CO_2 desorption process. Bougie et al. studied microwave irradiation for the desorption process using the MEA solvent (Figure 8).[104,106,107] CO_2 loading, viscosity, and heat capacity were found to be critical factors on the heating rate of the solvents. The relative energy consumption of a 50wt.% MEA solvent was found to be lower than a 30wt.% MEA solvent.[106] In a follow-up study, different non-aqueous solvents were tested for microwave regeneration.

For a 20wt.% MEA in diethylene glycol monoethyl ether (DEGMEE) solvent, the relative energy consumption reduced by 78% compared to a 30% MEA aqueous solvent. The low dielectric constant and high boiling point of the DEGMEE are on the basis of the low relative energy consumption. It was concluded that because of the low boiling point and high dielectric constant, non-aqueous solvents should be preferred over aqueous solvents when using microwave energy for heating and vaporization.[107] Li et al. regenerated CO_2-loaded tri-ethylene tetramine (TETA) and organic solvents by microwave irradiation.[108] The energy consumption was found to reduce by 70% for TETA in 1,4-butanediol (BDO) and TETA in poly(ethylene glycol) (PEG200) solvents compared to thermal regeneration of 30wt.% MEA in water solvent. Additionally, the CO_2 recovery increased by a factor 2.7 compared to the benchmark MEA solvent. For appropriate comparison, energy savings (ŋ) were calculated based on the microwave regeneration energy required to desorb CO_2 when using a 30wt.% MEA in water solvent as the benchmark (Equation 1). Note that the higher the energy saving values, the better the solvent for microwave regeneration.

$$\eta\ (\%) = \frac{E_b - E_{MW}}{E_b} \times 100 \qquad (1)$$

where E_b is the relative microwave regeneration energy of 30wt.% MEA in water, and E_{MW} is the relative microwave regeneration energy of non-aqueous solvents.

The additional evaluation of the microwave heating technology is thus made for aqueous and non-aqueous solvents when using similar microwave regeneration setups. Regeneration performances for different non-aqueous solvents based on the energy reduction are illustrated in Figure 9.[107,108] It can be noted that a higher relative energy-saving can be obtained for both MEA and TETA in organic solvents. While the use of microwave energy instead of thermal heating increased the energy consumption of aqueous MEA solvent (33.06%), energy consumption of TETA/BDO and TETA/PEG200 solvents decreased by 8.8% and 14.5%, respectively.[108]

Microwave heating technology could be a technology fit to tackle the high energy use/drawback of the CO_2 desorption process. Reference is made to Figure 3 where the microwave technology analyzed in light of existing technologies. As explained above the required regeneration energy value is high compared to the conventional reboiler regeneration technology. One of the main drawbacks is that significant efforts are needed to improve the scale-up of this technology.

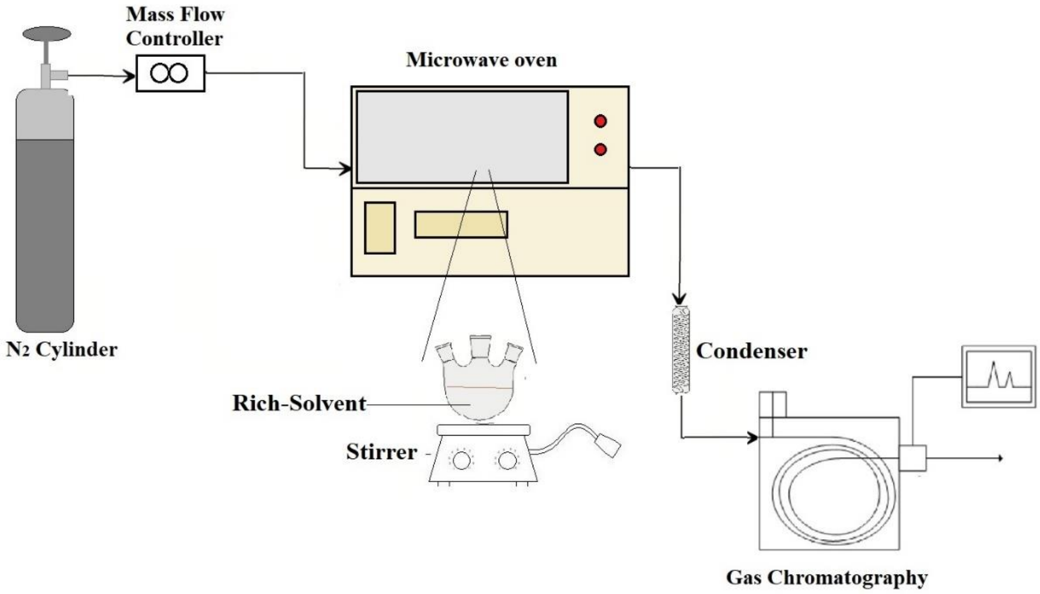


Figure 8. Schematic flow diagram for microwave assisted CO_2 desorption.

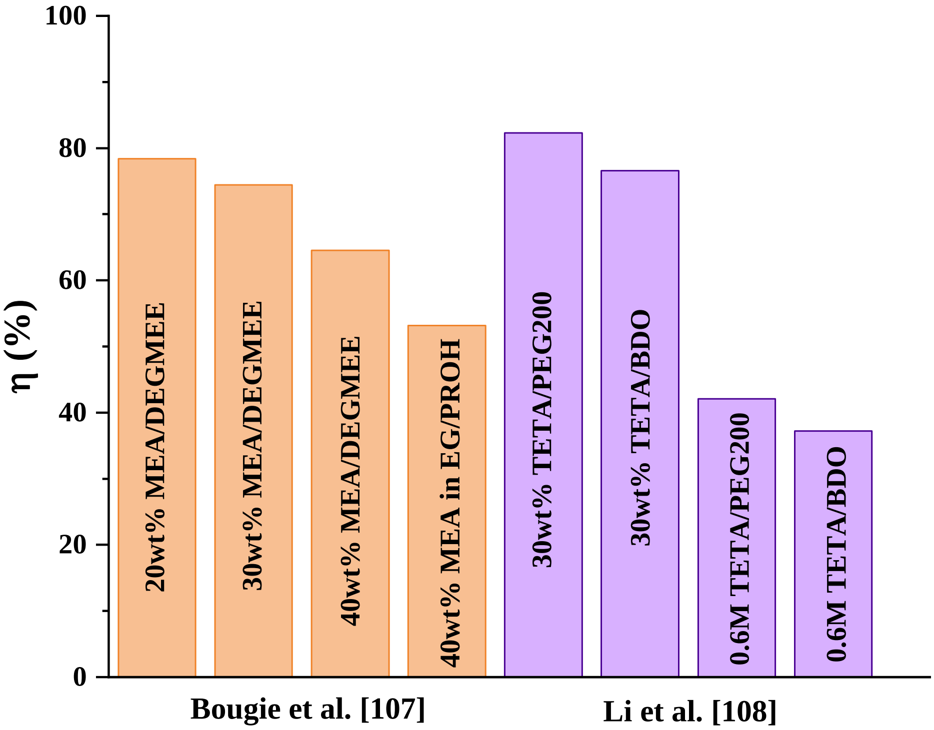


Figure 9. Relative energy savings for microwave regeneration of non-aqueous solvents compared to 30wt.%MEA.

3.3 Solvents Used for Intensified CO_2 Desorption

3.3.1 Intensified Packed Bed Technology

Intensified packed bed technology (i-PB) studies include studies discussing the use of different amine solvents, additional carriers replacing steam, and direct injection of steam in packed bed columns.

3.3.1.1 Novel Solvents/ Blended Solvents/ Non-aqueous Solvents

To reduce the regeneration energy and enhance regeneration efficiency the use of novel amine solvents and of alternative carriers other than steam in conventional packed bed technology has potential. Solvents such as MDEA, DEA, AMP, DETA and TETA have been extensively studied in the past. To decrease the reboiler heat duty, single and blended solvents of DEA, MDEA, and/or AMP have been benchmarked to MEA as the reference solvent by Sakwattanapong et al. Compared to the amine concentration, the heat duty was found to be inversely dependent on the lean and rich solvent loadings.[64] MEA has the highest heat of desorption and lowest operating CO_2 partial pressure, requiring the largest amount of steam to drive the CO_2 stripper. Thus, the required regeneration energy when using MEA is higher than compared to using DEA and MDEA. Moreover, several studies showed that regeneration of blended solvents like MEA-MDEA and DEA-MDEA requires less energy than when using a single solvent.[64] Also the effect rich solvent loading on the regeneration energy with solvent blends have been already studied.[75] As the rich solvent loading increases, the total regeneration energy consumption decreases. This was attributed to the higher equilibrium CO_2 partial pressure for rich solvent loading of 0.5 mol CO_2/mol MEA compared to rich solvent loading of 0.3 mol CO_2/mol MEA. Thus, the CO_2 stripping from high rich solvent loading leads higher driving force for transfer which needs less steam, implying that less energy is needed for water vaporization.[75]

The regeneration performance of the DETA solvent with higher absorption capacity and faster reaction kinetics was also investigated in a packed bed desorber.[74] It was reported that the regeneration energy compared to MEA was lower, but that the mass and heat transfer rates were reduced for higher concentrations (4 kmol/m^3) of DETA. The high concentrations result in increased solvent degradation and equipment corrosion.[74] As mentioned above, investigations on solvent development revealed that sensible heat and heat of vaporization have a large influence on the total regeneration energy. Since the blends of AMP-MDEA-DETA have lower sensible heats than a single MEA solvent, the regeneration energy required when using blends was also lower.[109] TETA was presented as a promising solvent for CO_2 capture. Rich TETA solvents with CO_2 loadings of 1.6 mol CO_2/mol TETA, which is significantly higher than the base case loading (0.5mol CO_2/mol MEA), results in a significant reduction of the required regeneration energy. However, longer contact times are needed to acquire high CO_2 loading of rich solvents from the bottom of the absorber columns. Thus, absorber columns with higher packing heights are needed, leading to a substantial increase in investment costs.[110]

Additionally, operating pressure of the desorber column is known to be effective on the reboiler heat duty. High operating pressures and reboiler temperature result in less water vapor, which means less heat is required from the reboiler. Thus, heat of vaporization becomes less significant because of the high pressures in the desorber column. This also results in less energy consumption for CO_2 compression.[111] To operate columns at high desorber pressures, solvents with high thermal stability are necessary. Piperazine, for instance, has a high absorption heat and can be operated at high desorber pressures. Rabensteiner et al. reported that increasing the operating pressure from 1.4bar to 2.6bar reduced the reboiler heat duty by 7.5% when using 37.6wt.% PZ.[112] Even though the energy reduction when using 30wt.% MEA was by 20.5%, the thermal degradation limitation of MEA at high temperatures and high pressures makes it unfavorable.[112]

3.3.1.2 Additional Carriers

As the heat of vaporization hugely contributes to the total heat duty, the use of a different carrier gas replacing steam can be beneficial. It was demonstrated that using pentane as carrier gas decreased the desorber outlet temperature and CO_2 partial pressure in the effluent gas stream.[73] Consequently, the use of pentane resulted in a reduction of the heat removal in the condenser following the desorber.[73] If water vaporization can be avoided in the reboilers substantial energy efficiency improvements could be feasible. Therefore, some studies focused on the development of non-aqueous desorption processes.[113,114] Injection of carrier gases such as nitrogen, methanol vapor and steam with a MEA/methanol solvent as absorbent, was evaluated.[113] The use of an inert carrier gas like nitrogen, as compared to the use of gaseous methanol and steam, resulted in irreversible reactions between CO_2 and the solvent. Among all carrier gases, injecting nitrogen resulted in the highest reduction of regeneration energy with a relatively low energy consumption of 2.38 MJ/kg CO_2. In a follow up study, the influence of a rich solvent flow rate, rich solvent loading, and regeneration temperature on the regeneration energy of MEA/methanol solvent was investigated in a pilot plant. Organic carriers such as pentane, hexane, and cyclohexane were also used to replace steam because they have a lower latent heat, boiling point, and heat capacity.[115] Among them, pentane was found to be the most promising alternative with a high regeneration rate and low regeneration energy consumption. The addition of an organic carrier increases the desorber pressure and eliminates the recompression energy for CO_2 storage downstream of the desorption column.[115] However, the utilization of an organic carrier also has its share of challenges such as its miscibility with the solvent, its more difficult removal of CO_2 downstream of the desorber, and its possible toxicity and flammability.[115]

3.3.1.3 Direct Steam Stripping

Direct Steam Stripping (DSS) refers to the direct injection of superheated steam generated in a low-pressure steam turbine. Unlike the Conventional Reboiler Regeneration (CRR) technology, the latent heat of the steam is more efficiently transferred to the solvent and the low-pressure steam generated by evaporation can be send back to the turbine.[58,116] A requirement is that the higher temperatures of the rich solvent and the carrier steam need to be maintained to prevent steam condensation during the desorption process. The regeneration energy consumption when using DSS, compared to the benchmark MEA solvent, was lowered by 23.2%. However, an economizer, a steam heat exchanger, and a gas compressor are required, resulting in an increase in the CAPEX.[58]

The recovery ratio of the carrier steam (χ) which is the amount of steam exiting the desorber column is significantly dependent on the temperature difference of the economizer and operating conditions.[42,58] Although recovery ratio is an important parameter for DSS, it is generally set constant as 0.8 when calculating steam energy. The influence of recovery ratio on regeneration, on the other hand, has only been studied using the Aspen Plus simulation software. It was found that increasing the recovery ratio up to 0.85 reduces the regeneration energy consumption. However, recovery ratios lower than 0.5 result in identical results to CRR in terms of regeneration energy consumption.[42]

3.3.1.4 Advanced Stripper Configurations

The reboiler heat duty can also be reduced by making modifications to the stripper column. These are cold-split bypass (CSB), interheater (IH) and lean vapor compression (LVC). They can be either applied separately or in combination with each other.[41,48,117] In the CSB configuration, the remaining cold-rich solvent was employed to extract the sensible heat from the hot lean solvent after a proportion of the cold-rich solvent was removed and injected into the top of the CO_2 stripper. IH involves extracting semi-lean solvent from the center of the column and heating it with the lean solvent from the bottom using an interheater. Then, it is transferred to the following stage. In both configurations, energy reduction relies on the steam condensation in the column. In the LVC configuration, a gaseous stream is produced by flashing hot lean solvent. It is compressed, then sent back to the column. The operating pressure with LVC is higher and the rich solvent temperature is lower than that of conventional desorber designs.[45] Liu et al. compared the energy saving performances of advanced stripper configurations with respect to the simple stripper design.[117] Advanced column designs were found to have lower top temperatures than simple strippers at the same operating pressures. Thus, less vapor was produced, which in turn reduced the reboiler heat duty. For CSB, IH and LVC configurations, energy saving values were 18%, 14% and 11%, respectively. When CSB was integrated with IH or LVC, energy savings were found to increase by 20-21%.[117]

3.3.2 Catalyst Aided Solvent Regeneration

Energy consumption during the regeneration process of a MEA is high due to the difficult proton transfer from the protonated amine to the neutral water and due to the highly endothermic desorption reactions, more specifically the deprotonation reaction of $MEAH^+$.[35] A catalyst can provide acid sites that promote the splitting of carbamate ions to release CO_2 at significantly lower temperatures (<100 °C) compared to conventional technologies, without altering the thermodynamics of the reaction.[118] Although the heat of desorption remains unaffected, the sensible heat and the latent heat of vaporization are reduced in catalytic solvent regeneration processes.[35]

Among several candidates, HZSM-5 and γ-Al_2O_3 are often employed as solid catalysts for MEA, blended MEA-MDEA, and blended MEA-DEA solvent regeneration at a temperature of 90-95 °C.[119] In fact, HZSM-5 was found to be the best performing catalyst when it comes to decreasing the regeneration energy and increasing the regeneration efficiency for the blended MEA-DEA solvent. The energy consumption was found to decrease from 3.5 to 1.56 MJ/kg CO_2 leading to a significantly reduced water vaporization heat.[119] As a consequence, HZSM-5 and γ-Al_2O_3 became the commonly used catalysts for follow-up studies on catalyst aided CO_2 solvent regeneration.[18,35,120,121] A study using different catalysts claimed that the energetic performance of the H-Y catalyst was worse than that of the HZSM-5 and γ-Al_2O_3 catalysts due to its lower surface area.[35] It is noted that the higher surface area and Brönsted/Lewis ratio of a catalyst result in a higher desorption rate and hence lower regeneration energy.[35] In addition to these commonly used industrial catalysts, SAPO-34 and SO_4^{2-}/TiO_2 catalysts were also evaluated for regeneration of MEA and a tri-blended solvent containing MEA, AMP and PZ.[18,122] Investigation of CO_2 desorption of the same tri-blend solvent showed that, in the presence of solid acid catalysts like H-mordenite, Hβ, HZSM-5, and Al_2O_3, the energy requirement reduced by 66.1% when compared to the desorption of a MEA without catalyst.[118] Osei et al. studied the effect of adding a solid acid catalyst on the liquid-side mass transfer performance for the desorption of CO_2 from a CO_2-loaded MEA solvent in a packed bed column.[123] An improved performance was observed in the presence of HZSM-5. This was attributed to the ability of that catalyst to act as a proton donor and thus facilitate the $MEACOO^-$ breakdown.[123] Coker et al. experimentally studied the mass transfer performance of catalyst-aided CO_2 desorption.[124] Experiments were performed in a full-cycle bench scale pilot plant to both improve the CO_2 absorption using MEA, MEA-MDEA, and BEA-AMP as blended solvents and to facilitate the desorption in the presence of HZSM-5 and K/MgO. The overall liquid side mass transfer coefficient for the blended BEA-AMP solvent was found to increase by 23.6% in the presence of HZSM-5 catalyst and by 45% in the presence of both K/MgO and HZSM-5 catalysts.[124]

Even though an extended number of catalysts that could be used for catalyst-aided solvent regeneration have been studied, challenges still exist when it comes to actual industrial application. This must be attributed to the bulk of the studies on the catalyst-aided desorption was performed employing a simple recirculation technology without heat insulators to prevent heat losses (Figure 10).[120,125] The latter explains the high values for energy consumption reported in literature for these catalyst aided solutions compared to other technologies (see Figure 3).

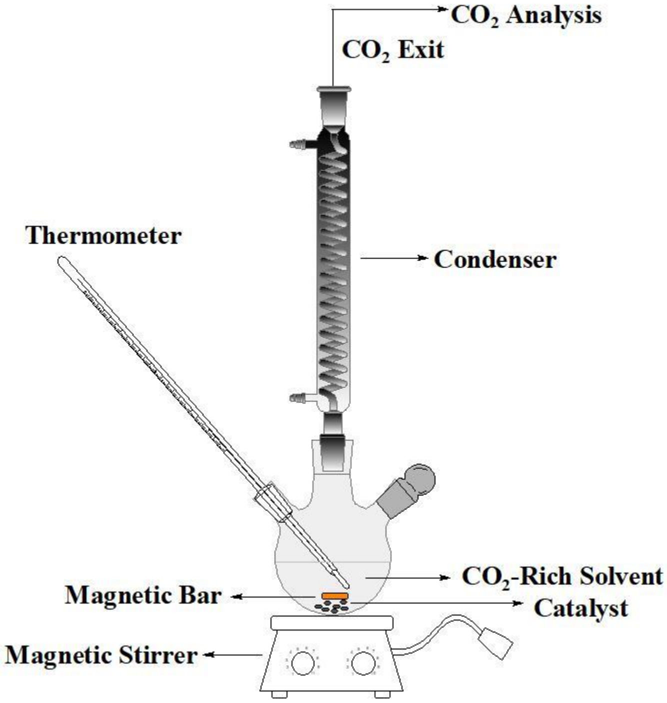


Figure 10. Experimental apparatus for catalyst-aided CO_2 desorption.

To evaluate the performance of various catalysts for catalyst-aided regeneration processes, the energy saving (ŋ) of each of them based on the equipment that was used was estimated using Equation 2. The higher the energy saving values, the better the catalyst aided solvent for CO_2 regeneration.

$$ŋ\ (\%) = \frac{E_b - E_{CA}}{E_b} \times 100 \tag{2}$$

where E_b is the regeneration energy needed for the solvent without catalyst and, E_{CA} is the regeneration energy needed for the solvent in the presence of a catalyst.

As mentioned above, high regeneration energy consumption values are reported for a catalyst-aided regeneration technology (Figure 3) due to the use of simplified recirculators where heat losses were not prevented. Figure 11 evaluates catalyst-aided regeneration studies based on the desorption equipment used. The analysis learns that for MEA or a blended MEA-DEA solvent, also in this case HZSM-5 was found to be the most effective catalyst (Figure 11). On the contrary, for tri-blended MEA-AMP-PZ solvent Hß zeolite should be preferred. Nonetheless, irrespective of the catalysts used, the energy savings compared to solvents without catalyst, are seen to decrease going from single solvents to tri-blended solvents.

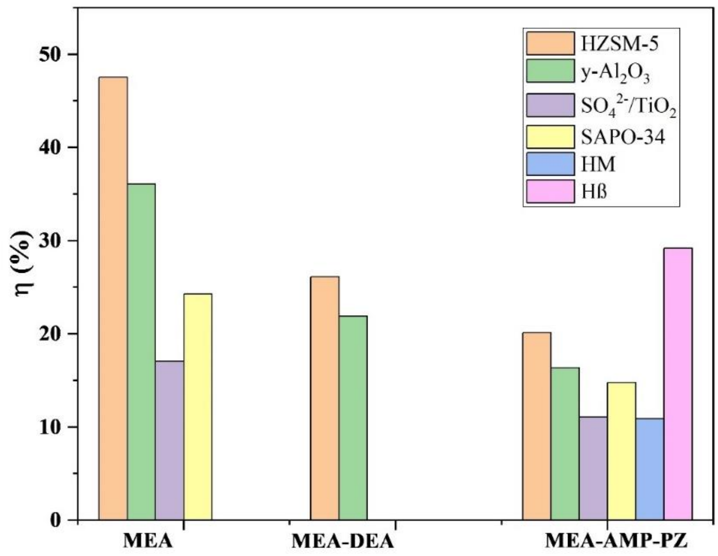


Figure 11. Energy savings for catalyst aided solvent regeneration compared to conventional solvent regeneration.

Bairq et al. tested the effect of various modified heterogeneous mesoporous catalysts on CO_2 desorption using MEA solvent at 97 °C. They found that the CMK-3-SiO_2 catalyst enhanced the desorption rate by 195% and decreased the energy requirement by 37.41% compared to blank MEA.[126] The enhanced performance due to the presence of the catalyst was attributed to the availability of a large mesoporous surface and an abundance of Brönsted and Lewis acid sites.[126] In yet another study it was found that, at 95 °C, the desorption rate increased by 28.9% when regenerating a CO_2-loaded rich solvent using a $TiO(OH)_2$ catalyst compared to the non-catalytic regeneration of that solvent.[127] It was confirmed that the strong acidity of the catalyst is responsible for these improvements. Similarly, Bhatti et al. studied the performance of CO_2 desorption for MEA and DEA solvents using two metal oxide catalysts (TiO_2 and V_2O_5) for a temperature range of 40 to 86 °C.[128] Both catalysts were found to improve the solvent regeneration performance, mainly because higher desorption rates were realized at lower regeneration temperatures. The desorption rate showed a marked increase by 86% and 50% for MEA and DEA solvents, respectively. The amount of the desorbed CO_2 increased by 17% and

13%, respectively.[128] In another study of the same group, the desorption rate for a MEA solvent was reported to increase up to 54% when employing ZrO_2 and ZnO catalysts.[125] Lai et al. found that a nanostructured $TiO(OH)_2$ catalyst drastically increased the CO_2 desorption rate from spent MEA by over 4500% (at 88 °C).[129]

3.3.3 Desorption Kinetics

Kinetic studies have been carried out in stirred cells, single sphere units, wetted columns, and laminar jets because interfacial areas are predictable, and hydrodynamics are known.[130] There is a vast number of studies concentrating on the absorption kinetics of CO_2 in amine solvents, however, the number of studies on CO_2 desorption kinetics is limited.[131] Nevertheless, a comprehensive understanding of desorption kinetics is of major importance when selecting a proper solvent for a specific application.[132] Desorption rate is an important aspect in desorption kinetics.[133,134] In studies prior to the work of Jamal et al., forward (absorption) and backward (desorption) kinetic rate parameters were assumed to be related.[135] Desorption rate parameters were calculated at conditions identical to those used to determine absorption rate parameters.[136] These first studies assumed CO_2 desorption kinetics to be pseudo-first order in an aqueous MDEA solvent.[137,138] However, in a study by Jamal et al., it was shown that only forward rate constants could be determined based on absorption experiments. Desorption experiments are needed to determine backward kinetic rate constants.[135] Reactions and rate expressions for CO_2 desorption in aqueous amine solvents were provided by Jamal et al.[130] The desorption reaction mechanism for carbamate amine forming solvents, including MEA, DEA, and AMP solvents, was described using the zwitterion mechanism.[130,135] For the MDEA solvent, however, the proposed desorption reaction mechanism is a base-catalyzed hydration reaction mechanism. This difference in kinetic mechanism is due to the absence of carbamate formation in the MDEA solvent to be regenerated.[130,135] The reaction pathway of MDEA is simpler because bicarbonate and protonated MDEA are formed by CO_2 hydrolysis. When using MDEA as the solvent, the desorption rate is lower which makes it the preferred solvent to study desorption kinetics.[139] Due to the lower number of hydrogen atoms, the chemical activity of MDEA is lower than that of primary or secondary amines. However, desorption rates are higher as the carbamates formed have a higher metastability.[133] Kierskowka-Pawlak et al. developed a simplified pseudo-first-order model for these reversible absorption-desorption reactions. The predictions of using the proposed model for a MDEA solvent were found to be in good agreement with the experimental desorption rate data.[131] A chemical enhancement factor for the desorption kinetics was introduced by Hamborg et al. investigating the effect of reaction kinetics on mass transfer using MDEA as solvent.[139] When the enhancement factor for the absorption kinetics was compared with the enhancement factor for the desorption kinetics, they were found to be very similar under similar operating conditions.[139] Tunnat et al. studied the desorption rates using MEA and DEA solvents. DEA gave a higher desorption performance when compared to MEA. This was attributed to the carbamate instability when using DEA as solvent.[140] A kinetic study for a novel blended solvent including butyl (amino)-ethanolamine (BEA) and AMP was performed by Afari et al.[30] The kinetic results for the BEA-AMP blend were compared with those for MEA and the MEA-MDEA solvent blends.[30] Because of the steric hindrance for AMP and the long BEA molecules, the carbamate instability increases. Correspondingly the desorption rate for the BEA-AMP blend was the highest. When using a MEA solvent, the hydrogen in the alkyl group makes the molecule very stable. CO_2 bonds with MEA are hard to break, which in turn results in slower desorption rates. Finally, it was observed that the addition of the tertiary amine MDEA to MEA, forming a blend, increases the desorption rate of the unblended MEA solvent due to the formation of metastable bicarbonate ions which promotes to release CO_2.[30]

Recently, Sunjhi et al. proposed to use thermogravimetric analysis (TGA) with model-free and model-fitting methods to determine desorption kinetic parameters for MDEA and MDEA-DEA solvents.[132] Observing identical TGA trends for both solvents at four different heating rates confirmed the repeatability of the method. Based on weight loss analyses, it was stated that at first CO_2 and H_2O were desorbed. When the regeneration temperature increases, MDEA or DEA partially evaporated. It was also noted that the desorption efficiency of MDEA was higher than that of the blended solvent while the absorption rate of the blended solvent on the contrary was higher than that of MDEA. This is due to the fact that the absorption activation energy of MDEA was lower than that of the blended MDEA-DEA solvent.[132] The effect on both absorption and desorption kinetics when blending the MDEA solvent with PZ and DEA activators was studied by Shunji et al. using the TGA method.[133] Although the activators increased the absorption rate of MDEA, the desorption rates were found to decrease compared to the unblended solvent. These results correspond with the fact that the desorption process is the reverse of the absorption process.

Table 1. Overview of PI Technologies for CO_2 desorption

Technologies	**Description**	**Improvements**	**Drawbacks**	**Ref.**
Rotating Packed Bed Technology	• Rotating doughnut-shaped device including the packing material. • Centrifugal field is produced by the high-speed rotor. • Gas-phase fed through the outer edge, flowing radially inwards passing through RPB to exit. • Liquid phase entering through nozzle radially outward.	• Control of residence time and thickness of mass transfer film. • Higher flooding velocities. • Improved mass transfer coefficients due to enhanced acceleration. • Increased gas-liquid contact area due to small droplets and liquid films. • Smaller volume.	• Short contact time. • Use of solvents with high amine content solvents. • Scalability.[48]	4,5,76,77
Microreactor Technology	• CO_2-rich solvent pumped into microreactor. • Due to desorption temperature, two phases (liquid and gas) are formed.	• High interfacial area. • Increased mass and heat transfer. • Thermal loss reduction. • Ease of operation and process control. • Reduced cost due to reduced volume.	• Low capacity. • Scalability. • Difficult hydrodynamics and transfer characteristics.	6,61,71, 83,84

	• A phase separator is installed at the microreactor’s outlet. • Gas and liquid phases are released from the upper and bottom parts of the separator, respectively.			
Membrane Stripping Technology	• CO_2 gas molecules permeate through pores of the membrane. • Membrane acts like a barrier controlling differential pressure between phases. • Sweeping gas used to strip CO_2 molecules.	• High interfacial area. • Nondispersive gas-liquid contact. • Improved operational flexibility. • Reduced CAPEX and OPEX. • Vacuum technology with low regeneration temperature. • Can be operated in any orientation.	• Reduced mass transfer and membrane efficiency due to pore fouling and wetting. • Cost increase due to increased length needed for higher contact area. • Low partial pressure reduces the permeability of CO_2. • Additional energy cost for the vacuum generation and compression of CO_2.	3,20,91-93

Ultrasound-Assisted Regeneration	• Inaudible ultrasonic waves, frequency 20-100kHz. • Micro-liquid jets and vortex produced by ultrasound. • Increased interfacial area due to millions of cavitation bubbles.	• Lower CO_2 loadings in a lean solvent. • Lower regeneration energy consumption. • Intensified liquid phase mass transfer. • Reduced regeneration temperature resulting in limited degradation. • Improved desorption capacity. • Reduced desorption time.	• High energy input needed. • Lack of literature studies.	98,99,100,141
Microwave Heating Regeneration	• Molecules having dipole moment absorb microwave energy and convert it to heat. • Direct molecular interactions with electromagnetic radiations.	• Faster heating and cooling. • Lower solvent regeneration temperature. • Reduced loss of solvent. • Instantaneous and volumetric heating.	• Ineffective for regeneration of aqueous MEA solvents. • Poor regeneration efficiency value due to low boiling point and high dielectric constant of solvents.	104,106-108
Intensified Packed Bed Technology	• Novel solvents like MDEA, DEA, DETA and TETA are used. • Non-aqueous solvents are used.	• Reduced regeneration energy with novel solvents. • Low regeneration temperature of non-aqueous solvents.	• Increased cost due to high-cost solvents. • Degradation and corrosion.	58,64, 73-75, 109,110, 113-116

	• Additional carriers like nitrogen, pentane and hexane are used. • Steam can be injected directly.	• Additional driving force with additional carriers.	• Toxicity and inflammability of non-aqueous solvents. • Increased CAPEX of DSS.	
Catalyst-Aided Solvent Regeneration	• Catalysts with acid sites promote the splitting of carbamate to release CO_2 at lower temperatures. • Introduction of metal ions catalysts in an amine solvent. • Chemisorption of carbamate on the catalyst surface. • With the proton donating catalyst, deprotonation step.	• Catalytic desorption results in faster kinetics. • Enhanced CO_2 desorption rate and more desorbed CO_2. • Reduced regeneration energy. • Lower regeneration temperature resulting in less solvent degradation.	• Catalyst powders decrease catalyst-solvent contact area reducing the catalyst activity. • Replacing conventional packing by catalyst needs attention. • New desorber design needed to prevent blockage in packing section.	18,35, 118,121, 126,142, 143

Table 2. Geometry and operating conditions for process intensifying equipment and methods

PI Method	Desorber Type	Operating Conditions	Rich Solvent Properties	Ref.
Rotating Packed Bed Technology	r_i (m)=0.025-0.078 r_o (m)=0.080-0199 h (m)=0.020-0.025 SS wire mesh	T_{des}=67-105 °C P_{des}=1-2.5 atm	30wt.% MEA, 10/20wt.% PZ-DETA, 3.6M AMP-AEEA-NMP	4,5,76,77
Microreactor Technology	Tubular geometry 0.001x0.0006 m, L(m)= 0.10 Channel geometry r_i (m)=0.0004, L (m)=0.035	T_{des}=55-95 °C P_{des}=1 atm	30wt.% MEA, 30wt.% DEA, 30wt.% MDEA	61,71,83
Membrane Stripping Technology	Aluminum Oxide, PP, hollow fiber PVDF	T_{des}=70-80 °C P_{des}= 0.2-0.5 atm	2M-5M DEA, 20wt.% MEA, 6wt.% SMM	3,92,93,144
Ultrasound-Assisted Regeneration	Ultrasonic tube reactor, semi-batch unit	T_{des}=40-60 °C, 105-125 °C Power: 50 W,	30wt.%-70wt.% MEA, 30wt.% MEA: MDEA (1:1)	98-100,141

		f: 20-28 kHz		
Microwave Heating Regeneration	Cylindrical quartz reactor, three-necked flask	MW=100-480 W T_{des}=70-90 °C	30wt.%-50wt.% MEA, MEA in DEGMEE TETA/BDO and TETA/PEG200	104,106-108
Intensified Packed Bed Technology	Packed bed column	T_{des}=70-130 °C P_{des}=1-2 atm	MDEA, DEA, DETA, TETA, AMP-MDEA-DETA	58,64,73-75, 109,110,113-116
Catalyst Aided Solvent Regeneration	Batch reactor Packed bed column Bubble column	T_{des}=60-105 °C T_{des}=85 °C T_{des}=80 °C	MEA, MEA-DEA, MEA-AMP-PZ with HZSM-5, y-Al_2O_3, SO_4^{2-}/TiO_2, SAPO-34; HM, and Hß MEA with HZSM-5 and y-Al_2O_3 Cu-MEA	18,35,118, 120,121,143

4. Performance Metrics

An overview of CO_2 desorption studies including both conventional and intensified technologies was presented above. Different process intensification methods, as previously stated, seek to enhance the mass transfer characteristics of the conventional technology. Thus, an overview of mass transfer coefficients for most of the discussed PI technologies provides a better understanding of their performances. As the mass transfer in the liquid phase has the largest influence on the overall mass transfer, gas side mass transfer resistance was mostly neglected. It results in equal volumetric overall liquid and liquid-only mass transfer coefficients, $K_La_V \cong k_La_V$.[63,83] Volumetric overall liquid (K_La_V) mass transfer coefficients for conventional and intensified technologies are compared in Figure 12a. K_La_V values in RPBs are higher than in most conventional packed beds but remain lower than the values in other PI equipment. The mass transfer coefficients in microreactor studies are found to be two orders of magnitude higher than in conventional packed bed technologies.[61,71,83] Due to higher liquid recirculation rates and smaller diffusional resistance at the gas-liquid interface, K_La_V values in microreactors are improved by increasing the solvent flow rate and the temperature.[71] The volumetric overall mass transfer coefficients for membrane stripping technology are one order of magnitude higher than for the packed bed technology due to their higher interfacial area. Volumetric overall mass transfer coefficients for catalyst aided technologies are comparable to those for the conventional packed bed technology. Mass transfer performance studies still need to be performed for ultrasound-assisted and microwave heating regeneration. Overall, for the desorption process, the number of mass transfer studies for the different PI technologies remains limited.

The volumetric flow rates of CO_2 stripped from the rich solvent for conventional and intensified desorption technologies based on lab-scale experimental data found in literature are shown in Figure 12b. Despite the fact that microreactor and membrane stripping technologies have considerably larger volumetric overall mass transfer coefficients than that of conventional and other intensified technologies, the volumetric flow rates of CO_2 stripped from a rich solvent are rather low due to the small desorption volumes. Rotating packed bed technology, on the other hand, provides the highest volumetric flow rate of stripped CO_2 of all technologies due to its high mass and heat transfer rates. The relative volumetric flow rates of the CO_2 stripped from a rich solvent for the different technologies is an important parameter to compare these intensified technologies as it indicates the capacity and scalability of the desorption units for industrial applications.

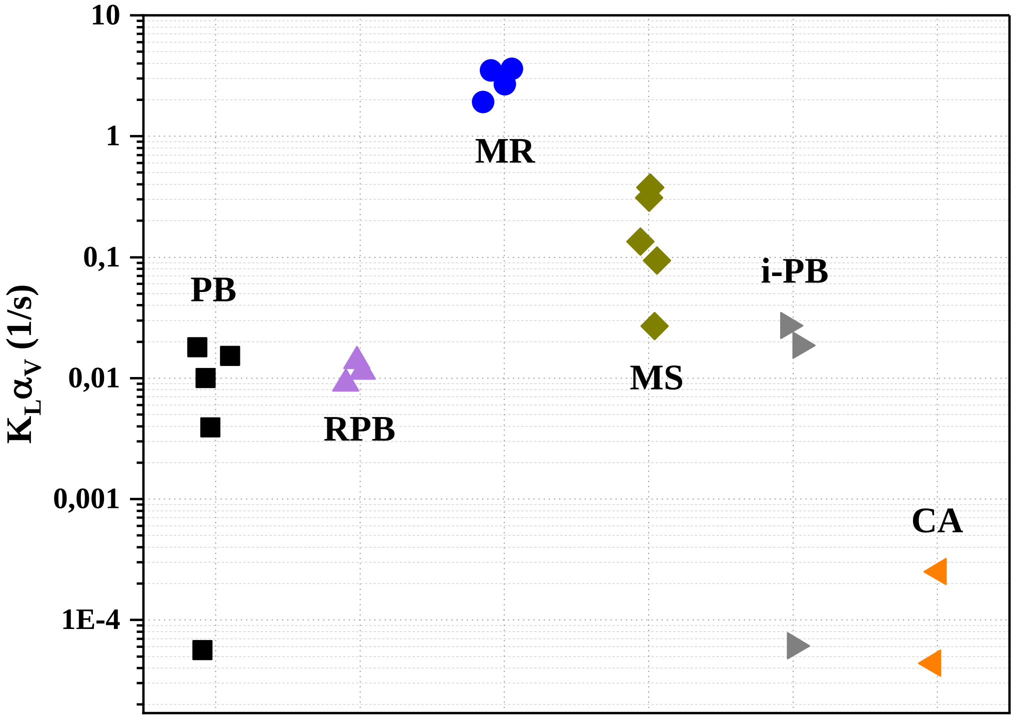
Enhancing mass transfer characteristics with different process intensification methods can improve desorption performance in terms of both regeneration efficiency and regeneration energy. We now introduce a single parameter to compare the different CO_2 desorption technologies which includes both regeneration efficiency and regeneration energy. Given the different geometries of the intensified CO_2 desorption equipment, the effect of the desorber volume on the regeneration performance is included as well. This parameter will allow to make a proper comparison of conventional and intensified technologies. A variable named *Regeneration factor* including the energy, efficiency, and volume is proposed in this section. CO_2 desorption technology is considered to be efficient when more CO_2 is desorbed, but at the same time the regeneration energy needs to remain limited and the volume of the desorber should be small. Based on these variables, the cost reduction of different technologies can also be analyzed. The regeneration factor is calculated as:

$$Regeneration\ factor\ (RF) \ = \ \frac{Regeneration\ Efficiency}{Regeneration\ Energy\ * Volume\ of\ the\ Desorber} \tag{3}$$

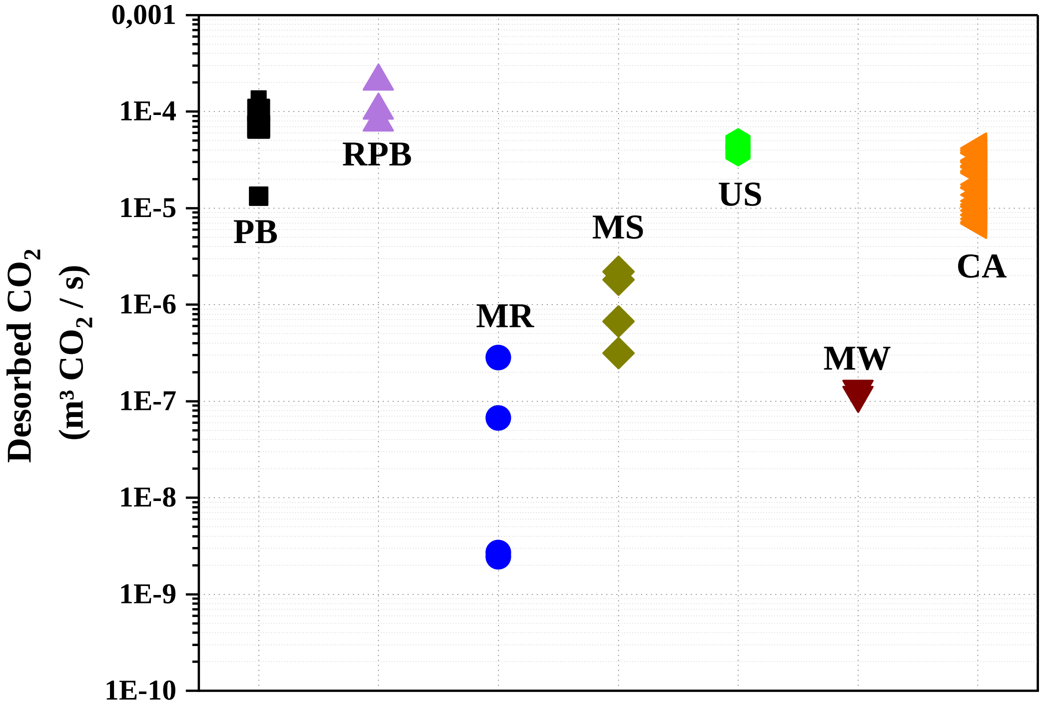
In Figure 12c, the regeneration factor for conventional and intensified technologies is compared. For Rotating Packed Beds, RF values are found to slightly improve compared to those of the conventional packed bed technology. The lower RF values of the RPB technology are due to low regeneration efficiency, which must be attributed to the short contact times in an

RPB. Thus, although the RPB regenerator consumes less energy and has a smaller volume than packed bed columns, the regeneration efficiency still needs improvement. Microreactors achieve the highest RF values of all technologies, conventional and intensified. The excellent mass and heat transfer characteristics of microreactors result in low regeneration energy consumption. The microreactor technology is thus very promising, also in terms of investment cost due to the small volume of a unit. Membrane stripping processes have high RF values, due to a low energy requirement and a small desorber volume. When alternative energy sources are used, microwave technology is found to improve regeneration performance compared to conventional packed bed technology. Ultrasound assistance was reported as a very promising technology to increase the CO_2 desorption performance.[100] However, the RF value is low. But the value is based on one study only. Further data is needed for a fair comparison. Intensified packed bed technologies have lower regeneration energy requirements and higher regeneration efficiencies than conventional packed bed technologies, when making the use of novel or blended solvents and of alternative carriers that are considered promising for CO_2 desorption. In catalyst-aided regeneration, high regeneration energy consumption due to high heat losses are the main reason for the low RF values. In addition, replacing the conventional packing by a bed of catalyst particles implies additional attention for possible blockage problems in the packing section. From Figure 12c it is concluded that microreactor and membrane stripping technologies are the most promising PI technologies for CO_2 desorption today as they have the high RF values. However, the drawbacks of these technologies for CO_2 desorption need to be considered as well. The use of microreactor technology in industrial plants is challenging since the volume of CO_2-containing flows, like flue gas flows, are very high, which makes scalability challenging. Membrane contactors in turn are prone to pore fouling and wetting, resulting in reduced mass transfer characteristics, an important limitation for applications on an industrial scale. Note that the RPB technology has higher desorbed CO_2 volumetric flow rates and higher

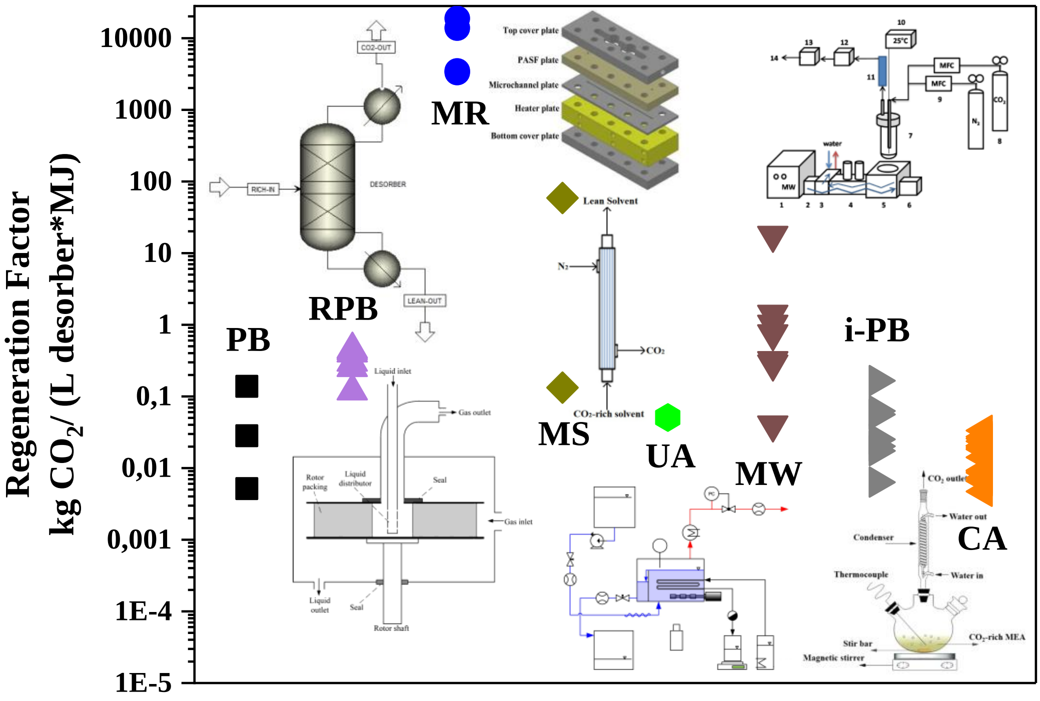
RF values than conventional packed bed technology. Additionally, the progress made for the design and optimization of the RPB technology for CO_2 desorption that is already made now makes it also a promising technology for industrial application.



(a)


Desorbed CO_2
(m^3 CO_2 / s)
0,001
1E-4
1E-5
1E-6
1E-7
1E-8
1E-9
1E-10
PB
RPB
MR
MS
US
MW
CA

(b)


Regeneration Factor
kg CO_2/ (L desorber*MJ)
10000
1000
100
10
1
0,1
0,01
0,001
1E-4
1E-5
PB
RPB
MR
MS
UA
MW
i-PB
CA
CO2-OUT
RICH-IN
DESORBER
LEAN-OUT
Liquid inlet
Gas outlet
Rotor packing
Liquid distributor
Seal
Gas inlet
Liquid outlet
Seal
Rotor shaft
Top cover plate
PASF plate
Microchannel plate
Heater plate
Bottom cover plate
Lean Solvent
N_2
CO_2
CO_2-rich solvent
water
MW
MFC
MFC
25°C
N_2
CO_2
CO_2 outlet
Water out
Condenser
Water in
Thermocouple
Stir bar
Magnetic stirrer
CO_2-rich MEA

(c)

Figure 12. Comparison of process intensification technologies using a) mass transfer coefficients b) desorbed CO_2 volumetric flow rates c) regeneration factor.

PB=Packed Bed, i-PB=intensified Packed Bed, RPB=Rotating Packed Bed, MW=Microwave Heating Regeneration, US=Ultrasound-Assisted Regeneration, MR=Microreactor, MS=Membrane Stripping, CA=Catalyst-Aided Solvent Regeneration.

5. Conclusions

The high capital and operating costs of the amine-based CO_2 capture process, especially the huge energy penalty of the solvent regeneration step, are major concerns for large scale industrial application. Despite substantial research into the CO_2 absorption step, research in and knowledge of the CO_2 desorption step is still limited. This study provides a comprehensive overview of the current state of the art of different technologies that can be used for CO_2 desorption, including conventional packed bed technology and high-performance equipment such as rotating packed beds, microreactors, membrane contactors, ultrasound assisted, and microwave assisted technologies. The performance of these technologies is evaluated based on the required solvent regeneration energy and the obtained regeneration efficiency. A new parameter, named *Regeneration Factor (RF)*, is introduced. RF accounts for the regeneration energy and for the desorption efficiency, but also for the volume of the desorber equipment, to establish a proper comparison of all technologies in terms of cost-efficiency. Based on their RF value, microreactors and membrane contactors are found to be the most promising technologies. This can be attributed to the high mass transfer coefficients, respectively, two orders and one order of magnitude higher than the conventional packed bed mass transfer coefficient. However, the proposed RF value does not take into account for the scalability of the desorption equipment, which is the main drawback of the microreactor and membrane stripping technologies. From the microwave-assisted desorption technology studies it is concluded that the relatively high RF values are a consequence of high regeneration efficiencies and small equipment volumes. It is found that the use of novel and blended solvents, or the use of carriers reduces regeneration energy consumption, but that overall regeneration efficiency needs further improvement. Rotating packed beds with enhanced mass and heat transfer perform slightly better than conventional packed beds. However, the fast improvement in design and optimization of this technology, compared to the other intensification technologies, makes it the more promising technology for CO_2 desorption on industrial scale.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Nomenclature

a_V	interfacial area per volume (m^2/m^3)
C_A	concentration of amine (kmol m^{-3})
$C_{L,i}$	concentration of solute in the inlet liquid (mol L^{-1})
$C_{L,o}$	concentration of solute in the outlet liquid (mol L^{-1})
d_i	inside diameter of membrane (m)
d_{ln}	logarithmic mean diameter of membrane (m)
d_o	outside diameter of membrane (m)
E	enhancement factor (dimensionless)
h	packed height of column (m)
H	Henry's constant (kPa dm^3 mol^{-1} or dimensionless)
k_G	gas-phase mass transfer coefficient (m s^{-1})
k_L	liquid only mass transfer coefficient (m s^{-1})
k_La_V	volumetric liquid only mass transfer coefficient (s^{-1})
K_L	overall liquid-phase mass transfer coefficient (m s^{-1})
K_La_V	volumetric overall liquid mass transfer coefficient (s^{-1})
k_M	membrane mass transfer coefficient (m s^{-1})
L_m	molar liquid rate (kmol $m^{-2}s^{-1}$)
P_{des}	desorption pressure (atm)
r_i	inner radius (m)
r_o	outer radius (m)
Q_L	volumetric flow rate of liquid (m^3 s^{-1})
S	stripping factor
T_{des}	desorption temperature (°C)
T_{reb}	reboiler temperature (K)

V	the volume of the microchannel (m^3)
wt.%	weight percentage
x	mole fraction of CO_2 in the liquid phase
x_e	mole fraction of CO_2 in the liquid in equilibrium with bulk gas
z	axial length of packing (m)

Greek letters

α	CO_2 loading of the solution, mol CO_2/mol amine
ŋ	energy saving
f	frequency
ρ_m	molar density (kmol m^{-3})

Abbreviations

AEEA	2-(2-aminoethylamino) ethanol
AMP	2-amino-2-methyl-1-propanol
BEA	butyl (amino)-ethanolamine
BDO	1,4-butanediol
CA	catalyst-aided solvent regeneration
CAPEX	capital expenditure
CCS	carbon dioxide capture and storage
CCU	carbon dioxide capture and utilization
CSB	cold-split bypass
CRR	conventional reboiler regeneration
DA2MP	1,5-diamino-2-methylpentane
DEA	diethanolamine
DEGMEE	diethylene glycol monoethyl ether
DETA	diethylenetriamine

DSS	direct steam stripping
HIGEE	high gravity
IH	interheater
LEAN	lean solvent
LHV	lean vapor compression
MDEA	methyldiethanolamine
MEA	monoethanolamine
MR	microreactor
MS	membrane Stripping
MW	microwave Heating Regeneration
NMP	N-methyl pyrrolidone
OPEX	operational expenditure
PB	packed bed
i-PB	intensified packed bed
PCC	post-combustion capture
PEG200	poly(ethylene glycol)
PI	process intensification
PG	potassium glycinate
PP	polypropylene
PTFE	polytetrafluoroethylene
PVDF	polyvinylidene fluoride
PZ	piperazine
RICH	rich solvent
RF	regeneration factor
RPB	rotating packed bed

SMM	surface modifying macromolecule
SS	stainless steel
TETA	triethylenetetramine
TGA	thermal gravimetric analysis
US	ultrasound-assisted regeneration

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Supporting Information

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