

Review

Emerging Solvent Regeneration Technologies for CO₂ Capture through Offshore Natural Gas Purification Processes

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Abstract: It is estimated that 40% of natural gas reservoirs in the world are contaminated with acid gases (such as hydrogen sulfide and carbon dioxide), which hinder exploitation activities. The demand for natural gas will increase by 30% from 2020 to 2050, with the rise of industrial activities and the lifting of travel restrictions. The long-term production of these high acid-gas fields requires mitigation plans, which include carbon capture, utilization, and a storage process to reduce carbon emissions. Absorption is one of the most established technologies for CO₂ capture, yet it suffers from extensive energy regeneration and footprint requirements in offshore operations. Therefore, the aims of this paper are to review and analyze the recent developments in conventional and emerging solvent regeneration technologies, which include a conventional packed-bed column, membrane contactor, microwave heating, flash drum, rotating packed bed, and ultrasonic irradiation process. The conventional packed column and flash drum are less complex, with minimum maintenance requirements, but suffer from a large footprint. Even though the rotating packed-bed column and microwave heating demonstrate a higher solvent flexibility and process stability, both technologies require regular maintenance and high regeneration energy. Membrane contactor and ultrasonic irradiation absorption systems are compact, but restricted by various operational issues.

Keywords: carbon dioxide; absorption; regeneration; CO₂ removal; gas separation process



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1. Introduction

The issues of global warming and climate change have become a subject of intense interest all over the world since the last decade. The warming of the climate system is now evidenced from observations of increasing global average air and ocean temperatures, the widespread melting of snow and ice, and the rising global average sea level. This scenario will undoubtedly give rise to the destruction of the ecosystem, biodiversity, and human economic activities, both in the short and long terms [1]. The anthropogenic (human-caused) driver of climate change is the increasing concentration of greenhouse gases (GHGs) in the atmosphere [2]. Carbon dioxide (CO₂) is the largest contributor to GHGs with regard to the amount present in the atmosphere, where it contributes to 65% of global warming effects, primarily as a result of burning coal, oil, and natural gas [3]. Despite the presence of other GHGs, CO₂ receives the most attention due to its long lifespan in the atmosphere, which makes it the most critical gas when tackling the issues of climate change. Archer et al. reported that the time needed for 70%, 20%, and 10% of emitted CO₂ to remain in the atmosphere is 100 years, 1000 years, and 10,000 years, respectively [4]. The International Panel of Climate Change (IPCC) predicted that the atmospheric content of CO₂ can reach up to 570 ppmv, leading to an increment in the mean sea level by 3.8 cm and a rise in the global mean temperature of about 2 °C by the year 2100 [5]. If the phenomenon remains unchanged, the devastating effects on the world's population will be unavoidable. The total CO₂ emissions gradually increased from 1990 to 2002, and drastically increased after

2019 due to rapid industrialization as reported by International Energy Agency (IEA) [6]. The CO₂ emission trend showed a notable decline of 9.7%, shifting from 33.4 GtCO₂ in 2019 to 31.5 GtCO₂ at the end of 2020. The observed trend was due to the reduction in CO₂ emissions in power generation, ground transportation, and the industrial sector, as lockdowns were implemented to curb the rapid spread of COVID-19. However, the CO₂ emission savings as a result of the drastic lockdown were only temporary and showed no significant impact on the global atmospheric CO₂ levels [7]. It is expected the CO₂ emissions will increase again with the rise in industrial activities and the lifting of travel restrictions. Thus, it remains crucial to study and mitigate about 50% of CO₂ emissions by 2030 to prevent the increase in the global mean temperature of more than 1.5 °C [8]. In order to prevent major climate change, CO₂ concentration in the atmosphere should be reduced either by the CO₂ uptake from the atmosphere, biologically, or the high CO₂ emissions from the sources of fuel-combustion activities, industrial processes, and natural-gas processing [9].

By virtue of the world's dependency on oil and gas as the main sources of energy that is expected to rapidly increase, the usage of level of natural gas globally is approximately 128 billion cubic feet and keeps increasing year by year, as reported by the International Energy Statistic [10]. The energy industry motivates an exploration of the high CO₂ content and impurities of the natural gas reservoirs due to the high demand worldwide. To date, more than 40% of natural gas reservoirs in the world are contaminated with acid gases (such as hydrogen sulfide and carbon dioxide), which hinder the exploitation activities [11]. This paper focuses on the global attention to reduce carbon emissions through offshore natural gas processes, mainly the CO₂ that is present in the original reservoir gas composition.

One crucial strategy for meeting CO₂ emission reduction targets around the world is carbon capture and sequestration (CCS) technology. CCS has been a commercially safe operation for over 45 years, through which almost more than 20 million ppm of CO₂ have been captured and injected into the reservoir since 1972 [12]. There are three steps in CCS, which involves capturing CO₂ at the source, compressing it for transportation via pipelines, and injecting it deep into a rock formation at a carefully selected and safe site, where it is permanently stored. The carbon capture involves the development of CO₂ capture technology for offshore natural gas purification. Recently, a wide range of conventional technologies, such as absorption, adsorption, membrane and cryogenic separation have been further explored to enhance carbon capture technologies for offshore natural gas purification [13].

The adsorption method has started to gain more attention as a method to remove CO₂ from the natural gas streams. This technology has been identified with having a low energy penalty and high energy efficiencies [14]. Solid adsorbents, such as zeolites and activated carbon, allow the gas to accumulate on their surface, forming a film of molecules or atoms due to molecular attraction to the solid surface in a wide range of operating conditions. The process depends upon the intermolecular forces between the adsorbent and the CO₂. The adsorbent is set in the form of a bed in a column and CO₂ is then assimilated. The concerns for this technology are the scale-up and need to develop CO₂-specific adsorbent materials. The adsorbent has shown rapid deactivation, and a longer desorption time is required during the regeneration process. The development of CO₂ adsorbents should satisfy low-cost raw materials, low heat capacity, fast kinetics, high CO₂ adsorption capacity, high CO₂ selectivity, and thermal, chemical, and mechanical stabilities under extensive cycling [15]. In addition, an emerging CO₂ adsorption process using helium has been proposed to mitigate the energy penalty for CO₂ capture by reducing the process temperature [16].

The membrane system includes thin barriers that allow for the selective permeation of one component in the gas stream to pass through faster than the others [17]. It is a pressure-driven process in which the pressure exerted on the solution at one side of the membrane serves as a driving force to separate it into a permeate and a retentate [18]. It offers several advantages over other technologies, including its relatively smaller size and simplicity, which has been identified as moderate technology readiness for commercialization [19].

Nevertheless, membrane separation is accompanied by several challenges in achieving high degrees of separation, so multiple stages and/or the recycling of one of the streams is necessary. This leads to an increase in the complexity, energy consumption, and costs [20]. The good membrane for post-combustion CO₂ capture should have some characterizations, such as high CO₂ permeability, high selectivity, high thermal and chemical stability, resistance to plasticization, resistance to aging, and cost effectiveness [21,22].

Cryogenic separation is a technology that is mostly used for CO₂ removal from natural gas streams with high CO₂ concentrations (typically > 50%) [13]. The cryogenic method involves the separation of the gas mixtures by fractional condensation and distillation at a lower temperature [23]. The process is better than other technologies in several ways, for example, it consumes less energy duty and does not require regeneration and chemical utilization [13]. However, CO₂ removal from natural gas via the cryogenic process needs a highly dehydrated gas stream, which increases the operating and capital cost for the separation process. Additionally, the existence of other gases may obstruct the cooling process and contribute to corrosion problems. The cryogenic process also faces the risk of solid formation due to the complex phase behavior range of CO₂ [24].

Hydrate-based CO₂ separation is an alternative technology, in which the natural gas containing CO₂ is exposed to water at low temperature and high-pressure conditions [25,26]. The basic mechanism of the process is a selective partition of the target components between the hydrate and gas phases. To be specific, CO₂ can form hydrates easier than other gases, such as N₂, due to the differences in the phase equilibrium. The potential barriers of this technology are the ability to release CO₂ from the hydrate in an energy-efficient manner, the efficient capture of CO₂, stable prehydrate, and trace contaminants interfering with the hydrate formation [27]. The need to accelerate the hydrate formation rate and increase the capacity of CO₂ uptake has become a limitation for the use of hydrate-based CO₂ capture technology [28].

To date, absorption technology is one of the most widely used forms of technology used to remove sour gas from natural gas or post-combustion streams. CO₂ separation via absorption is widely used in the industry, due to its capability to be operated under a broad range of CO₂ compositions and lower hydrocarbon losses. In this separation process, CO₂ dissolves into the absorbent and a low temperature is required to ensure a high rapport of CO₂ absorption. The distribution of CO₂ occurs between the gas–liquid interface and the bulk gas, prior to the reaction with the solvent [29]. In the physical solvent process, an organic solvent, such as methanol, was used to allow CO₂ to permeate and dissolve the gas in the liquid phase [15]. The chemical absorption process using aqueous alkanolamine solutions is proposed to be the most applicable technology for a relatively low CO₂ partial pressure system, compared to a physical solvent, as illustrated in Figure 1 [30]. However, the physical solvent could perform well at high CO₂ partial pressure, which leads to a better absorption capacity when compared to the chemical solvent. Hence, the physical absorption has an advantage in the system with a higher CO₂ partial pressure. The aqueous alkanolamine solvent is widely used in gas purification plants, due to its characteristics that are good in relation to physicochemical parameters. Alkanolamine has a high reactivity towards CO₂, a low production cost, and high CO₂ loading [31]. In the amine gas processing operation, the gas stream and liquid amine solution are contacted by a countercurrent flow in an absorption tower. However, the process suffered several drawbacks, including a high corrosion rate, amine degradation, high energy consumption, and requires a larger size of equipment [32]. Thus, the researchers actively made an effort to develop innovative solvents to overcome the drawbacks of the conventional sorbents [33,34]. In the past decade, ionic liquids (ILs) have been proposed as promising alternatives, due to their inherent structure tunability, good affinity with CO₂, and low volatility liquids [35,36]. Haghbakhsh and Raeissi investigated the feasibility of one of the IL classes, deep eutectic solvents (DESs), to be a new generation of green solvents for CO₂ separation [37,38].

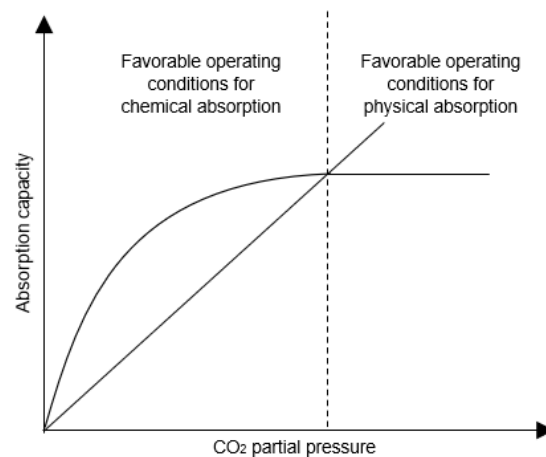


Figure 1. Absorption capacity of physical and chemical solvents [30].

The selection of specific CO₂ capture technologies has become crucial, which heavily depends on CO₂ partial pressure, energy consumption, and footprint requirement [24,39]. Figure 2 illustrates the comparison of CO₂ capture technologies with respect to the CO₂ concentration in flue gas, energy consumption, and footprint [40–42]. For offshore natural gas purification, energy consumption and a compact footprint requirement are among the crucial factors to be considered. Even though the absorption has been proven to be the most established CO₂ capture technology, the conventional solvent regeneration process generally requires excessive energy and footprint/tonnage, which make the overall operation not economically viable for offshore natural gas purification [5].

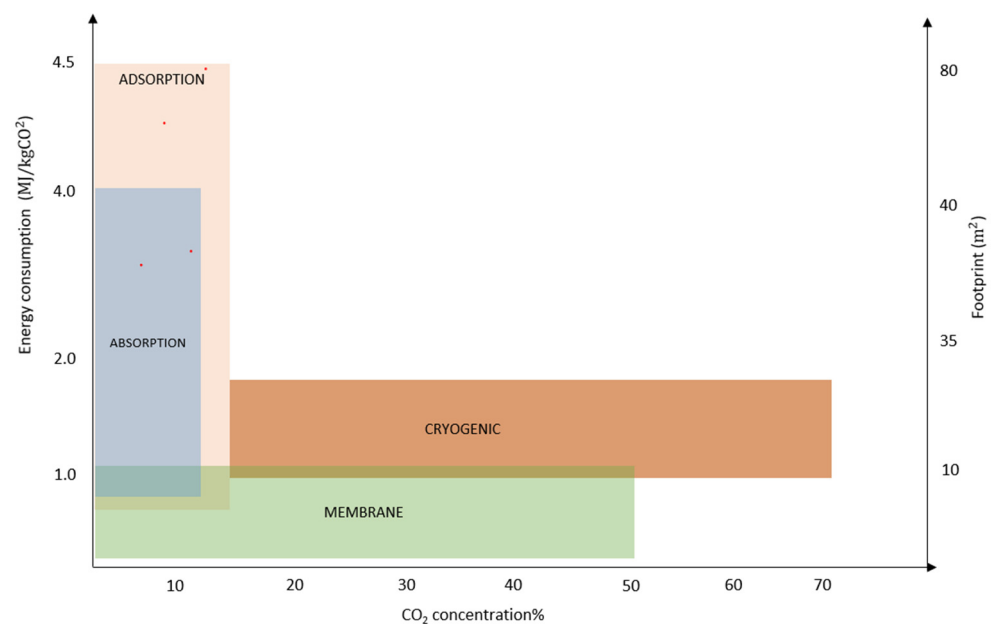


Figure 2. Comparison of CO₂ capture technologies (adapted data [40–42]).

Thus, the conventional and emerging solvent regeneration technologies are critically discussed and analyzed in this review paper. In this work, the flash drum is compared to the developing regeneration technologies, such as membrane contactor, microwave heating, rotating packed bed, and ultrasonic irradiation, in terms of footprint and energy consumption. It is crucial to determine the most promising solvent regeneration technology, which has the capability to overcome the limitations of conventional stripper columns.

2. Conventional Absorption and the Regeneration Operation

In the petroleum and chemical industries, the technology based on liquid absorption is the most mature and commercially available amongst the above-mentioned CO₂ capture options. Absorption by using a chemical solvent is developed from the reaction between CO₂ with one or more basic absorbents that have a specialty to absorb the CO₂ in a high capacity. Several solvents have been adopted for capturing CO₂ from the gas streams, in which amines with alkaline properties are most extensively used to absorb acid gases [43,44]. The treatment of amine gas using a monoethanolamine (MEA) solution is the most advanced process in coal-fired power plants for CO₂ removal, which produces more than 90% of absorption efficiency [45]. In terms of the unit operation, the absorption process is carried out in an absorber that acts as a gas–liquid contractor. The various absorber types, including the packed-bed, tray-tower, and bubble columns, can be used for the CO₂ absorption process.

A stripper, normally designed as a packed-bed/tray column, is the primary device for the regeneration process, which involves the physical mass transfer process and reactions. The absorbed CO₂ in the rich solvent is separated by altering the temperature and pressure in the stripper tower [46]. Figure 3 shows the basic absorber and stripper configuration. The feed gas passes through the absorber column from the bottom to the top, in which approximately 85–90 vol% of CO₂ is absorbed by the solvent [47]. The CO₂-rich solvent is further passed to the stripper to regenerate the solvent, prior to recycling, back to the absorber. The temperature inside the stripper needs to increase for the solvent regeneration process to occur. This thermal regeneration is the most basic and common technique used for CO₂ removal. The regenerated solvent is then sent back to the absorber for the adsorption process, thus creating the continuous recycle process. However, the conventional packed-bed/tray regeneration column suffers from several operational issues, including high energy requirements for stripping, excessive footprint/tonnage, and solvent thermal degradation, during the offshore purification process [48].

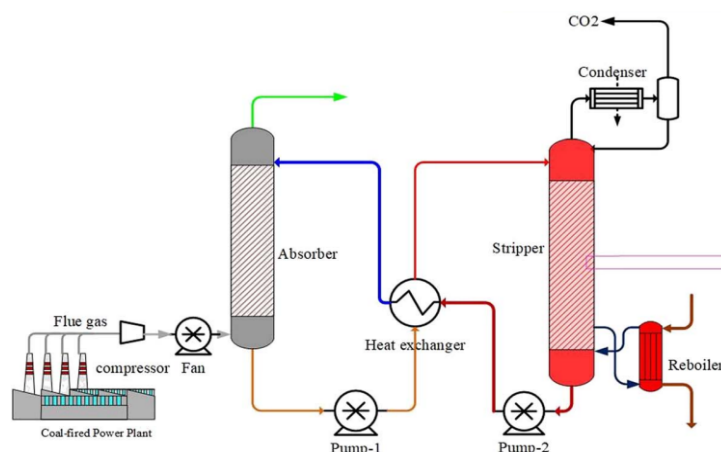


Figure 3. Typical absorber–stripper configuration for the CCS system [49].

3. Emerging Solvent Regeneration Technologies

During the past decades, various regeneration technologies have been proposed to minimize the generation energy and footprint requirement, compared to the conventional packed-bed/tray regeneration column. Therefore, it is crucial to review the development and improvement of each emerging solvent regeneration technology here, to further evaluate their advantages and limitations in this area of research.

3.1. Membrane Contractor

A membrane contactor, having the advantages of a high surface area and high operational flexibility for ease of modularization, is a form of technology that has become a potential candidate for CO₂ desorption [50]. The membrane contactor also possesses

additional benefits as the gas and solvent have separate flow paths that can prevent foaming, flooding, entrainment, and channeling issues [51]. The membrane contactor technology has also been extensively studied by numerous researchers as it has the advantages of a large surface area for the mass transfer, it is lightweight, and has a small footprint for the system [52].

In the desorption process, the membrane is used as the non-selective barrier that is placed between two phases to keep these phases in contact and separated at the same time [53]. The selected gas is diffused to the permeate side through the membrane pores. Then, a sweep gas or steam sweeps out the CO₂ gas on the permeate side to enhance the driving force for stripping the CO₂. The steam is condensed to produce a high purity of CO₂. Figure 4 illustrates the mechanism of the membrane contactor during solvent regeneration [54].

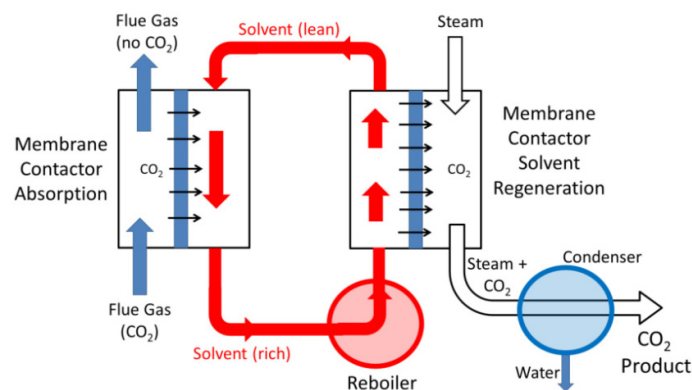


Figure 4. Membrane contractor configuration for separating and purifying CO₂ [54].

The materials used in the membrane play an important role in controlling the efficiency of mass transfer for both the absorption and desorption of CO₂ gas. The porous membrane contactor has been comprehensively studied for its property as a solvent absorption, and the CO₂ is in direct contact with the pores of the membranes to facilitate the mass transfer. Nobuhide Takahashi et al. performed a study on the effect of membrane pore sizes towards the performance of CO₂ desorption. Three different pore sizes, 0.2, 0.5, and 1.0 μm, of a microfiltration membrane (MF) were used to investigate the rate of CO₂ desorption in 30 wt% monoethanolamine (MEA) solvent regeneration at a temperature of 40 °C. They found that the smallest membrane size (0.2 μm) has the highest rate of CO₂ desorption, which is $4.6 \times 10^{-6} \text{ m}^3/\text{m}^2\text{s}$ for the pressure difference of 94 kPa, with a consideration of the volumetric flux as the desorption rate. The foaming phenomena was also recorded at this pressure difference. Additionally, the different membrane thicknesses produced the same trend as the pore size, as the rate of CO₂ desorption increased with the increase in the membrane thickness, where a 2.9 mm membrane thickness with a pore diameter of 1.6 μm produced $450 \times 10^{-6} \text{ m}^3/\text{m}^2\text{s}$ of CO₂ desorption rate [55]. It is crucial to maintain the permeate area at high pressures to avoid the steam from entering the solvent area due to the pressure difference. The phenomena causes the solvent to wet the membrane pores, which can lead to mass transfer reduction due to the slower diffusion rate [56–61].

As the porous membrane could lead to mass transfer reduction due to the wetted and foaming issues, several studies were conducted on non-porous membrane contactors, which, it is believed, could overcome this issue [62–64]. The non-porous membrane separates the gas and liquid phases, where the CO₂ passes through the high permeate polymer as a gas [61,65,66]. Colin et al. performed the regeneration of MEA using a non-porous polydimethyl siloxane (PDMS) membrane. The study was carried out at a temperature range of 90–110 °C with a 20 kPa pressure difference and a 4 μm membrane pore size. The highest temperature (110 °C) led to the highest mass transfer coefficient of CO₂ to the permeate side, which was 0.3 cm/s, while the mass transfer coefficient of CO₂ for the other temperatures mostly demonstrated a similar value of 0.01 cm/s [67].

Membrane contactors have a potential to be used in both absorption–desorption considerations, however the solvent selectivity for the separation should be good to avoid the membrane wettability and to reduce solvent degradation for long-term applications. This hybrid pressure-driven technique requires absorbent regeneration to occur in a continuous mode of absorption processes in series or parallel configurations [68,69]. The process offers the potential of a reduction in the energy regeneration in the series case and capital costs in the parallel case. The membrane contactor permits the mass transfer from a liquid phase to a gas phase, or vice versa for the gas–liquid membrane separation. The separation occurs through the membrane due to the driving force generated by the difference of partial pressure or difference of concentration at both sides of the membrane. Forcing the dispersed phase through the membrane created a constant quantity of droplets that produced uniform emulsions. This phenomenon could reduce the energy input as well as the wall shear stress to the membrane [70]. Witchipan et al. performed a modeling study to analyze the energy requirement for CO₂ desorption from a CH₄ solvent. It was reported that the combination of the membrane contactor with a slightly vacuum condition on the permeate side could provide a high-net energy, which is 0.178 MJ/m³. The value of this net energy is equivalent to the 85.37% reduction from the total energy required to generate up to 90% of CH₄ regeneration from the CO₂-rich solvent [71].

The membrane contactor has been recognized to provide a better mass transfer due to the large contact area, compared to the conventional process. Additionally, the membrane contactor provides 500 times more contact area than the liquid–liquid extraction column, and 30 times more compared to the gas absorber [72]. Normally, a conventional packed-bed column provides an active interfacial area between 100 to 250 m²/m³. The hollow fiber membrane could cover the same value of interfacial area with the compact size in the design. However, the usage of the membrane contactor faces some constraints. Firstly, the gas and liquid flow is constrained by the pressure drop coming from the packing density of the fiber membrane. Secondly, the porosity of the membrane could reduce the mass transfer area due to the occurrence of a chemical reaction within the boundary layer [54].

Karl Anders Hoff et al. performed modeling work to study the size reduction in the membrane contactor, compared to the packed column in the absorption–desorption cycle process [73]. Figure 5 shows the potential of a membrane contactor to replace the absorber and desorber unit for the typical gas treatment system. The rectangular modules were used for the post-combustion capture with a dimension of width, length, and depth were 3 m, 3 m, and 1 m, respectively, while the high pressure modules are used for the gas treatment with a 2-to-2.5 m maximum diameter and a 5 m maximum length. They found that the total volume of the separator is reduced by 75%, compared to the conventional absorber and desorber tower. The overall size reduction obtained from the modeling results is shown in Table 1.

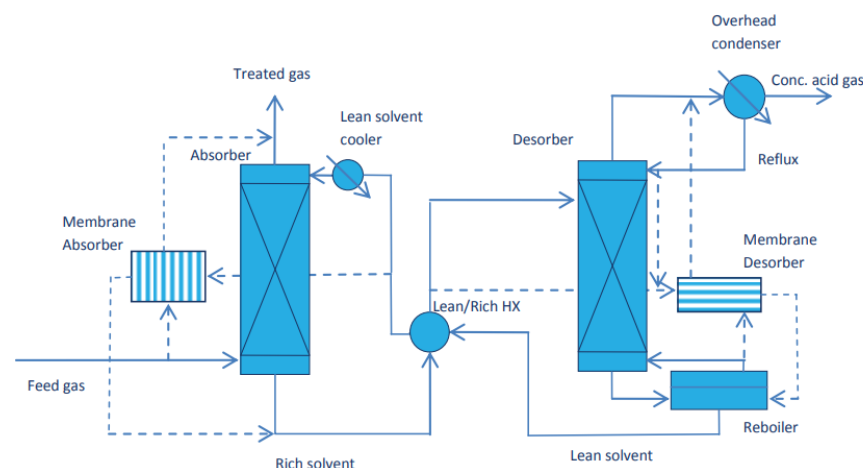


Figure 5. Simplified absorption and desorption processes that have the potential to be replaced with a membrane contactor system [73].

Table 1. Adaptation from the simulation results of the membrane contactor vs. packed tower [73].

Scenario	Post-Combustion Capture	Natural Gas Treatment	Natural Gas Treatment with the Presence of Promoter
Solvent used	30% MEA	30% MDEA	30% MDEA 5% piperazine
Liquid flow mode	Shell side	Shell side	Shell side
Fiber length (meter)	3	5	5
Total packed volume (m ³)	675	52	113
Volume reduction	75%	75%	54%

3.2. Microwave in Solvent Regeneration

Recently, the idea has been proposed of using microwave heating as an alternative approach to overcome solvent regeneration in CO₂ removal. Microwave (MW) irradiation uses the same process as conventional thermal heating, with a slight difference in that it consumes less energy. The performance of MW irradiation depends on the ability of molecules with a dipole moment to absorb the MW energy and convert it into heat [74]. Figure 6 shows that MW heating instantly heats the molecules in the solvents through dipolar rotation and/or ionic conduction, which produce heating that is more efficient and conducted in a safer mode [75]. Chronopoulos et al. concluded that MW could offer the possibility of a more accelerated overall CO₂ desorption process, contributing to a process that is four times faster, compared to conventional heating desorption [76].

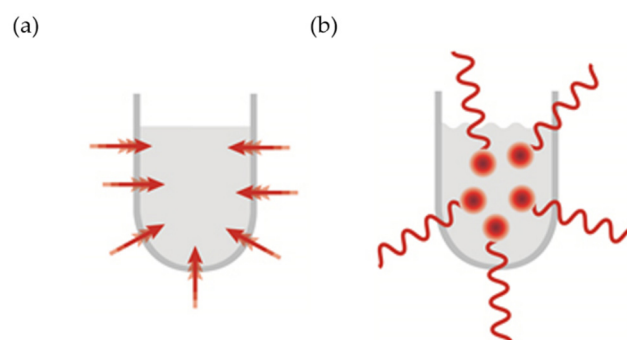


Figure 6. Graphic illustration of heat induction for (a) conventional heating and (b) microwave heating (adapted from [75]).

There are two significant constants involved in the MW approach, which are the dielectric constant and dielectric loss factor. The dielectric constant is used to measure the ability of a substance to store energy, while the ability of that substance to convert the electromagnetic energy into heat can be determined by calculating the dielectric loss factor [77,78]. These two constants are in the functions of the MW frequency, temperature, and molecule interaction inside the solution [79]. Thus, MW has been known as an instantaneous and volumetric heating process with an unlimited heat transfer capability.

Chudburry et al. conducted a study on the direct and indirect effects of microwaves on the Na-ETS-10 solvent [80]. Indirect microwave regeneration is defined as the desorption that occurs in the water solvent first, and is then followed by microwaved drying. However, direct microwave regeneration is defined as the constant microwave power that is supplied from the beginning to the end. They found that direct microwave regeneration could produce a 22% higher desorption capacity and lower power consumption of 16.6%, compared to indirect microwave heating after 5 cycles of CO₂ desorption [80]. This phenomenon, as a result of high energy, is required when performing indirect microwave regeneration, as CO₂ is first desorbed in the conventional method, before the introduction of the microwave thermal regeneration approach [80].

Bougie and Fan highlighted in their study that MW irradiation could reduce solvent regeneration energy by up to 33% for the 50 wt% of the MEA solvent, compare to the value found for the benchmark 30 wt% MEA [74]. They concluded that the MEA concentration

significantly influenced the cycling absorption–MW regeneration process. Energy saving could only be achieved if the absorption–regeneration is cycled at least 3 times to achieve that outcome. Furthermore, a study conducted by McGurk stated that the MW could rapidly regenerate the MEA at low temperatures, which is about 70–90 °C, compared to the conventional thermal regeneration at high temperatures, 120–140 °C, which leads to the overall cost reduction [81]. However, solvent regeneration by using MW is a slow process as it requires absorption–regeneration cycling, and the operating temperature (70–90 °C) is still considered to be high.

3.3. Flash Drum

The flash drum is a simple separator that can be used to separate/flash out the absorbed gas in CO₂-rich solvents. Typically, an intermediate pressure flash drum is used as the absorbed product gas is desorbed and transformed into a vapor phase due to the flashing effect. Therefore, the volume of the gas phase increases and leads to a higher gas flow rate, whereas the produced gas that remains in the solvent is reduced to an insignificant level and reduces the waste of the product gas. The flash gases from the intermediate pressure flash drum are recycled to recover the product gas, as it contains a significant amount of hydrocarbons. The successive flash drum at a lower pressure mainly releases CO₂ and flash regeneration is carried out at a near-atmospheric pressure [22]. The lean solution is cooled and returned to the high-pressure absorption column after the regeneration process. The regenerated lean solution is used again for the CO₂ absorption process in the high-pressure absorption column. Technically, to prevent the liquid from coming out of the top of the gas outlet, the flash drum is installed with de-entrainment mesh pad.

In comparison to other technologies, the flash drum is the simplest form of technology that does not have complex accessories and operations, which lead to the ease of maintenance. Typically, the simplicity of the technology leads to the reduction in the operation expenditure (OPEX). In general, the sizing of the flash drum depends on the flow rate of the gas and liquid that is released from the tank [82]. The higher the flow rate of the gas and liquid released from the flash drum, the larger the size of the flash drum required. In addition, the major drawback of this process is that a series of the flash drums is required for having a high quality of regenerated solvent. A single column of the flash drum is not usually capable of regenerating the solvent as well as the other forms of technology. Moreover, one of the disadvantages of using the flash drum in the regeneration process is the pressure loss of CO₂ during the recovery process. Therefore, the flash drum is usually operated at an ambient pressure, which requires an additional compression system to meet the requirement of transferring the gas and liquid product to the pipeline. Figure 7 illustrates the employment of two-stage flash for CO₂ regeneration to improve the efficiency and make the process more economical [83]. Despite the excessive footprint, the flash drum still remains the most widely used method for physical solvent desorption.

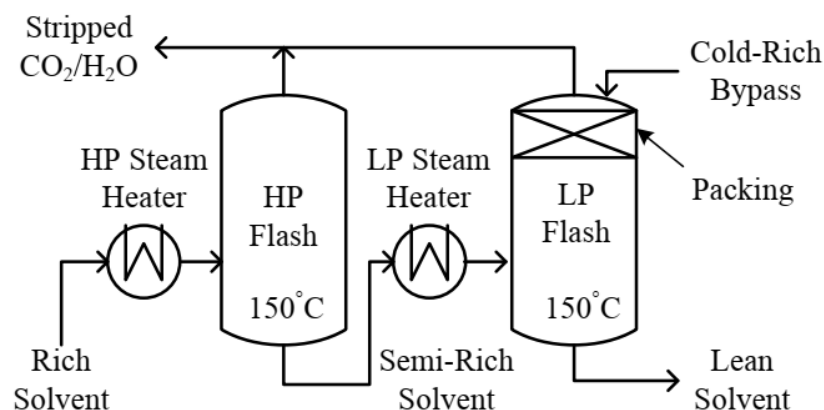


Figure 7. Simplified two-stage flash process flow diagram [83].

3.4. Rotating Packed Bed (RPB)

Rotating packed bed (RPB) has gained attention for the potential of process intensification in CO₂ separation. RPB was invented by Ramshaw and Mallinson in 1981 to intensify the mass transfer process, compared to the conventional packed bed [84]. As the conventional packed bed in real industry application faced several challenges, such as a large footprint and low mass transfer coefficient, several studies have been conducted on RPB for the potential of replacing the packed bed in the post-combustion application. The RPB was developed to reduce the column size and the complexity of the CO₂ absorption plant. Ramshaw reported that the RPB provides a volume of reduction by 2–3 orders of magnitude; additionally, the height equivalent of a theoretical plate (HETP) of RPB can be as low as 1–2 cm, when compared to the conventional packed-bed column [85].

In the typical RPB for the desorption process, the rich solvent flows through a rotating packed bed from the middle part of the RPB and is distributed through the thin liquid films generated by the centrifuge force, which leads to a greater mass transfer [86]. In addition to the absorption and desorption processes, the RPB has been used in distillation and material production. Figure 8 illustrates the cross-section of the RPB for the desorption process. The standard counter-current RPB is equipped with the casing, liquid solvent distributor, dynamic seal, and rotor shaft with the packing. The liquid solvent enters the RPB via the liquid inlet and is distributed in the packed-bed section. Then, the solvent is pushed out from the packed bed by the centrifugal force. The lean solvent is later collected at the bottom section of the RPB. Meanwhile, the gas flows inward to the packed bed and leaves the RPB through the central section, as shown in Figure 9. A reboiler is required to heat the solvent for a better desorption process rate.

Atuman et al. performed simulation work to study the size reduction in the RPB, compared to the conventional packed bed [87]. This simulation was validated using an experimental study conducted by Jasim et al. [88]. The simulation study of the CO₂ desorption process from MEA was conducted under some fixed conditions, such as rich-MEA solvent flow rate (0.3 kg/s), temperature (104 °C), pressure (202 kPa), and solvent loading (0.482 mol CO₂/mol MEA) for both the RPB and conventional packed bed. It was found that the packing volume for the RPB was 0.015 m³, while the conventional packed bed was 0.659. The results show that there is a 44 times reduction in the packing volume for the RPB, compared to the conventional packed bed. Additionally, the calculated height of the transfer unit for the RPB is 1.7 cm, while the conventional packed bed is 20.8 cm. This result contributes to the smaller size of the RPB, compared to the conventional packed bed [87]. Based on the finding by Agarwal et al., the casing volume of the RPB and the packed bed was 4.5 times its packing volume [89]. Hence, the overall volume of the RPB is concluded to be 9.7 times smaller than the conventional packed bed.

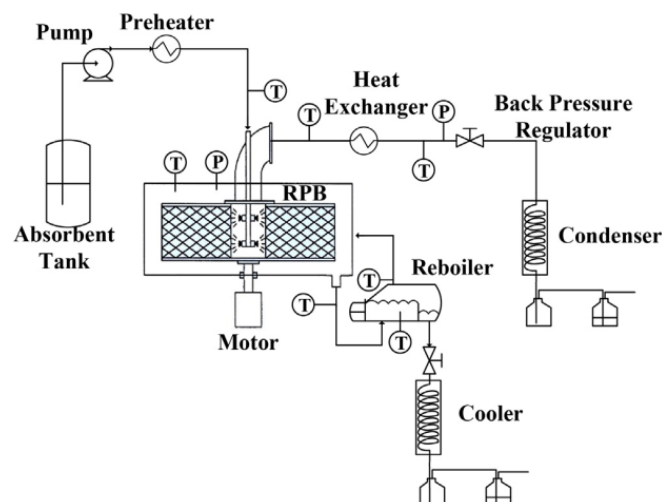


Figure 8. Experimental apparatus of the rotating packed bed for solvent regeneration [90].

The RPB is also known as the energy-intensive system for the solvent regeneration process. The regeneration energy refers to three types of heat required during the desorption or solvent regeneration process, which are the heat of the reaction with CO₂, the sensible heat of the solution, and the heat of vaporization. For the energy reduction strategy, these three types of energy should be minimized to as low as possible. The heat of the reaction with CO₂ can be adjusted through the heat energy required for the thermal liquid solvent regeneration. Meanwhile, the sensible heat of the solution and the heat of vaporization can be reduced via the optimization of process conditions with a good process design [90]. Apart from these three types of heat required during generation, the reboiler energy and the power of the rotor rotation are crucial to be included for the overall energy consumption.

Chamchan developed a model to study the energy consumption for conventional packed bed and RPB which have been validated from pilot plant at China Steel Corporation (CSC) [91]. This packed-bed pilot plant was equipped with 16mm of packed SUS pall ring, where the diameter of the packed-bed column was 0.1 m with a 2 m height. Meanwhile, the RPB has a 0.06 m height, 0.36 m outer diameter, and 0.12 m inner diameter, which use a 5428 cm³ wire-mesh packing volume. The comparable energy consumption for the conventional packed bed and RPB were obtained using an ASPEN Plus Rate-Based model. The authors found that the energy consumption for the RPB was slightly higher than the packed bed. The RPB consumed about 7.5 GJ/tons of CO₂, while the packed bed consumed about 6.5 GJ/tons of CO₂ for the regeneration of the MEA solvent [91]. Additionally, Tan and Chen performed a study to identify the power consumption rate for the RPB by using the equation developed by Singh et al. [92]. The results from the study showed that 1.2 kW of power was required for the flow rate of 90 cm³/min. However, this power consumption value is unacceptable for a techno-economy CO₂-removal process [93]. Thiels et al. proposed to combine the RPB with a packed-bed column as a method to reduce the energy consumption of the RPB [94].

3.5. Ultrasonic Irradiation

Recently, ultrasonic irradiation has been found to be a potential alternative for a high mass transfer rate without having any moving part [95,96]. This advantage was applied to the gas–liquid absorption and desorption process. Ultrasonic irradiation is a sound wave that has a higher frequency (20 kHz to 500 MHz) than the limit that can be heard by human beings. Furthermore, the ultrasonic irradiation is usually used to enhance the gas–liquid phase reaction mostly involving the physical kinetic effect, as it can produce chemical and physical effects on the liquid phase. In the natural gas purification process, especially in the desorption process, the ultrasonic wave is able to perform the cavitation and nucleation in the liquid solvent and leads to the bubble's formation. The gas diffuses in these bubbles and causes the sour gas to escape through the bubbles. The more bubbles that are created, the higher the occurrence of the gas desorption rate. The cavitation effect is more favorable for the desorption process rather than the absorption process, which relies on the atomization and the fountain effect from the solvent vibration [97]. Hence, the development of ultrasonic columns for desorption or solvent regeneration always fulfilled the cavitation effect.

The cavitation effect is typically produced by a low frequency of ultrasonic irradiation [98,99]. The cavitation or the bubble formation can only be produced under the rarefaction phase of the ultrasonic irradiation [100]. A stable microbubble formation is required to improve the desorption process, which can either be stable at a certain average formation size in a many-cycle rate when growing and exploding, or collapse during the ultrasound wave compression [101–105]. Based on this theory, the desorption or the degassing process could be further improved using ultrasonic irradiation. Figure 9 shows the ultrasonic irradiation column for the gas purification adapted from a study by Tay et al. [106].

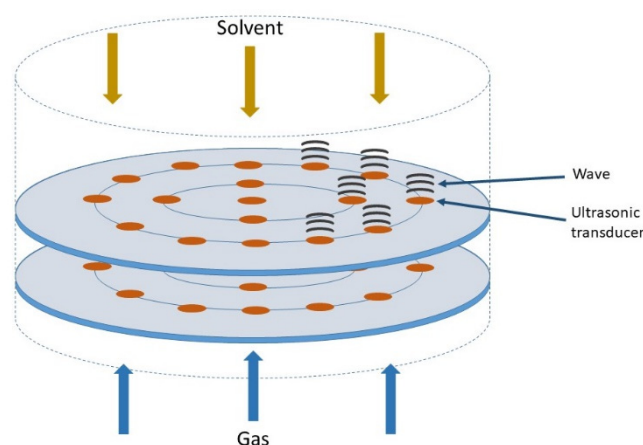


Figure 9. Ultrasonic irradiation column [106].

The concept of using ultrasonic irradiation can be used to enhance the desorption rate as well as the desorption capacity of the flue gas, which is due to the high mass transfer contributed to by the physical kinetic effect in the liquid phase. Several studies proved that ultrasonic irradiation has the potential to improve the mass transfer process during the desorption process [107–109]. Jiru Ying et al. conducted a study to investigate the effect of ultrasound on the CO₂ desorption from a 30 wt% MEA solvent using a 28 kHz ultrasonic transducer [110]. The authors found that the desorption rate of CO₂ increased as they introduced the ultrasound, compared to the CO₂ desorption from the heating treatment. The desorption rate of CO₂ was measured in terms of the CO₂ flow rate vented from the vessel using a flow meter. The flow rate of CO₂ desorption measured in the presence of ultrasonic irradiation was 1.51 g/min, while the flow rate of CO₂ desorption measured with the usage of heating treatment was 0.30 g/min. The results show that ultrasonic irradiation can enhance the desorption rate by about 5 times, when compared to the direct heating process. This was due to the fact that more bubbles are formed in the vessel equipped with the ultrasonic wave, which remarkably provides a large surface area for the gas–liquid contact area [111]. Moreover, this study also indicated that the ultrasonic irradiation could reduce the energy consumption for the CO₂ desorption, as it did not require any heating duty. However, the other energy consumed, such as electrical energy, needs to be considered to prove the power reduction during the CO₂ desorption, while using the ultrasonic irradiation.

Additionally, S. Gantert and D. Moller [112] performed a study on power consumption during the CO₂ desorption in a batch reactor. The study focused on the desorption of CO₂ from 30 wt% of MEA and compared multiple ultrasonic intensities (10–50 W) with the same ultrasonic frequency. The authors found that the highest ultrasonic intensity, which was 50 W, produced the highest CO₂ desorption flow rate (3.2×10^{-2} m³/s). This indicates that the highest intensity of ultrasonic irradiation led to the highest amount of bubble formation. Additionally, no desorption process occurs at 40 °C, while desorption starts to occur at the temperature of 45 °C, and achieved the peak level at 80 °C. The typical packed column required 100–120 °C to initiate the desorption process [113]; hence, ultrasonic irradiation is capable of reducing the energy consumption during the desorption process. As a low temperature (80 °C) is required to initiate the process, the degradation of MDEA can also be reduced. Davis and Rochelle reported that the degradation of MEA is reduced by a factor of 4 when decreasing the temperature for every 17 °C [114].

To date, several researchers tried to perform a study to reduce the footprint of the desorption column. A small footprint of the desorption column is essential, especially for the offshore natural gas purification process, due to the space limitation. Based on the previous works [97], the intensification factor for ultrasonic irradiation can be high, up to 16 times, compared to the packed-bed column. Due to this, this technology can also be used with a slow kinetic solvent, such as MDEA. Typically, the commercial ultrasonic transducer

disc is only about 2 mm thick, which contributes to the minimal footprint requirement for a single ultrasonic vessel. Due to the compact and simpler design, the manufacturing cost for the ultrasonic irradiation column is believed to be less, which is mainly contributed to by the vessel body, piezoelectric ultrasound transducer, and the power supply [115].

4. Advantages and Challenges of the Emerging Regeneration Technologies for the Offshore Natural Gas Purification Process

The offshore natural gas purification process should be energy efficient and compact. In order to fulfill these requirements, various emerging solvent regeneration processes have been established. Table 2 shows the comparative analysis of the emerging solvent regeneration technologies for the offshore natural purification process, in terms of the energy consumption and size reduction. As a summary, membrane contactor technology is the most promising emerging solvent regeneration technology, since this form of technology can significantly reduce the energy consumption and footprint requirements, followed by ultrasonic irradiation technology, microwave, and rotating packed-bed technology. Yet, all these emerging technologies suffer from several drawbacks in terms of their operational and maintenance difficulties, and material reliability. To ensure the feasibility of these technologies, these highlighted issues should be addressed prior to their full commercial application.

Table 2. Comparison analysis between the emerging regeneration technologies.

Technologies	Energy	Size	Advantages	Disadvantages
Membrane contactor [71,73]	−85%	−54%	<ul style="list-style-type: none"> • High mass transfer coefficient • Compact size 	<ul style="list-style-type: none"> • Wetting and foaming issues lead to a reduction in the mass transfer • Short life cycle of the membrane • Low solvent flexibility limits the range of solvent usage
Flash drum [82,83]	Not reported	Not reported	<ul style="list-style-type: none"> • Less complexity, which means that it is easy to operate • Less maintenance needed • Less OPEX 	<ul style="list-style-type: none"> • Large footprint • Required a series of flash drums for better efficiency • Required additional systems for transferring the product
RPB [87,89,91]	+115%	−9.7%	<ul style="list-style-type: none"> • High solvent flexibility offers a high range of solvent usage • High stability 	<ul style="list-style-type: none"> • Large regeneration energy • Regular maintenance due to moving parts
Ultrasonic irradiation [112,115]	−66%	−16%	<ul style="list-style-type: none"> • High solvent flexibility offers a high range of solvent usage • High stability • Compact size • High mass transfer coefficient 	<ul style="list-style-type: none"> • High electrical energy leads to high OPEX • Difficulty to replace the piezoelectric transducer
Microwave [74,80,81]	−16.6%	−22%	<ul style="list-style-type: none"> • Provide an instant heating effect • High solvent flexibility 	<ul style="list-style-type: none"> • Slow process to obtain the lean solvent

5. Conclusions

The choice of CO₂ capture technology heavily depends on CO₂ partial pressure, energy consumption, footprint requirement, and the targeted quality of the gas product. These factors are crucial to be considered in offshore applications. CO₂ separation via absorption is widely used in the industry, due to its capability to be operated under a broad range of CO₂ compositions and lower hydrocarbon losses. Despite being the most mature form of technology, the application of absorption for the offshore natural gas purification process is

hindered by a high solvent regeneration energy and excessive footprint requirement. The development of emerging regeneration technologies in CO₂ capture, such as membrane contactor, flash drum, rotating packed bed, microwave heating, and ultrasonic irradiation, contributed to the closure of these technology gaps. Generally, the conventional technologies, such as the packed column and flash drum, produce a large footprint and tonnage, which restrict their application in the offshore natural gas purification process. The RPB column and microwave heating demonstrate a higher solvent flexibility and process stability, but suffer from extensive maintenance and high regeneration energy. The membrane contactor and ultrasonic irradiation absorption system are compact, yet their application in the offshore natural gas purification process is constrained by various operational issues. Thus, future research is necessary to fully comprehend the viability of these technologies for achieving the most efficient process for solvent regeneration through the offshore natural gas purification process.

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