BMC Chemical Engineering

REVIEW Open Access

Process intensification technologies for CO_2 capture and conversion – a review

Abdullahi Adamu, Fernando Russo-Abegão and Kamelia Boodhoo*

Abstract

With the concentration of CO_2 in the atmosphere increasing beyond sustainable limits, much research is currently focused on developing solutions to mitigate this problem. Possible strategies involve sequestering the emitted CO_2 for long-term storage deep underground, and conversion of CO_2 into value-added products. Conventional processes for each of these solutions often have high-capital costs associated and kinetic limitations in different process steps. Additionally, CO_2 is thermodynamically a very stable molecule and difficult to activate. Despite such challenges, a number of methods for CO_2 capture and conversion have been investigated including absorption, photocatalysis, electrochemical and thermochemical methods. Conventional technologies employed in these processes often suffer from low selectivity and conversion, and lack energy efficiency. Therefore, suitable process intensification techniques based on equipment, material and process development strategies can play a key role at enabling the deployment of these processes. In this review paper, the cutting-edge intensification technologies being applied in CO_2 capture and conversion are reported and discussed, with the main focus on the chemical conversion methods.

Keywords: Process intensification, CO_2 conversion, Carbon capture, Photocatalytic, Electrochemical, Thermochemical, Plasma, Membrane, Renewable fuels

Background

Since the beginning of the industrial revolution in the late 1700s, the global demand for energy has continuously risen, and fossil fuels have been used to cater for this demand [1, 2] with a projected demand of about 778 Etta Joule by 2035 [3]. According to the British Petroleum Statistical Review of World Energy in 2019, energy consumption rate has, in the last 10 years, almost doubled to 2.9% in 2018, while oil production rate grew at 2.2%, more than double the average growth of 1% from 2007 to 2017 [4]. The major repercussion of using fossil fuels is the emission of CO_2 into the atmosphere which has surpassed 400 ppm since 2015 (Fig. 1) [5]. To restore the carbon cycle, approaches such as switching to renewable energy sources [6], efficient energy usage [7] and carbon sequestration and utilisation [7, 8] have been considered.

While the carbon capture approaches can remove on average 50–94% of the emission from cement and fossil fuel-fired plants [9], technologies to capture CO_2 released from energy production and transport sectors are less well developed although these account for 25% [10] and 50% [9] respectively of the global greenhouse gas emission. There is a necessity to develop new ways to capture atmospheric CO_2 originating from these various sources. In 1999 Lackner et al. [11] demonstrated the feasibility of large-scale direct CO_2 capture from air and this technology is now at early stages of commercialisation [9]. Currently Climeworks in Switzerland, Global Thermostat in collaboration with Exxonmobil and Infinitree LLC in USA, Giaura in Netherlands, Oy Hydrocell Ltd. in Japan and Carbon Engineering [12] are actively engaged in establishing commercial-scale direct air capture. All these companies, except Carbon Engineering, employ a cyclic absorption-desorption process. The advantage of such a system is it requires low energy and capital input. However, the challenges involve the need for large facilities with a periodic sealing from air during regeneration [13].

Carbon Engineering [13] reported details of their direct air capture process which involves pulling in atmospheric air through a contactor device, followed by CO_2 absorption in aqueous sorbent (KOH). As shown in Fig. 2, the process consists of two linked chemical loops. Not only do they capture CO_2 from air, they also transform the CO_2

* Correspondence: kamelia.boodhoo@newcastle.ac.uk
School of Engineering, Newcastle University, Newcastle upon Tyne NE1 7RU, UK

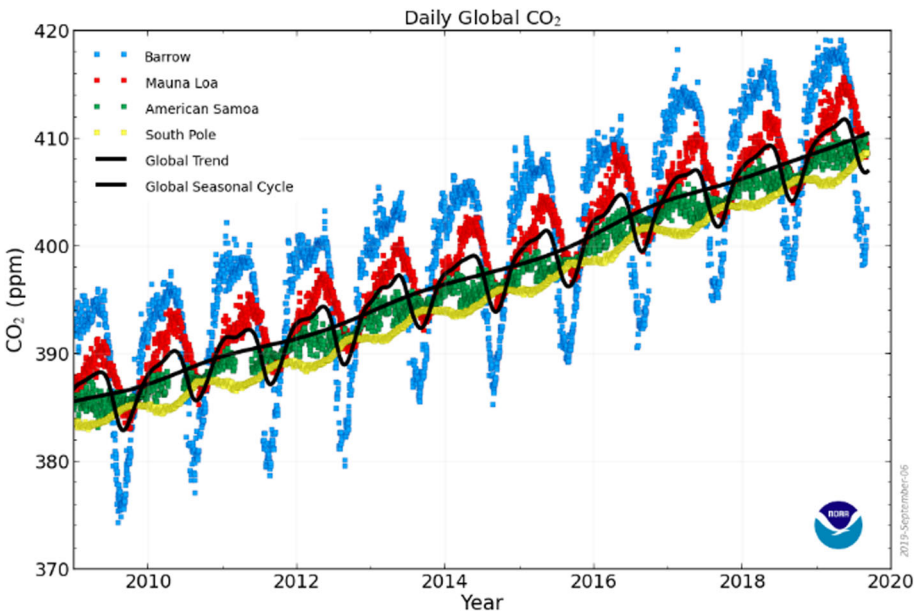

Fig. 1 Daily averaged CO_2 from four GMD Baseline observatories. Image provided by NOAA ESRL Global Monitoring Division, Boulder, Colorado, USA (http://esrl.noaa.gov/gmd/) [5]

back into fuel, creating net-neutral carbon based fuels. Despite being a promising technology, direct air capture is powered by natural gas and it is at the early stage of realisation, creating uncertainties due to lack of detail specifications [13].

Storing captured CO_2 underground is currently the most readily available option. A more sustainable solution to underground storage of captured CO_2 which poses risks of leakage [14] is to create an artificial carbon cycle shown in Fig. 3, where renewable energy sources drive the CO_2 conversion into fuel and chemicals or fuel precursors [8, 15].

With CO_2 being poorly soluble in water and thermodynamically stable, converting it to added-value products in aqueous medium requires a high energy input. Furthermore, CO_2 reduction via electrochemical, thermal, biochemical and photochemical routes coupled with heterogeneous catalysts/enzymes suffers from the drawback of poor selectivity, low efficiency and cost-intensive processing [16] arising from the multitude of products that can be formed, as shown in Fig. 4.

Role of process intensification in CO_2 capture and conversion

Process intensification (PI), a technique aimed at modifying conventional chemical processes into more cost-effective, productive, greener and safer processes [17],

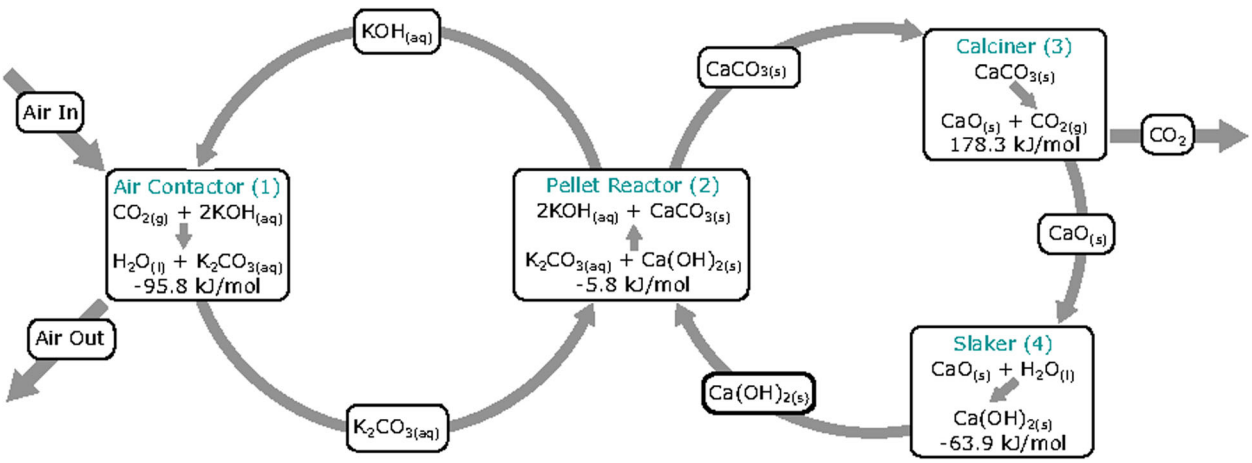

Fig. 2 Process Chemistry and Thermodynamic of Direct Air Capture by Carbon Engineering. Reprinted from [13] DOI: https://doi.org/10.1016/j.joule.2018.05.006; licensed under the CC BY-NC-ND 4.0 license

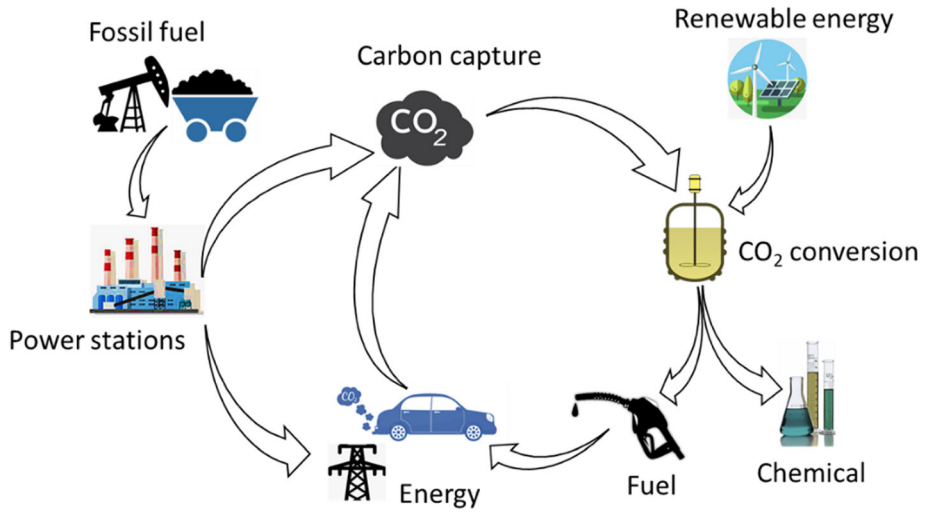

Fig. 3 CO_2 conversion cycle

offers the opportunity to address some of the challenges encountered in CO_2 capture and conversion.

PI technologies are characterised by equipment size reduction through enhanced and targeted mixing, and mass/heat transfer, leading to improved selectivity, high energy efficiency, reduced capital cost and waste reduction [17]. The smaller processing volumes handled in intensified systems offer the potential to reduce material costs and improve safety.

Even greater intensification levels can be realised by astutely combining synergies of equipment, materials and methods (Fig. 5) [18], and by combining two or more technologies in a given process. Such combinations are able to utilise the specific advantages of each component, whilst aiming to supress any associated constraints of a particular aspect of the process. For instance, in the case of CO_2 reduction, electrochemical reduction can be incorporated with photocatalysis, which provides the driving force to initiate the process [19]. Similarly, replacing an intensive energy source with a more efficient and ideally renewable source can lead to intensification of CO_2 reduction. For instance, compared to CO_2 activation using high-cost thermal energy, high-energy non-thermal plasma has shown an improved performance [20]. For such combinations or substitutions to be effective, it is important to understand the important properties in material and/or devices that will efficiently and affordably reduce CO_2 to value-added products. Herein, the intensification of carbon capture and of key CO_2 reduction

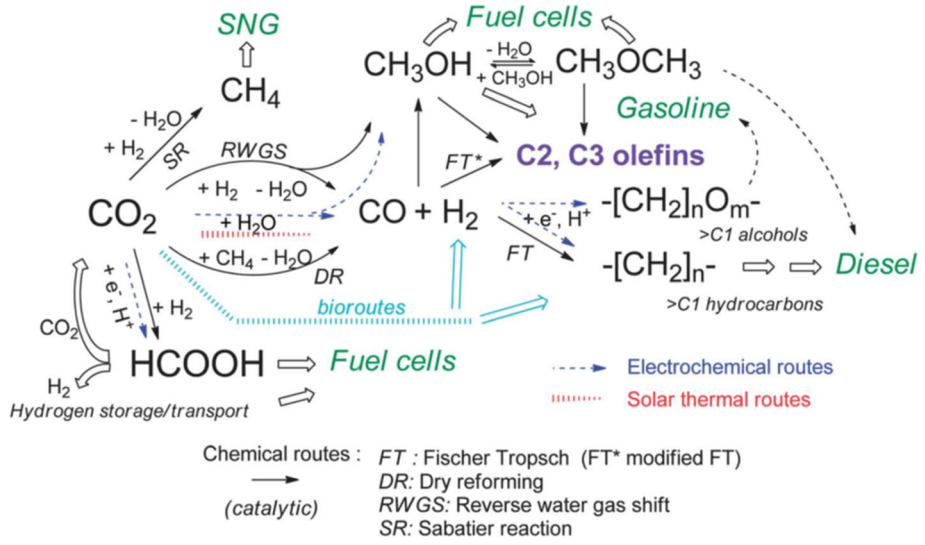

Fig. 4 Schematic overview of CO_2 conversion routes. Reprinted from [6], Copyright (2013), with permission from The Royal Society of Chemistry

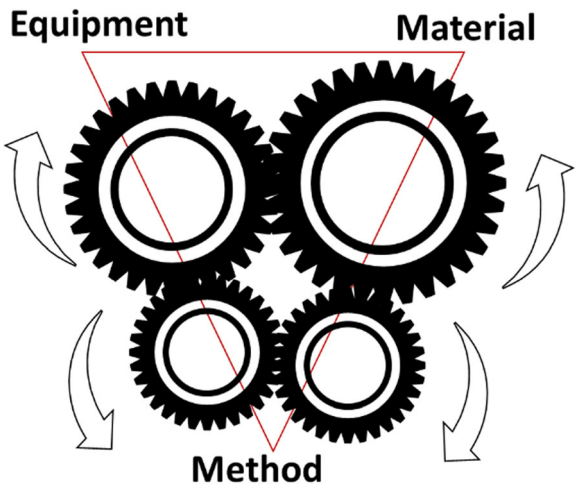

Fig. 5 Integrated approach of process intensification

methods including photocatalytic, electrochemical, biochemical and thermochemical processes is reviewed, focusing on the integration of the three PI approaches of equipment, materials and methodology shown in Fig. 5.

Intensification of CO_2 capture and sequestration

Among the methods that have the potential to reduce the atmospheric CO_2 concentration is carbon capture (CC). Pre-combustion, post-combustion (PCC) and oxy fuel capture are the approaches used for CC applying different processes such as membrane separation, adsorption, chemical absorption, physical absorption, chemical looping and calcium looping [21]. In PCC where CO_2 is captured from exhaust streams in fossil fuel-fired plants, intensification of chemical absorption has been extensively investigated and will form the basis of this review on CC. Readers interested in pre-combustion capture and oxyfuel capture are invited to consult appropriate references [22–24].

Chemical absorption

Chemical absorption involves CO_2 captured by contacting the exhaust gas with an aqueous absorbent in a conventional packed column. This approach is widely used in PCC in power and cement plants using conventional amine and ammonia based absorbents [25]. Figure 6 depicts the simplified process diagram for conventional PCC. In these systems, once the absorbent in the column becomes saturated with CO_2, it is passed onto a stripper/regenerator column, where a stream of superheated steam at around 100–120 °C [27] is passed to regenerate the absorbent and releases the captured CO_2, making the absorbent CO_2 lean. The lean absorbent is sent back into the absorber for another cycle. PCC is cost intensive [28], however it is currently the most fully developed

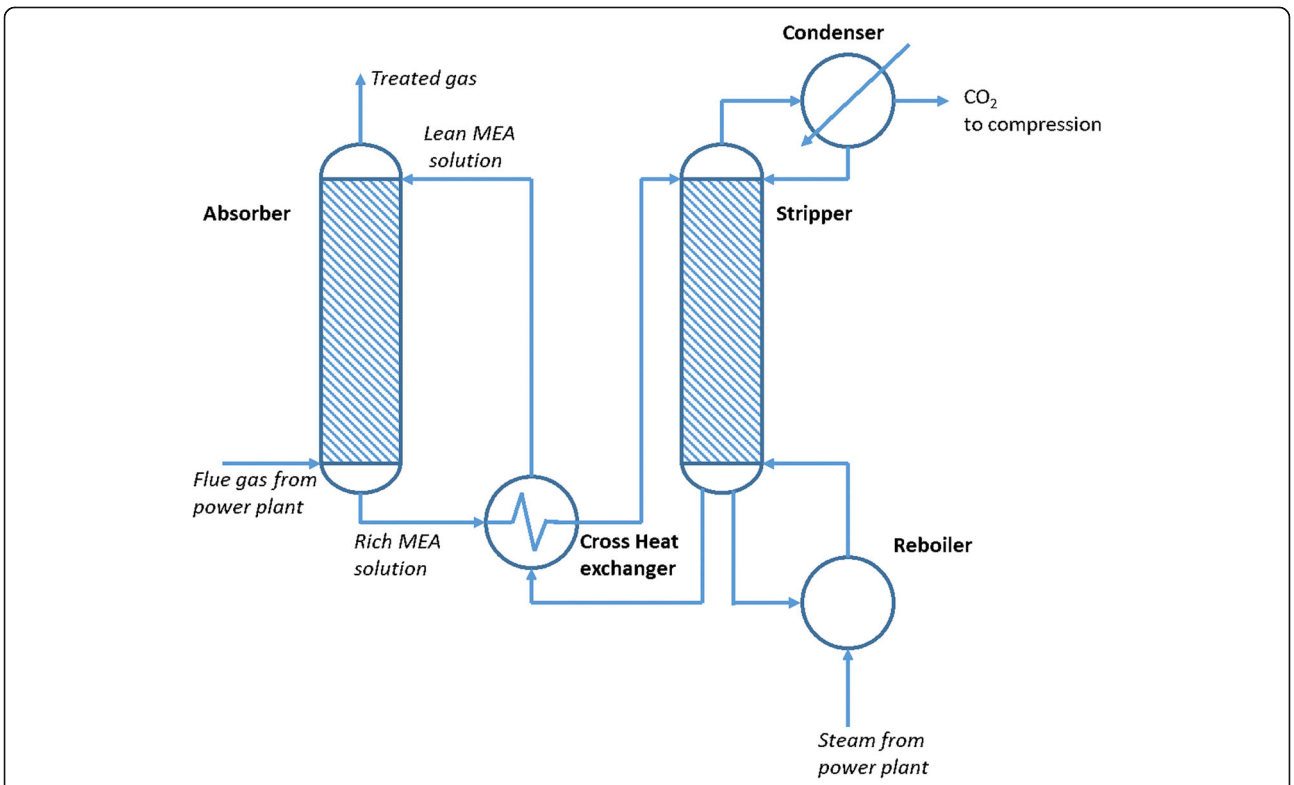

Fig. 6 Simplified schematic process flow of conventional post-combustion CO_2 capture using chemical absorption. Adapted from [26], Copyright (2015), with permission from Elsevier

and studied process [26]. It can handle large volumes of gas and can be easily retrofitted into an existing plant as end-pipe treatment [28]. In solvent based PCC, the absorber and stripper cover ~ 72% of the total equipment cost [29]. Using PI technologies could substantially improve the cost-efficiency of solvent-based PCC.

Enhancing mass transfer between liquid absorbent and CO_2 is essential in intensifying CC using chemical adsorption. Different strategies have been reported to enhance gas-liquid mass transfer, including the use of rotating spiral contactors [30], spinning disc reactors (SDRs) [31], micro-reactors [32] and rotating packed beds (RPB). In rotating spiral contacting, as the name suggests, the rotation of a spiral channel causes a centrifugal acceleration, forcing the gas and liquid to flow in parallel layers of uniform thickness. By controlling the spiral geometry and the operating conditions, it is possible to minimise the layer thickness and reduce the mass transfer resistances in the system. In SDRs, the same is achieved by flowing the liquid and the gas over a horizontal spinning disc. The centrifugal forces in the system lead to formation of thin films and high shear rates over the disc; grooved and corrugated disc surfaces can further passively enhance mixing and mass transfer by inducing turbulence within the film. Microreactors make use of micrometric channel diameters to create small diffusion distances where mass transfer resistances are minimised. Finally, in RPBs, a rotating bed filled with large surface area packing material creates a highly sheared thin film with enhanced gas-liquid contact and mass transfer rates. RPBs are discussed in more detail in Rotating packed bed absorbers section as these are the most extensively investigated intensification strategy at pilot scale due to its ability to handle large volumes of gas. Table 1 summarizes the CC technologies and possible intensification techniques.

Absorbent selection

The conventional alkanolamine based absorbents are corrosive and operate at high pressures, demanding costly corrosion resistant materials. Due to mass transer limitations, large equipment is required to treat vast volumes of flue gas [38]. Several researchers have reported the use of monoethanolamine (MEA) absorbent in carbon capture [34, 39, 40]. This requires high energy during regeneration [41] and reacts fast with CO_2 [42] compared to other solvents reported. Fast reactions are often mass transfer limited, even at molecular level, as molecules do not have enough time to diffuse before they react, leading to a micromixing controlled system [26]. Developing technologies with enhanced mass transfer capabilities is therefore necessary to address such restrictions.

The choice of optimum absorbent loading, concentration, and stripper operating conditions have also been shown to signficantly improve capture performance and reduce energy consumption in conventional packed columns [29, 43]. However, high MEA concentration imposes a greater corrosion risk, hence it is necessary to optimise this or develop less corrosive absorbents. Modifications of conventional absorbents through combination of common amines [44–46] and introduction of porous materials [47] have been reported to enhance CO_2 absorption. Combination of amines is aimed at harnessing the advantages of individual amine and supressing their disadavantages. For example, thermal and oxidative stability of MEA can be improved by introducing other amines (e.g. peperazine) with high resistance to oxidative and thermal degradation. Additionally, introduction of a porous packing material with a Lewis acid nature can suppress protonation of absorbent, prolonging its lifetime and performance.

The energy penalty during absorbent regeneration could significantly be reduced using novel absorbents, including biphasic absorbents, enzymatic-based and -encapsulated absorbents [36]. In particular, liquid-liquid biphasic absorbents separate into two immiscible liquid phases: a CO_2-rich phase and a lean phase, at high temperature or during CO_2 absorption. As only the CO_2-rich phase is sent to the stripper, this leads to process intensification by reducing the stripper size and energy consumption for regeneration [36]. This absorbent is classified as third generation absorbent and

Table 1 Intensification techniques in carbon capture

Technology	Contact medium	Intensification technique	Intensification Parameter	Ref.
Adsorption	Adsorbent	Novel adsorbent	CO_2 uptake	[33]
		MOFs	Selectivity	
Chemical absorption	Solvent	Rotating Packed Bed	Absorber and stripper size; Mass transfer	[34]
		Membrane Reactor	CO_2 permeance and selectivity	[35]
		Spinning Disc Reactor	Mass transfer; CO_2 absorption	[31]
		Microreactor	Mass transfer	[32]
		Novel Solvent	Energy efficiency; CO_2 absorption	[36]
		Thermal integration	Energy efficiency	[37]

consists of a mixture of amine dissolved in alcohol [48]. Ultrasound assisted stripping can enhance reactivity and mass transfer through cavitation [49]. This technology has the potential to reduce capital cost by intensifying absorbent regeneration. Ying et al. [50] have shown that ultrasound-assisted absorbent regeneration intensified the regeneration 3 times faster than using only heat and achieved 19% cost reduction and low lean amine solution of 0.2 mol/mol.

Rotating packed bed absorbers

When using amine-based solvents, which react rapidly with CO_2, mass transfer intensification has been demonstrated using RPB technologies (Fig. 7), generally referred to as HIGEE. This technology uses centrifugal fields to form highly sheared thin films where highly efficient gas-liquid mass transfer can occur [34], resulting in equipment volume reduction, improve efficiency [52] and safety owing to its high gas-liquid contact area and compactness. With its intense fluid dynamics and mass transfer, low absorbent concentrations can be used in RPBs to achieve similar performance to that of a large conventional absorber column. Chamchan et al. [53] observed that both the RPB and conventional packed bed absorbers demonstrated similar absorption performance and energy consumption in CO_2 capture at pilot scale but the RPB was associated with a 1/3 volume reduction compared to the conventional packed bed. In CC from flue gas with low CO_2 concentration, Xie et al. [52] demonstrated the RPB to be capable of achieving a mass transfer coefficient around 2.7x higher than in a packed column, with a corresponding 2.6x reduction in equipment volume. Mass transfer rates are greatly affected by gas-liquid contact area influenced by the packing type used in RPBs, with blades [39] and structured [54]. Over 4 times faster gas phase mass transfer at high liquid flow in RPB with blade packing compared to structured packing has been reported by Lin et al. [39]. This was ascribed to the formation of smaller liquid droplets and thin films on the blade packing leading to large gas-liquid interfacial area and fast dissolution as well as diffusion of CO_2 into MEA solution.

Stripper columns have also been intensified using RPBs. Cheng et al. [55] found that to achieve the same thermal regeneration efficiency in a conventional stripper and in an RPB, the size of the RPB was 10 times smaller than the conventional packed bed. Jassim et al. [34] demonstrated that to achieve the same performance as that of RPB, conventional stripper height and diameter have to be increased by 8.4 and 11.3 factor respectively. Table 2 highlights the reported extents of intensification achieved using RPBs for CC and absorbent regeneration. With the reduction in equipment size achieved by employing RPBs, cost savings

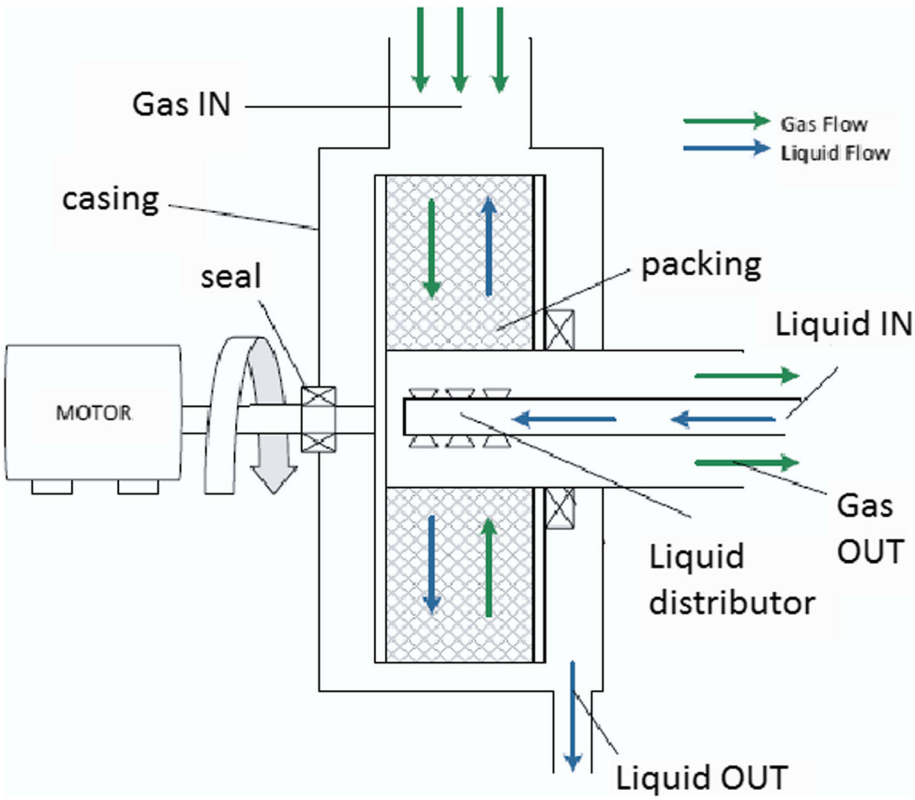

Fig. 7 Schematic of a counter-current flow RPB. Reprinted with permission from [51]. Copyright (2016) Royal Society of Chemistry

Table 2 Intensification achieved in RPB for CO_2 capture and absorbent regeneration compared to conventional packed columns

Absorbent	Conditions	PI Achieved	Ref.
MEA	20–40 °C 1 atm 600–1000 rpm	• Reduction in absorber height of transfer unit by almost 13 times • Enhanced mass transfer • Stripper volume reduction	[34]
DETA, PZ, DEG	27 °C 1 atm	• 96.8% height of transfer unit reduction	[45]
MEA, DETA, PZ	50 °C 1600 rpm	• 22.7% regeneration energy reduction using 40.8 wt% MEA. Different configuration of RPB with packed bed were investigated • 1.5 times RPB volume reduction to achieve similar performance with packed bed column • RPB in series have 10.5% lower regeneration energy and 20.6% higher treated gas compared to single RPB	[46]
Potassium sarcosine	20–70 °C 1 atm 600–1400 rpm	• 2.6 times reduction in packing volume • Possibility to treat large volume of flue gas within a smaller device • 29 times increase in volumetric mass transfer coefficient	[52]

DETA Diethylenetriamine, *PZ* Piperazine, *DEG* Diethylene glycol

can be envisaged. The capital cost for a CO_2 absorption process involving a more compact RPB (1.3 m diameter, 2.6 m length) has been estimated to be 30% lower than a large conventional packed bed column (1.37 m diameter, 13.7 m length) with the same performance [56]. Further reductions in capital cost of an RPB- based CO_2 capture process can be achieved by integrating several functionalities in one equipment e.g. integrating the reboiler within the RPB desorber unit. The overall cost of CO_2 capture in a power plant deploying an RPB-based process has been evaluated to be marginally lower at 61€/ton CO_2 vs. 65€/ton CO_2 for a conventional packed column process [57]. Clearly, there is scope for further decreases in operating costs in such PI based processes via greater integration of energy across the absorption and desorption cycles and reduced energy consumption in the reboiler for the desorption process [56].

Adsorption

Aside from liquid absorbents, solid adsorbents are also utilised to capture CO_2 from exhaust gas. Adsorption involves coupling of CO_2 with a solid adsorbent, which is subsequently regenerated upon heating or by processing at a reduced pressure to release the adsorbed CO_2. Although this type of approach is capital intensive and operates at a high pressure, high CO_2 removal can be achieved [58]. Nevertheless, common adsorbents such as activated carbon, carbon molecular sieves [59] and zeolites [60], among other common adsorbents, are associated with low CO_2 adsorption capacity especially at large scale processing [27].

Metal organic frameworks (MOFs) are emerging adsorbents that are characterized by high surface area, high porosity and tunable surface functionality [61, 62]. MOFs are composed of metal ions or metal cluster vertices joined by organic ligands resulting in a network. The metal ions and/or organic ligands can be altered to enhance CO_2 capture capacity and selectivity of MOFs [63]. Zheng et al. [33] developed an expanded 4,4-paddlewheel-connected porous MOF-505-type from nanosized rectangular diisophthalate linker containing alkyne groups with surface area of 3038 m^2/g. Under 20 bar, this material demonstrated a CO_2 uptake capacity of 23.83 and 19.85 mmol/g at 0 and 25 °C respectively, 74.5% higher than typical zeolite 5A at 14 bar [64]. There is growing research on ways to further improve the performance of MOFs for CC [61].

Biochemical capture

Indirect capture and sequestration of CO_2 through its consumption by photosynthetic microorganisms such as microalgae for bioenergy and biorenewable chemicals production is a promising technology for CO_2 valorisation. The photo-efficiency of these microorganisms is low, thus there is a technology development need in this area to improve their efficiency. Some of the conventional intensification technologies shown in Table 1 can be deployed in conjunction with more novel techniques of cell immobilisation such as in biocomposites where highly concentrated, living but non-growing microorganisms are incorporated within the structure of either non-porous substrates (polyesters, metals) or non-woven porous substrates (papers) [65–67].

Moreira et al. [68] evaluated the potential of using algal culture to capture CO_2 from atmosphere. Green algae like *Chlorella* sp. was reported to have been used to enrich the CH_4 content of biogas [69], while Cheng et al. [70] used *Chlorella vulgaris* to capture CO_2 in a photobioreactor. The integration of the photobioreactor with a membrane module has intensified the process, achieving 69% higher CO_2 fixation rate. The same microalgae was immobilised within a porous biocomposite paper to demonstrate the intensification potential of CO_2 biofixation in a spinning disc bioreactor (SDBR) [71]. High cell retention (> 99.5%) even under conditions of high shear and consistently high microalgae photoreactivity were recorded over a period of 15 h of continuous operation at 300 rpm

under illumination with cool white LED panels. The CO_2 biofixation in the SDBR was almost doubled that achieved in a 2 L flat panel suspended cells photobioreactor reported in the literature [72], with an almost 1000-fold processing volume reduction in the SDBR [71]. The low volume and therefore thin film formed over the photoreactive biocomposite paper in the SDBR allowed efficient light penetration to reach the high concentration of entrapped cells in this device. An assessment of a similar biocomposite-based bioreactor operated as a falling film reactor for a syn-gas processing highlight the promising process intensification potential of 66% smaller reactor volume and 96% lower power input requirement than a CSTR [73]. It is feasible to expect that such advantages can be replicated with a light-induced CO_2 absorption process using a microalgae biocomposite, leading to a more cost-effective process.

One important consideration of the biocomposite-integrated SDBR or falling film reactor for producing valuable chemicals or fuels is that the micro-organisms employed in the biocomposite structure should be able to secrete the products of interest into the surrounding medium for easy extraction and purification without disrupting the biocomposite. This may require genetic engineering of some bacterial species to make their wall permeable. One other limitation is the small throughput, which is imposed by the need for thin films in order to derive the benefits of improved gas-liquid mass transfer and light penetration efficiency. Processing at larger throughputs would require scaling out methods to be implemented such as multiple rotating discs or falling film tubes operating in parallel in a given unit.

Intensification of CO_2 chemical conversion

The captured CO_2 is most often stored underground in a process called geological sequestration [74], which involves pumping CO_2 into geological formations. The CO_2 is stored under pressure, enough to keep it as a supercritical fluid. Alternatively, the captured CO_2 is sunk under pressure deep below the ocean. In order to reduce our reliance on geological sequestration, and also the continuous extraction of more fossil fuels, it is of utmost importance to look for technologies that can convert the captured CO_2 to added-value fuels and products. Such technologies can either use CO_2 in a circular way, or can sequestrate the CO_2 in long duration materials, replacing chemicals and materials currently derived from fossil sources. CO_2 conversion processes have been exhaustively discussed in literature [7, 16, 19]. To achieve an optimum conversion while being cost effective and competitive with fossil-fuel production routes, process intensification is essential. In this section, the intensification aspects of photochemical, electrochemical, biochemical and thermochemical routes that have been developed in recent years have been reviewed. Table 3 summarizes some intensification techniques being used in photocatalytic, electrochemical and thermochemical CO_2 reduction.

Photocatalysis

The sun illuminates the Earth with a broad range of electromagnetic energy, primarily concentrated within the infrared and visible regions. This energy is utilised by plants to produce carbohydrates from CO_2 and water in the presence of chlorophyll (Fig. 8a). However, the overall efficiency of this process does not exceed 4.5% [83]. Photocatalytic processes taking place in the presence of an engineered photocatalyst, can be used to artificially mimic this process and reduce CO_2 into solar fuels with the potential to increase the overall process efficiency (Fig. 8b).

Photocatalysts are usually semiconductors with relatively low band gap energy compared to insulators. When exposed to electromagnetic radiation with energy equal to or higher than the band gap, the valence electrons in the materials are excited into the conduction band, creating positively charged holes in the valence band. The redox thermodynamics of a photocatalyst is determined by the electrode potentials of the generated charge carriers, while kinetic aspects are determined by

Table 3 CO_2 conversion methods with their possible intensification techniques

Process	Driving force	Main products	Intensification technique	Ref.
Photocatalytic reduction	Light	CH_3OH, CO, CH_4, H_2	• Photocatalyst modification	[75]
			• Monolith reactor	[76]
			• Micro-reactor	[77]
Electrochemical reduction	Electricity	CH_3OH, HCOOH	• Microfluidic device	[78]
			• Coupling with photocatalysis	[79]
			• Gas diffuse electrode	[80]
			• Membrane reactor	[81]
Thermochemical reduction	Heat	CO, H_2,	• Plasma	[20]
			• Reactive coupling	
			• Membrane reactor	[82]

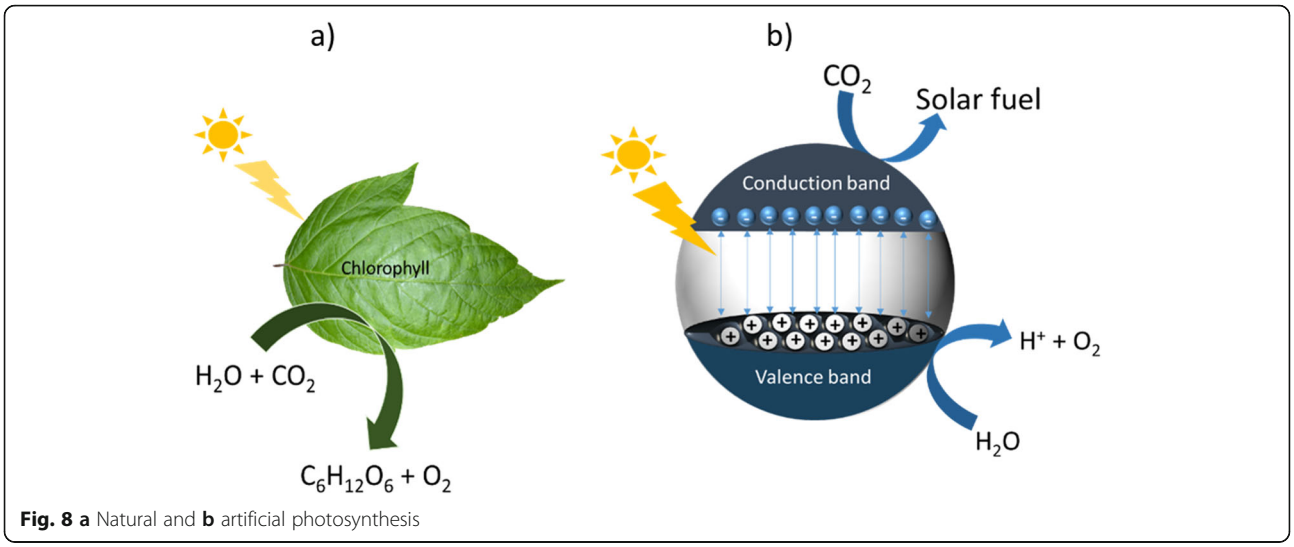

Fig. 8 **a** Natural and **b** artificial photosynthesis

not only the redox chemistry involved, but also by the light driven processes, including charge generation, migration and recombination.

Despite the promising future of this process, there are challenges associated with photocatalytic CO_2 reduction:

- Charge-carrier recombination: When the charge-carriers are generated, the redox kinetics competes with the kinetics of charge recombination. If the charge-carriers recombine too quickly, there is not enough time for the redox reactions to progress, and the light energy is dissipated as heat;
- Low selectivity: several factors including photon energy, conduction band edge, light intensity and photocatalyst active sites influence product selectivity [84];
- Low yield: the process is still far from practical application due to low yield arising from slow reactions and unfavourable thermodynamic equilibria;
- Light penetration: A high concentration of photocatalyst in suspension within the reactor prevents efficient light penetration into the medium due to light absorbed by the catalysts, leading to "dark" areas as the radiation is absorbed close to the light source. Optimisation of catalyst concentration is required to enhance kinetics both through catalysis and optimal light intensity distribution;
- Batch photoreactors with low surface area-volume-ratios are often used, leading to slow kinetics, and to mass and heat transfer limitations;
- Poor mixing efficiency in standard photoreactor systems.

Addressing these challenges through PI, with emphasis on photocatalyst and reactor design, has been reviewed and discussed below.

Photocatalysts

Improving photon absorption by photocatalysts remain one of the biggest challenges. Several strategies have been reported aiming to either reduce the band gap and shift the optical response from UV to visible, or to improve the stability of charge-carriers by minimising recombination. Some of the common and novel materials are reviewed here alongside the strategies being used to enhance their performance.

Even though a wide variety of photocatalysts have been used, such as Ta_3N_5 [85], Ga_2O_3 [86], ZnS [87], In_2O_3 [88], TiO_2 remains the most commonly used of all of them. The photocatalytic activity of TiO_2 was first discovered 90 years ago, but breakthrough was made when Fujishima and Honda used TiO_2 electrode to split water over 40 years ago [89–91]. The advantages of TiO_2 is that it is less toxic, low cost, stable [92] and simple to prepare [89]. However this material has low photocatalytic efficiency [89, 92] due its relatively high band gap (3.2 eV). As a result, TiO_2 requires UV radiation for activation, using a very small amount of the total solar radiation available [92]. It also suffers from low photocatalytic stability due to electron-hole recombination [93].

Doping is being used to enhance the performance of photocatalysts aiming to: reduce band gap energy, minimise charge carrier recombination, and/or increase the levels of surface-adsorbed species [94]. Doping is the process of modifying band structure of semiconductor by adding impurities [95]. In TiO_2, dopants can promote or inhibit anatase to rutile transformation by penetrating into the anatase lattice and change the level of oxygen vacancies [94].

Metals dopants such as e.g. V [85], Na [96], Ni [97] help decrease the electron-hole recombination phenomenon by trapping the excited electron and reducing the conduction band edge, thereby improving the visible light response of

the photocatalyst. Noble and transition metals are being used as dopants to improve the spectral response of TiO_2. Due to the cost of noble metals, transition metals are preferred. Selectivity and optical response of TiO_2 can be equally affected by non-metal doping. Upon doping with non-metals, an energy level is created above the valence band resulting in narrowing the band gap energy of TiO_2 [98]. Carbon, nitrogen, fluorine, sulphur and iodine have been reported to narrow the band gap energy of TiO_2 and improve the visible light response of TiO_2 [75, 99].

A synergetic effect of both metal and non-metal dopants can be utilized to improve the visible light response of TiO_2. Incorporating Fe and N in TiO_2 nanoparticles, Khalilzadeh et al. [98] were able to achieve a visible light response on gaseous CO_2 reduction. An increase of two orders of magnitude in methanol and methane yield was recorded upon doping TiO_2 nanoparticles with 0.12%Fe-0.5%N at the fixed pressure of 75.8 kPa and 15.5 kPa for CO_2 and H_2O respectively. The band gap energy of this photocatalyst was estimated to be 2.93 eV against the 3.19 eV of the pure TiO_2.

An alternative strategy is to employ semiconductors heterojunctions, which are designed by combining two or more semiconductors. This has been reported to be an effective way to facilitate charge carrier transfer and separation in semiconductors [100, 101], improving their performance as photocatalyst. Photocatalyst nanoparticles are reported to have enhanced optical response due to their high surface area and short charge carrier transport channels [102]. No redshift was reported for single nanostructured TiO_2, unless incorporated with dopant.

Another photocatalyst material that is currently being investigated is graphitic carbon nitride (g-C_3N_4). g-C_3N_4, a metal-free photocatalyst, is attractive due to its low cost, high light absorption, charge carrier stability [103], low toxicity [104], ease in preparation and easy coupling with other photocatalyst materials [105]. This material can further be enhanced to reduce its high chance of carrier charge recombination resulting from low band gap energy (2.8 eV). Compared to TiO_2, it requires visible radiation for activation. g-C_3N_4 can be prepared very simply by heating urea [103, 106] or melamine [105] to 550 °C in a muffle furnace. The high reduction potential of g-C_3N_4 can be related to its conduction band edge (− 1.13 eV). However, its valence band potential is insufficient to oxidise water, leading to lower yield of CO_2 photoreduction products when using it alone [107]. Usually, g-C_3N_4 is incorporated with TiO_2 to fully explore its advantages [108].

Photocatalytic reactors

Process intensification in photocatalytic reactors can be achieved through maximisation of the radiation intensity reaching all catalyst surfaces. It is possible to achieve this by intensification of the surface to volume ratio in the reactor while designing adequately illuminated surfaces to illuminate all exposed surfaces. Alternatively, mixing enhancement can play two intensification roles. On the one hand, mixing intensification increases the exposure of the catalyst to regions with high-light intensity, helping to overcome the effects of non-uniform light distribution present in most common light sources, and hence increasing the light usage by all catalyst particles [109]. On the other hand, mixing intensification leads to a reduction in mass and heat transfer resistances [110], especially in aqueous slurry system where catalyst particles tend to settle down and CO_2 solubility is low. The most common photoreactor types for CO_2 reduction are slurry, fixed bed, annular and surface coated reactors [111]. Slurry reactor types have low light penetration due to light scattering and absorption effect in particle suspended medium [112] and bear additional cost of separating catalyst particles [111]. In fixed bed reactors high photoactivity is achieved for plug flow regime, less pressure drop [112] that enable it to operate under reduced cost. However, this reactor type is mass and photon transfer limited as transfer of molecules within the coated catalyst is diffusion-limited. Alternative intensified reactors that can overcome some of these limitations are discussed below.

Monolith photoreactors Monolith photoreactors consist of a bundle of optical fibres in a honeycomb-like structure that transmits light into the reactor core and serves as photocatalyst support (Fig. 9). High photocatalytic efficiency and production yield in monolith photoreactors can be ascribed to their improved light distribution, large surface area to volume ratio, low pressure drop and high catalyst loading [113, 114]. The limitation here lies in the ineffective utilisation of reactor volume and hence thin fibres with relatively large surface area are often used [115]. Another interesting feature of this reactor type is that it does not require additional costs for downstream catalyst separation since the catalyst is immobilised by coating over the surface of the monoliths. However it is recommended that as thin a catalyst layer as possible is employed (no more than a few microns thickness) as thicker catalyst layers may result in significant product adsorption within the catalyst structure and can lead to catalyst deactivation [116]. It is also important to limit operation to a film flow regime in the monolith reactor in order to avoid problems arising from gas bubbles interfering with the uniformity of light distribution in the channels [116].

Although a light source with higher intensity than in conventional batch reactors was used in monolith reactor studies, Ola et al. [76] reported that the quantum efficiency in the monolith reactor was higher than that of the conventional batch reactor owing to its large

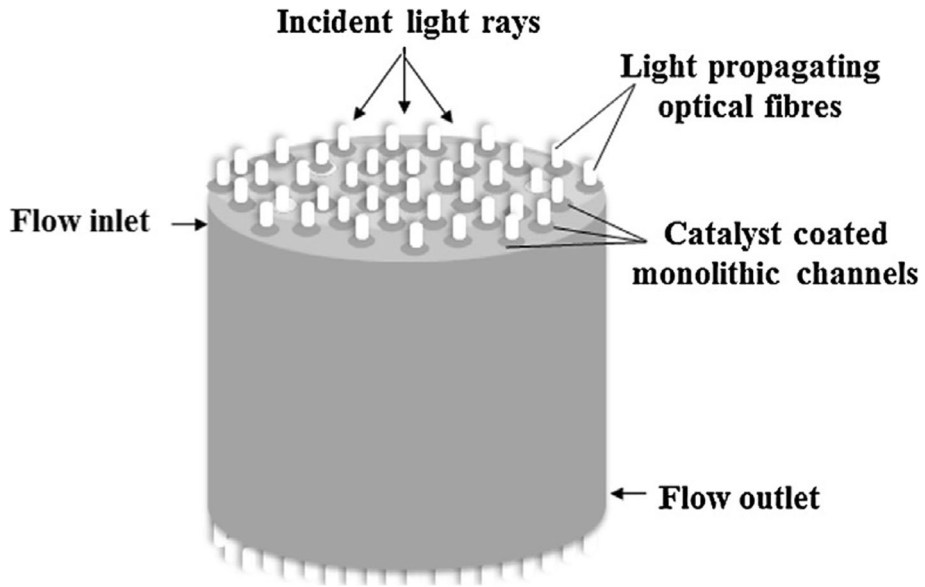

Fig. 9 Schematic of internally illuminated monolith reactor. Reprinted from [112], DOI: https://doi.org/10.1016/j.jphotochemrev.2015.06.001; licensed under the CC BY license

surface area and uniform light distribution. Monolith reactors have been shown to outperform conventional reactors owing to their high illuminated surface area (Table 4). Performance of monoliths can be further intensified by applying material intensification as discussed earlier on.

Microreactors Lokhat et al. [119] defined microreactors as "*reactors with reaction channels of the order of micrometers, at which diffusion is the dominant mixing mechanism rather than turbulent eddies.*" The small size nature of microreactor operating in continuous regime have made it promising in intensification of thermally driven [120] and photocatalytic reactions [121], among other applications. In photocatalytic reactions, in addition to overcoming thermal and mass diffusion limitations, in applications where light distribution determines the yield [122], microreactors allow for uniform light distribution owing to its small size, short optical paths and large surface area to volume ratio [123]. Given the high photon density in micro-reactors, it is clear that short reaction times are needed compared to conventional large scale vessels [122]. Energy consumption and light efficiency can be further improved using low power LED light sources, which offer a high and unidirectional radiant flux. Additionally, reaction parameters such as flow rate, temperature and pressure can easily be adjusted and fine-tuned in microreactors operating in continuous regime, unlike batch reactors where some parameter can only be adjusted after the end of each run consuming much time.

Photocatalytic reduction of CO_2 in aqueous phase is usually conducted at high pressure to increase the solubility of CO_2 in H_2O, which can pose safety concerns. Due to the small size of microreactors and short diffusion resistances, operation pressure and safety concerns are minimised. Another advantage of microreactors is the easy integration with micro-mixers leading to improvement of mixing efficiency and reduction in premixing requirements. Taking into account the limitations of photoreduction of CO_2 associated with mass transfer and light distribution, and the advantages of microreactors highlighted, it is clear that microreactor technology has the potential to address those limitations.

The drawback of microreactor processing is scaling up. Although a numbering up approach can be implemented

Table 4 Performance for CO_2 reduction monolith photoreactors compared to batch photoreactors

Medium	Catalyst	Light intensity, mW/cm^2	Main Products	Intensified parameter	Ref.
Aqueous	Pd/Rh doped TiO_2 nanoparticles	41.62	CH_3OH HCHO	95.7% higher quantum efficiency	[76]
Gaseous	V and W doped TiO_2	44.6	CO CH_4 H_2	57.8% higher quantum efficiency compared to Cu/TiO_2 in conventional reactor	[117]
Gaseous	TiO_2	150	CH_4	91% higher quantum efficiency	[118]

to increase processing throughput, the challenge of incorporating uniform illumination in such systems is far from trivial and much research is still needed in this area [109]. Moreover, in multiphasic reactions involving precipitation or suspended catalyst particles, the potential for channel blockage is a disadvantage in microreactors. Strategies have been developed to overcome such problems. Delacour et al. [124] introduced pulsed ultrasound to prevent blockage of micro-channels by solid particles. Alternatively, photocatalysts can be immobilised onto the micro-channel walls. There have been many reports of the application of immobilised photocatalysts for abatement of organic pollutants [125–130], however there is room for many more studies on CO_2 photoreduction.

Electrochemical conversion

In electrochemical conversion methods, the electrons discharged during the redox reactions at the surface of electrodes drive the electrochemical reduction of CO_2 in H_2O to a plethora of chemicals and fuels, the most common being carbon monoxide, formic acid and methanol [131]. The reaction between CO_2 and H_2O occurs into two half-cells, with H_2O oxidation taking place at the anode and CO_2 reduction at the cathode [132]. Thermodynamically, water oxidation (reaction 1) takes place at the potential of 1.23 V while CO_2 reduction (reaction 2) takes place at near 0 V, making the reduction half reaction difficult [133]. Applying an overpotential can force the reaction to proceed forward, although making the reaction less energy efficient. A number of products are formed in this process because of the multi-electron transfer imposing difficulties in controlling the selectivity of the process [134]. Also a competing reaction leading to hydrogen evolution takes place at 0 V leading to Faradic efficiency reduction [135] and low catalyst stability.

$$H_2O \rightarrow 4H^+ + O_2 + 4e^- \quad (1)$$

$$aCO_2 + bH^+ + be^- \rightarrow C_aH_{b-2}O_{2a-1} + H_2O \quad (2)$$

Novel and highly stable electrocatalysts are currently being developed and tested; these include quantum dot [136], carbon nanostructure-based [137] electrocatalysts, among others. More investigations are needed to quantify the intensification level of these potential materials.

Electrochemical reactors

Prior to the development of novel electrochemical reactors, traditional fuel cell-based reactor designs with the electrodes separated by a polymer membrane were commonly studied [78]. Mass transfer limitation between gas-liquid-solid phases and interphases in electrochemical CO_2 reduction can be reduced using gas diffusion electrodes [80, 81]. Introducing polytetrafluoroethylene (PTFE) into gas diffuse electrode with Nafion bonded Sn catalyst layer in electrochemical CO_2 reduction systems by Wang et al. [138] showed an enhancement of the Faraday efficiency by 25.4%, which resulted from the increase in the catalyst active surface area. Jimenez et al. [80] investigated the effect of current density, temperature, CO_2 flow rate and anolyte concentration on the selectivity of CO_2 reduction in gaseous phase on Pt nanoparticles deposited on carbon nanotubes (CNT) using a Proton Exchange Membrane (PEM) cell. Increasing temperature to 80 °C was shown to favour formic acid and methanol production presumably due to increase in proton transfer though the PEM and diffusivity of liquid products. Methane and methanol were produced at low CO_2 flow rate. In contrast to this, under similar conditions but on Pb nanoparticles on CNT, Garcia et al. [139] observed that low temperatures favoured formic acid, and neither temperature nor CO_2 flow rate favoured methane production. However, methanol selectivity remained the same as on Pt/CNT.

Further enhancement of mass transfer can be achieved using microfluidic devices due to their large surface area [140]. Microfluidic electrochemical reactors are flexible because of the easy control of reaction parameters, such as residence time, pressure and temperature, using a single reactor set up. This means that evaluation of the effects of operating conditions can be easily studied, being possible to scan through different operating conditions without disassembling the reactor set up and with minimal downtimes between experiments. Lu et al. [141] established the optimum microfluidic channel thickness, electrolyte flow rate and catalyst composition ratio for an enhanced performance of membraneless microfluidic reactor with dual electrolyte for CO_2 reduction. 94.7% Faradaic efficiency was achieved at 100 μm channel thickness and above 50 ml/min flow rate.

Co-electrolysis of CO_2 and H_2O vapour can be conducted in solid oxide cell (SOC) to generate electricity and produce syngas with high conversion and selectivity [16]. The SOC converts electrical energy to chemicals when operating in electrolytic mode, and vice versa in fuel cell mode [142]. The SOC operates at an elevated temperature (≥800 °C) [16], and suffers from concentration polarisation and degradation of electrode [143]. Call et al. [144] have studied the use of fluidic oscillators to disrupt gas flow boundary layers and intensify mass transfer, leading to the development of a highly energy efficient system. The same researchers have also attempted to couple plasma with the SOC to improve energy efficiency of CO_2 activation while fluidic oscillation reduce concentration polarisation leading to an enhanced mass transfer.

The CO_2 reduction in electrochemical systems can be cost-effective depending on the chemical targeted and prevailing economic conditions. For instance, Jouny and

co-workers have demonstrated that CO_2 conversion to CO and formic acid can be highly profitable in such systems provided key electrocatalytic performance targets such as high selectivity and overpotential are met [145]. PI approaches can be directed at improving these desirable performance characteristics as highlighted above and thus making electrochemical processes more economically viable especially at large scales. The additional energy input often associated with active PI techniques such as fluid oscillations, centrifugal processing etc. should nevertheless be included in any economic analysis for a realistic assessment of operating costs and profitability.

Currently the main drawback of electrochemical CO_2 conversion in general is the lack of studies demonstrating the capabilities of the technology at scales large enough for industrial implementation [131]. Implementing PI techniques such as operating in microfluidic channels for instance may exacerbate these scale-up challenges, although the potential for scaling out (i.e operating in a large number of parallel channels) which have been demonstrated for other processes may provide a possible solution.

Plasma conversion

The high-cost thermal energy required for CO_2 reduction through thermochemical routes can be by-passed using non-thermal plasma technology generated through electric discharge. Non-thermal plasma, otherwise known as 'cold plasma', is characterised by high average energetic electrons (1–10 eV) with an average temperature of $10^4 - 10^5$ K [146] while the gas temperature remains near ambient. Compared to thermal plasma where operating temperatures can reach over 1000 K [147], non-thermal plasma is significantly more energy efficient and therefore more cost-effective as an energy source. Using non-thermal plasma to activate catalysts can facilitate thermodynamically uphill reactions [148] leading to an increased yield and selectivity at ambient temperature and atmospheric pressure avoiding catalyst sintering [149]. The synergetic effect of plasma and catalysts is shown in Fig. 10. Although the interaction between plasma and catalyst surface is not always clearly understood [151], apart from operating at low temperature, it enables quick start-up and shutdown cycles. A typical Dielectric Barrier Discharge (DBD) set up is shown in Fig. 11.

Zeng et al. [20] reported an increase in conversion of CO_2 hydrogenation by more than 30% upon incorporating plasma with Ni/Al_2O_3 catalyst in relation to plasma only at 4:1 H_2 to CO_2 ratio and 30 W discharge power in DBD reactor. The electron temperature of plasma can be increased by reduction of the discharge gap upon addition of packing material [146]. For wider discharge gaps in industrial applications, a filamentary discharge is typically observed. Filamentary discharges are transient and occur due to increase in the insulation medium (gas) between the electrodes which leads to ionization of the gas, being possible to visually observe the filaments arcs formed. The addition of a packing materials with a high dielectric constant can transform the discharge nature as a result of a decrease in discharge gap. This can lead to a significant enhancement of discharge characteristics. For example, Mei at al [153]. introduced $BaTO_3$ and glass beads packing materials into DBD reactor, observing a transition to a mixture of surface and filamentary discharge due to decrease in discharge gap. They also observed that the average electric field and mean electron energy increased.

Selecting a packing material with appropriate size can lead to energy efficiency and conversion improvements.

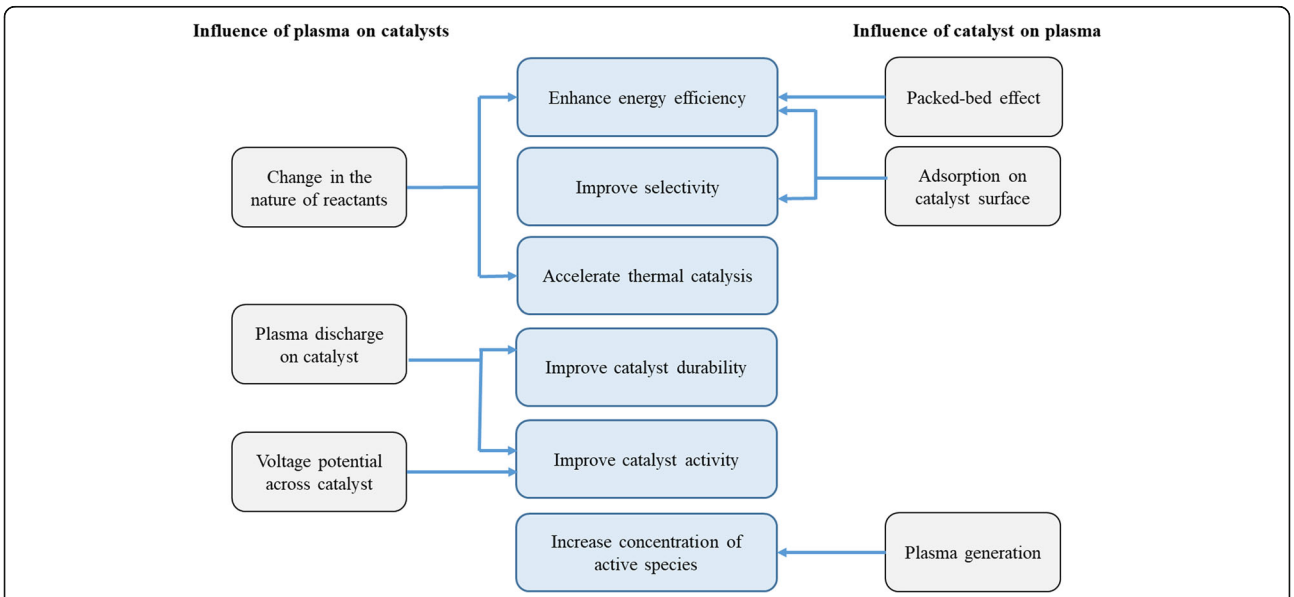

Fig. 10 Influence of plasma and catalysts on each other. Adapted from [150], Copyright (2009), with permission from American Chemical Society

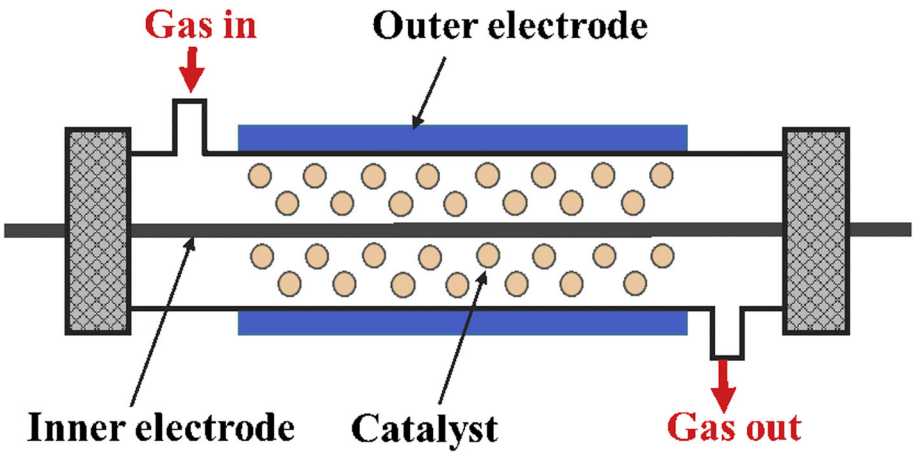

Fig. 11 Plasma assisted catalytic DBD reactor set up. Reprinted from [152], Copyright (2016), with permission from Elsevier

In plasma reactors, for the same packing material, large particles with small surface area are characterised by a high energy density, leading to high conversion. However, too large particles, will often create large gaps resulting in filamentary discharges. Therefore, an optimum packing size should be established to achieve maximum intensification in the process. Michielsen et al. [151] investigated the influence of SiO_2, ZrO_2, Al_2O_3 and $BaTiO_3$ dielectric beads sizes on conversion and energy efficiency of CO_2 decomposition in packed bed DBD reactor. At different sizes of $BaTiO_3$, a high conversion was recorded compared to without packing material. As the bead sizes of Al_2O_3 and $BaTiO_3$ increase, conversion and energy efficiency increase.

Fluidised [154], tubular DBD and coaxial dielectric packed-bed [146] plasma reactors are some of the reactors used for CO_2 decomposition/conversion. These reactors can be set up in different configurations. Zhang et al. [146] investigated the influence of reactor configuration on CO_2 decomposition using a coaxial DBD reactor packed with 3 mm glass and $BaTiO_3$ beads. The reactor consists of 2 concentric quartz tubes and it was configured in so that the stainless steel mesh electrodes used were either exposed to or insulated from the plasma activated species, thus affecting the electrical characteristics of the plasma reactor. It was observed that the highest conversion at the highest plasma power achieved when the electrode was insulated almost equals the conversion at low power when exposed to the plasma activated species. This increase in conversion was related to possible interaction between the electrode and plasma. Despite the promising future of plasma technology, it suffers from low overall energy efficiency [146] and selectivity [20].

Intensification using membrane separators and reactors

In a system involving gas-liquid, liquid-liquid and gas-liquid-solid where usually mass transfer resistances are high, a membrane contactor can be used to maximise the mass transfer rate without dispersion of one phase into the other [155]. Most membrane processes are driven by pressure difference that require less energy compared to thermal processes, making the overall processes high energy efficient. The membrane is characterised by high level of compactness, ability to address thermodynamic limitations [156], high contact area [157] owing to drastic reduction in the size of the unit [158] at the expense however of generally high membrane cost. This technology has been employed for carbon capture [159], in photochemical [160, 161], electrochemical [162], and thermochemical [82] CO_2 conversion processes aiming to overcome mass transfer resistance and enhance energy efficiency. With multifunctional units such as these membrane-integrated reactors, combining two functions into one unit should reduce the capital cost of the single unit compared to the individual reactor and membrane separation unit [163]. However, this technology suffers from limitations which include operating under high pressure [58], high membrane cost, cathode flooding, fuel crossover, membrane degradation in electrochemical systems [141].

Membrane-based gas absorption integrates both chemical absorption and separation [164]. Scholes et al. [22] have reviewed different types of selective membranes that can maximise the energy efficiency of pre-combustion carbon capture. A liquid mass transfer coefficient increase of 5 times when compared to a conventional packed column for CO_2 absorption in water at superficial liquid velocity of 1.25 cm/s has been reported [165], hence it is classified as one of the promising process intensification strategy [166]. Intensification of the CO_2 stripper by reducing the energy penalty can also be achieved by using membrane contactor [158].

Conclusions and outlook

This review provides an overview of the current progress in process intensification for carbon capture and conversion.

In terms of carbon capture, RPBs have been shown to give a significant reduction in absorber size and impovements in gas-liquid mass transfer efficiency in CO_2 capture via chemical absorption, which together with the development of more efficient chemical absorbents, represents a promising technology for CO_2 capture and sequestration.

The intensification of carbon capture and utilisation technologies have focused on developments relating to photochemical, electrochemical, thermochemical and biochemical routes. In photochemical process intensification, microreactors, monolith reactors and development of novel materials, such as graphitic carbon nitride, are approaches being investigated to intensify photocatalytic CO_2 reduction. Gas-diffusion electrodes, ion exhange membranes, microfluidic devices, as well as the development of highly stable electrocatalysts, are leading the way in improving Faradaic efficiency, current density and selectivity in electrochemical CO_2 conversion. There is also a growing number of research studies focused on replacing cost intensive thermal energy sources with cold plasma for catalyst activation in thermochemical CO_2 conversion. The development of biocomposite structures applied to intensfied reactor technologies offers one promising pathway of intensifying CO_2 capture and potentially conversion via biochemical routes.

Despite the progress achieved to date, there is great need for further research to be conducted to increase the technical and economic feasibility of many of the technologies highlighted. In CO_2 capture for example, intensification of the reboiler unit, development of novel absorbents, and further reduction in energy penalties in the stripper for absorbent recovery, deserve further investigation and development. In catalytic CO_2 conversion processes, there is limited understanding of mechanism of interaction between CO_2 and catalyst surface and more fundamental research is warranted in this area in order to arrive at optimal catalyst designs, particularly through the use of materials that can couple efficiently with the kinetic and thermodynamic requirements of CO_2 reduction. Intensification of radiation fields, products separation and mass transfer in photoreactors are also crucial. Finally, despite the recent advancement in plasma assisted catalysis, interaction between plasma and catalyst is yet to be understood, particularly to determine how catalysts and reaction conditions can be used to control selectivity and product synthesis routes. The energy efficiency of plasma technology still remains low and also needs addressing. Further development of this promising technology is much needed to realise its full potential in its application to CO_2 conversion.

Abbreviations

CC: Carbon capture; CNT: Carbon nanotubes; DBD: Dielectric Barrier Discharge; DEG: Diethylene glycol; DETA: Diethylenetriamine; MEA: Monoethanolamine; MOFs: Metal Organic Frameworks; PCC: Post-Combustion Capture; PEM: Proton Exchange Membrane; PI: Process Intensification; PTFE: Polytetrafluoroethylene; PZ: Piperazine; RPB: Rotating packed bed; SDBR: Spinning Disc Bioreactor; SDR: Spinning disc reactor; SOC: Solid oxide cell

Acknowledgements

Not applicable.

Authors' contributions

AA, FRA and KB made significant contribution in developing the ideas and structure of the manuscript. AA drafted the manuscript, while FRA and KB revised and finalised it. All the authors read and approved the final manuscript.

Funding

AA would like to acknowledge the Petroleum Technology Development Fund (PTDF), Nigeria, for funding his postgraduate studies at Newcastle University.

Availability of data and materials

Not applicable.

Competing interests

The authors declare that they have no competing interests.

Received: 24 October 2019 Accepted: 27 December 2019
Published online: 07 January 2020

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