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# Microfluidic-based chemical absorption technology for CO<sub>2</sub> capture: Mass transfer dynamics, operating factors and performance intensification

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## Abstract

Carbon capture, utilization, and storage (CCUS) is a crucial strategy for achieving  $CO_2$  emission reduction targets and mitigating the impacts of global warming and climate change. Among various CCUS technologies, chemical absorption of  $CO_2$  has proven to be a mature and widely-used technique in various industrial sectors. However, the current  $CO_2$  chemical absorption process involves large-scale equipment with low efficiencies, making it difficult to control. To address these issues, microfluidic devices have emerged as a promising technology to intensify the  $CO_2$  absorption process by providing a smaller required volume, enhanced mass transfer, cleaner and safer operations, higher productivity, and more efficient energy use.

This paper aims at presenting a comprehensive literature review on research advances of the microfluidic technology for CO<sub>2</sub> chemical absorption. The review covers various aspects, including microchannel geometries, two-phase flow patterns, mass transfer models, effects of operating factors, and measures to intensify the CO<sub>2</sub> absorption process. In addition, the paper discusses the measurement of interfacial and local parameters, such as liquid film thickness, velocity field, and local CO<sub>2</sub> concentration, which are primordial for understanding the transport phenomena and for optimizing the CO<sub>2</sub> absorbers. This paper may serve as an essential reference that contributes to the development and exploitation of highly-efficient microfluidic-based CO<sub>2</sub> chemical absorption technology for future large-scale industrial applications

## **Graphical abstract**



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## Highlights

- Recent researches on microfluidic-based CO<sub>2</sub> chemical absorption are reviewed.
- Absorption performance is determined by the mass transfer between CO<sub>2</sub> and absorbent.
- Effects of influencing operational factors are discussed.
- Optical techniques are frequently used to measure local & interfacial parameters.
- Different techniques used to intensify the CO<sub>2</sub> absorption process are surveyed.

## Keywords:

Carbon capture; CO<sub>2</sub> chemical absorption; Microchannel; Gas-liquid two-phase flow; Mass transfer enhancement; Emission targets

## Nomenclature

Latin letters			Abbreviations			
а	Specific surface area, 1/m	CCUS	Carbon Capture, Utilization and Storage			
Α	Liquid – gas contact area, m²	CFD	Computational fluid dynamics			
С	Concentration, mol/m <sup>3</sup>	ECBM	Enhanced Coal Bed Methane			
C <sub>e</sub>	CO <sub>2</sub> equilibrium concentration, mol/m <sup>3</sup>	IGCC	Integrated gasification combined cycle			
$d_h$	Hydraulic diameter, m	IL	Ionic liquids			
D <sub>diff</sub>	Diffusion coefficient, m <sup>2</sup> /s	LFT	Liquid film thickness			
E	Enhancement number	LIF	Laser induce fluorescence			
g	Gravitational acceleration, m/s <sup>2</sup>	LFDM	Laser focus displacement meter			
He	Henry constant, Pa.m <sup>3</sup> /mol,	М	mol/L			
h	Microchannel height, m	Mn-PTV	Nano-particle tracking velocimetry			
i	Amount of bubble in microchannel	PIV	Particle image velocimetry			
K <sub>G</sub>	Overall gas phase based mass transfer coefficient, kmol /kPa.m <sup>2</sup> .h	PTV	Particle tracking velocimetry			
$k_G$	Gas side mass-transfer coefficient, m/s	TPMS	Triply periodic minimum surface			
$k_L$	Liquid side mass transfer coefficient, m/s	VOF	Volume of fluid			
$k_L^0$	Physical mass transfer coefficient, m/s	AEEA	Aminoethylethanolamine			
k <sub>ov</sub>	Overall chemical reaction rate, 1/s	AMP	Aminomethylpropanol			
L	Length, m	BDA	1,4 butanediamine			
n	Molar flow rate, kmol/h	CO <sub>2</sub>	Carbon dioxde			
$n_i$	Fitting constants	CH <sub>4</sub>	Methane			
N	Molar flux, kmol/m².h	ChCl	Choline chloride			
p	Absorption pressure, kPa	DEA	Diethanolamine			
$\Delta p_F$	Friction pressure drop, kPa	DAB	1,4-diamidiniumbenzene			
Q	Volumetric flow rate, m <sup>3</sup> /s	DEEA	Diethylaminoethanol			
R	Gas constant, J/( K.mol)	DMEA	Dimethylethanolamine			
r	Radius of bubble in rectangular corner, m	EG	Ethylene glycol			
S -	Channel cross-section area,m <sup>2</sup>	HMDA	Hexanediamine			
	Experimental temperature, K		Polyethylene glycol dimethylether			
0	Local flow velocity, m/s	MEA	Monoethanolamine			
V	volume, m <sup>3</sup>	MDEA	2-(Dimethylamino)ethanol			
w	Microchannel width, m	ΡZ	Piperazine			
У	Molar fraction	TEA	Triethanolamine			
$Y_{CO_2}$	CO <sub>2</sub> loading rate, mol/mol	SDS	Sodium dodecyl sulfate			
z	Relative position in flow direction, m					
		<b>_</b>				
Greek	etters	Dimension	liess numbers			
$\eta_{CO_2}$		Ca	Capillary number, $Ca = \mu U/\sigma$			
δ	Liquid film thickness, m	Da	Damköhler number, $Da = k_{ov} \cdot d_h^2 / D$			
τ	Gas-liquid contact time, s	Re	Reynolds number, $Re = \rho U d_h / \mu$			
ρ	Fluid density, kg/m <sup>2</sup>	vve	Weber number, $We = \rho U^2 d_h / \sigma$			
μ	Dynamic viscosity, Pa.s	ScL	Schmidt number, $Sc_L = \mu/\rho D_{CO_2}$			
σ	I wo-phase interface tension, N/m	Sh <sub>L</sub>	Sherwood number, $Sh_L = k_L d_h / D_{CO_2}$			
β	Dimensionless structure parameter	Во	Bond numbers, $Bo = \Delta \rho g d_h^2 / \sigma$			
Subscri	ipts/superscripts					
G	Gas	b	Bubble			
L	Liquid	e	Equilibrium			
in	Inlet	F	Friction			
out	Outlet	TP	Two-phase			
с	Channel	0	Initial state			
L						

## 1. Introduction

The rapid development of the world economy has been at the large expense of fossil energy resources, resulting in the deterioration of ecology and environment. Researches[1,2] show that the main reason for the global warming during the past 50 years is related to the increased emission of greenhouse gases (GHGs) originated from human activities, such as industrial production, transportation, electricity generation, and homes & commercial businesses.  $CO_2$  is the main GHG and its global monthly mean concentration has increased rapidly from 340 ppm in year 1980 to around 419 ppm in July 2022[3]. The disastrous consequences of global warming due to excessive ( $CO_2$ ) emissions have been well-recognized, including drought, flood and extreme storm[4]. Since then, the carbon reduction has become an international consensus[5], evidenced by the signing of the Paris Agreement in year 2015[6]. In order to reach the 2050 carbon neutrality (net-zero) target [7], significantly increasing efforts have been devoted to the development and exploitation of Carbon Capture, Utilization and Storage (CCUS)-related technologies (cf. Fig.1) to balance the  $CO_2$  emissions with its removal[8,9].

As the basic and indispensable step of CCUS process, the development of efficient and low energyconsuming  $CO_2$  capture technologies has long become the research focus<sup>[5]</sup>. Conventional  $CO_2$  capture methods include chemical absorption, physical absorption or adsorption, membrane separation, cryogenic distillation, biological assimilation and others<sup>[10,11]</sup>. Their basic operation principles, advantages and disadvantages are summarized and compared in Table 1.

Among these CO<sub>2</sub> capture technologies, the cryogenic distillation is featured by the maturity and high capture efficiency, but involving high equipment investment and intensive energy consumption. The biological assimilation is much natural, environment-friendly and low energy input. But the slow process and limited scalability hinder its large-scale industrial application. The capture efficiency of physical adsorption and membrane separation depends strongly on the physical properties of material, and usually the recycling of these materials is a real problem. Compared with the above-mentioned methods, the chemical absorption seems to be the most adapted and widely used one for large-scale industrial applications[12,13]. This method uses chemical solvent(s) to react with the CO<sub>2</sub>; the absorbed gas could then be released by heating the product for the downstream processing (storage or utilization). It is a mature technology featured by high  $CO_2$  selectivity, low solvent consumption and high cost-effectiveness. Therefore, it has covered the CO<sub>2</sub> capture market of coal-fired power plants accounting for 60% of the global capacity[14]. Over the past few decades, the large-scale absorption equipment such as absorption tower has played a very important role in industry  $CO_2$  capture, featured by many advantages such as simple process control and stable equipment operation. However, the use of such large-scale equipment inevitably brings problems like high investment costs, excessive amount of solvent needed, high energy consumption, large occupied space and additional security measures, thereby hindering its further market development. One of the promising solutions to solve the above-mentioned drawbacks is through the process miniaturization, i.e., by the employment and implementation of microfluidic devices.

Microfluidic-based CO<sub>2</sub> capture is a technology developed to control and manipulate the CO<sub>2</sub> (gas) absorption process by a chemical solvent (liquid) in miniaturized absorbers/reactors with channel characteristic size in the order of micro or millimeter. Numerous studies have shown that the gas-liquid two-phase flow in microchannel reactors, when properly configured and handled, could exhibit numerous interesting features, including enhanced mass transfer due to high volumetric interfacial surface area, better repeatability and stability, safe manipulation with low leakage risk, and easy upscaling by the numbering-up[15,16]. The microfluidic technology has already been applied in many industrial fields, including medicine and pharmaceutical analysis[17–19]. Using microchannel reactors for CO<sub>2</sub> chemical absorption is also considered as a promising alternative technology pathway of macro-sized conventional equipment.

#### Table 1. Summary of conventional CO<sub>2</sub> capture technologies.

Technology	Definition & Operation principle	Advantages	Disadvantages	Application fields	Performance indicators*	Refs.
Chemical absorption	Using a solvent to absorb CO <sub>2</sub> from flue gas or other industrial emissions	<ul> <li>High contact surface area, high ca efficiency and selectivity;</li> <li>Mature technology;</li> <li>Low investment cost;</li> <li>Low carbon leakage;</li> <li>Standardized and easy process co</li> </ul>	pture - Corrosive; - High energy consumption; - Chemical solution post- treatment.	Large-scale coal-fired power plants and other fossil energy based industries	$\eta_{CO_2}$ >90%; Energy consumption: 1-3 GJ/t(CO <sub>2</sub> ); Cost: 10-50 \$/t(CO <sub>2</sub> ).	[20,21]
Physical absorption	Dissolving CO <sub>2</sub> in a liquid at high pressure and low temperature	<ul> <li>Cheap absorbent;</li> <li>Mature technology;</li> <li>High security.</li> </ul>	- Low selectivity; -Slow process; - Extra high energy consumption.	Large-scale energy conversion stations, such as integrated gasification combined cycle (IGCC) power plants, natural gas processing, coal chemical industry, etc.	η <sub>CO2</sub> >90%	[22]
Physical adsorption	Using solid material to adsorb CO <sub>2</sub> from flue gas or other industrial emissions by intermolecular forces	<ul> <li>High contact area, high capture efficiency;</li> <li>Mature technology;</li> <li>High security;</li> <li>Simple process.</li> </ul>	<ul> <li>Low selectivity;</li> <li>Low carbon loading rate of solid adsorbent;</li> <li>Low adsorption flux;</li> <li>Low material recovery;</li> <li>Energy-intensive carbon separation.</li> </ul>	Ammonia synthesis, hydrogen production, natural gas treatment, etc.	η <sub>CO2</sub> >85%	[23,24]
Membrane separation	Using a membrane to selectively separate CO <sub>2</sub> from flue gas or other industrial emissions	<ul> <li>High selectivity;</li> <li>High CO<sub>2</sub> purity;</li> <li>Low energy consumption.</li> </ul>	<ul> <li>High processing cost;</li> <li>Recycling of membranes;</li> <li>Larger flux resistance.</li> </ul>	Small waste heat recovery plant, biomass power plant	η <sub>CO2</sub> >90%; - Energy consumption: 0.01- 0.04 GJ/t(CO <sub>2</sub> ).	[25,26]
Cryogenic distillation	Separating CO <sub>2</sub> from flue gas or other industrial emissions using low- temperature distillation	<ul> <li>Mature technology;</li> <li>High selectivity;</li> <li>Low CO<sub>2</sub> transport cost.</li> </ul>	- Large-scale equipment investment; - Intensive energy consumption to maintain the low temperature.	IGCC power plants	η <sub>CO2</sub> >98%; Energy consumption: 2.4 GJ/t(CO <sub>2</sub> ).	[27,28]
Biological assimilation	Using plants and other photosynthetic organisms to absorb CO <sub>2</sub> and convert it into organic compounds.	-Low energy requirements; -Natural process; -Potential for carbon sequestration.	-Limited scalability; -Slow process; -Strong environmental dependence.	Biofuel production, soil pollution bioremediation	Absorption efficiency depends on environment and biological species; No requirement of external energy inputs	[29– 31]



Figure 1 CCUS flow chart and related technologies. (Data of CO<sub>2</sub> source from [1])

The microfluidic-based CO<sub>2</sub> absorption has long been the hot spot of the CCUS research community. In literature, a great number of researches have been focused on developing adapted absorbents[32–34] with expected features such as high CO<sub>2</sub> selectivity, stable, low corrosive, low toxicity and non-flammable. The two-phase flow patterns in microchannel and suitable working conditions for CO<sub>2</sub> absorption have also been broadly investigated[35–38]. Different mass transfer models have been proposed and developed to characterize and predict the CO<sub>2</sub> absorption performance in various absorbents[39]. Moreover, extensive research efforts have been devoted to the micro reactor design and structure optimization with the aim of mass transfer intensification[35,40–42], other researchers focus on multi-phase mass transfer process in micro reactor, such as Yao et al.[43] reviewed in detail the two-phase mass transfer mechanism and dynamics, but lack of the specific application on CO<sub>2</sub>

absorption. Relevant reviews have also been published, as summarized in Table 2. Nevertheless, some of them are focused on the absorbents for  $CO_2$  capture[44], with special focus on calcium oxide[45], potassium carbonate[46], ionic liquids[47], biphasic solvents[48], etc., while others try to review the factors affecting  $CO_2$  absorption, such as absorber structure, dimensions, operational temperature, pressure, etc., but more focused at the macroscopic scale[49,50]. Recently, Afzal et al.[42] presented a comprehensive review on the microchannel geometry, fabrication methods and applications in prospective industries. Noteworthy is the recent paper of Pasha et al.[5] reviewing the hydrodynamics and mass transfer characteristics of  $CO_2$  absorption in micro reactors. The two-phase flow patterns, the microreactor designs, the influencing factors and the economic aspect are specially focused and described. Some mass transfer intensification methods were mentioned, but only focusing on the overall structure of the reactor while more attempts on the local structuration were still absent. In brief, a systematic and comprehensive state-of-art literature review to keep track with the relevant studies and new developments on the microchannel  $CO_2$  mass transfer models, the impact of different operating conditions, the measurement of local & interfacial parameters and the process intensification measures is still lacking.

Study (year)	Main contents	Key findings
Zhang et al.[45] (2015)	Calcium oxide-based sorbents for CO <sub>2</sub> capture under high temperature.	Under high temperature, water vapor can augment the humidity which enhances the $CO_2$ adsorption capacity of CaO. Detailed mechanism of the influence of steam during carbonation remains to be clarified.
Mumford et al.[44] (2015)	Solvent-based $CO_2$ capture technologies, and insight on the new $CO_2$ absorption solvent.	Importance of new absorbents and absorber miniaturization is highlighted from the aspects of cost expenditure and absorption efficiency.
Hu et al.[46]	Potassium carbonate absorbents used for $CO_2$ absorption.	Optimum condition of potassium carbonate absorbents is discussed for practical
(2016)	Reaction efficiency improvement.	industrial CO <sub>2</sub> absorption.
Mehdi et al.[51] (2018)	Gas-liquid and liquid-liquid mass transfer in microchannel reactor.	Effects of aspect ratio, surface properties of microchannel wall etc. on mass transfer
	Influencing factors on microchannel based mass transfer performance.	need to be further clarified.
Aghaie et al.[47] (2018)	Current status and future prospects about $CO_2$ capture by ionic liquids solvents.	Ionic liquids solvents show good absorption performance and economic advantages. Influence of different parameters are clarified.
Ochedi et	CO <sub>2</sub> capture based on the liquid absorption methods.	Phase-change absorbents and absorbents with nanoparticles show great advantages in
al.[50] (2021)	Absorption performance, absorption mechanism and challenges.	CO <sub>2</sub> chemical absorption with low energy consumption.
Yao et al.[43]	Two-phase flow and mass transfer mechanism in microchannel.	Microfluidic technology enhances multi- phase reactions owning to the enhanced
(2021)	Some application cases involving CO <sub>2</sub> capture.	heat and mass transfer, and enables precise control of the reaction process.

Table 2. Summary of some published reviews related to CO<sub>2</sub> chemical absorption

Pasha et al.[5] (2022)	Hydrodynamics, mass transfer characteristics, designs, influencing factors and economic analysis on CO <sub>2</sub> absorption in micro reactors.	Micro-absorber shows great advantages on $CO_2$ absorption with a higher capital expenditure. Parallelized microchannels can augment the capture throughput, but the two-phase misdistribution issue remains to be overcome.	
Meng et al.[49] (2022)	Aqueous amine solution for $CO_2$ chemical absorption.	Amine scrubbing technology is rather promising for CO <sub>2</sub> capture; Several issues should be overcome including environment risks and high energy consumption.	
Afzal et al.[42] (2022)	Microchannel fabrication methods and applications in large-scale and prospective industries.	Compared with straight microchannel, the fabrication and two-phase flow analyze of sinusoidal microchannel are more complicated and need furtherly investigated.	
Gautam & Mondal[48] (2023)	The characteristics, applications and prospects of novel biphasic solvents.	Biphasic solvents are featured by fast reaction kinetics, high absorption capacity, and low regeneration energy required.	
	Two-phase flow patterns and mass transfer models;	Optimizing the micro-absorber structure can greatly improve the $CO_2$ absorption	
	Effect of operating parameters on the CO <sub>2</sub> absorption in microchannel;	performance and extend the potential for industrial application.	
This article	Identification and measurement of some interfacial & local parameters;	The measurement of interfacial and local parameters is a key issue for CO <sub>2</sub> absorption model building and structure optimization	
	Measures to enhance the CO <sub>2</sub> absorption performance in microchannel-based reactors.	model building and structure optimization.	

This review paper is thereby expected to fill the literature gap by gathering and surveying previous works on the microfluidic-based  $CO_2$  absorption technology, a research topic with rapid progress in recent years. In particular, the paper has the following objectives:

- A combined understanding of basic flow patterns, mass transfer dynamics and microreactor geometries;
- A complete survey on the effects of various operational factors on the CO<sub>2</sub> absorption performance in microchannel, including operating temperature and pressure, gas and liquid velocity, gas composition, and the absorbent concentration;
- A special focus on the local & interfacial parameters (e.g., liquid film thickness, local CO<sub>2</sub> concentration and velocity profiles) and their measurement by experiments, which are firstly documented;
- An overview of the CO<sub>2</sub> absorption performance intensification measures proposed and implemented by different researchers, essential for the applications in large-scale industries of this technology.

This paper may serve as an essential reference that contributes to the development and exploitation of highly-efficient microfluidic-based CO<sub>2</sub> chemical absorption technology for its future application and propagation in different industrial sectors.

## 2. Fluid flow and mass transfer of CO<sub>2</sub> chemical absorption in microchannel

This section presents basic elements about the microfluidic-based CO<sub>2</sub> chemical absorption process. Since CO<sub>2</sub> is the targeted gas phase, different chemical absorbents have been proposed and used as the liquid phase, including ammonia and its derivatives[52], alkaline[53], amine absorbent blends[54], ionic Liquids functioned absorbents[55] and phase change absorbents[56]. Developing highly-performant chemical absorbent with expected features has become a very important subject in CO<sub>2</sub> chemical absorption, but beyond the scope of this paper. Interested readers are invited to consult ref.[50] for more details.

## 2.1 Two-phase flow patterns for CO<sub>2</sub> absorption in microchannel

The gas-liquid two-phase flow pattern plays a determinant role on the mass transfer characteristics of CO<sub>2</sub> chemical absorption in microchannel. Basic flow patterns identified for CO<sub>2</sub> chemical absorption in microchannel include bubbly flow, slug (Taylor) flow, slug-annular flow, and churn flow. Detailed description and illustration of these flow patterns may refer to the review paper[5]. Some other specific flow patterns have also been reported, such as slug-bubbly flow, distorted Taylor flow, complete and partial bubble break flow, etc.[57–59]. Most of them are linked to the channel structuration and will be introduced in the later section 5.2.

Two-phase flow maps are then constructed to better predict and determine the flow regimes in microchannel and their possible transition within a certain range of working conditions. Common flow maps use superficial velocity (U) or Weber number (We) of the gas and liquid phase as coordinates, each flow pattern occupying a specific region on the map. Detailed and systematic studies on the basic two-phase flow maps in microchannel may be found in ref.[60]. Specially for CO<sub>2</sub> absorption, the chemical reaction rate would affect the shift of regime transitions under certain conditions[61,62].

Among the above mentioned common flow patterns, the slug flow (Taylor flow) seems to be the most suitable one for microchannel-based  $CO_2$  absorption because of the stable gas-liquid mass transfer interface, simple manipulation and easy controllability. As a result, it has received much attention and has been widely used in relevant researches reported in the literature (cf. Table 3).

## 2.2 Microchannel geometry

The fabrication method of microchannels, such as photolithography, soft lithography, or micromachining[63], can be used to set the geometry of microchannel absorber such as size, cross-sectional shape & junction type. These geometry parameters have been found to have a significant influence on the two-phase flow patterns which is essential for the CO<sub>2</sub> chemical absorption. The hydraulic diameter ( $d_h$ ) of the microchannel used for CO<sub>2</sub> absorption is usually between 200  $\mu$ m and 1000  $\mu$ m, with a cross-sectional shape of circular, rectangular, square, triangle, trapezoid, etc. (Fig.2a). The length of microchannel ( $L_c$ ) used for CO<sub>2</sub> absorption study in laboratory is usually shorter than 400 mm depending on the visualization window of the instruments. More details of microchannel geometry are summarized in Table 3.

The cross-sectional shape of microchannel directly affects the distribution of liquid film thickness (LFT) between the gas phase and the wall, thereby influencing the generation and the moving of Taylor

bubbles[64]. Choi et al.[65] reported the increased Taylor bubble velocity with the decreasing aspect ratio of the cross-section due to the corner effect. Cantu-Perez et al.[66] numerically investigated the effect of sectional shapes (e.g., flat, circle-arc etched, rectangular-shape etched and V-shape etched) on the CO<sub>2</sub> absorption by NaOH solution. They demonstrated much more stable flow pattern in flat rectangular shape microchannel because of the thin, spatially uniform and stable liquid film caused by corner capillary effect. For better visualization purpose, flat rectangular micro-channels become more frequently used for CO<sub>2</sub> chemical absorption experiments.

The microchannel size directly influences the energy dissipation of whole CO<sub>2</sub> absorption process[67–69]. Yue et al.[70] experimentally demonstrated that the increased pressure drop (caused by smaller  $d_h$ ) promoted the slug flow pattern in a much larger gas velocity range. Ganapathy et al.[71] demonstrated that the mass transfer rate could also be strengthened, e.g., liquid side volume mass transfer coefficient ( $k_L a$ ) increased by 2.6 times when the microchannel size decreased from 762  $\mu$ m to 254  $\mu$ m.



Figure 2 Conventional cross-section shapes and junction types of microchannel reactor for CO<sub>2</sub> chemical absorption

Many researchers have reported that the junction (mixing) structure could directly affect the gas phase bubble generation and growth in the microchannel. The most widely used junction types include T-junction, Y-junction, cross-junction, etc. (Fig.2b). It has been reported that the blocking-squeezing mechanism of T-junction at low liquid flow rate could generate stable bubble and Taylor flows[72,73], therefore preferred for microchannel-based CO<sub>2</sub> absorption experiments (cf. Table 4). Tan et al.[74] tested the mass transfer rate of different gas-liquid inlet angle (60°-150°) for a rectangular microchannel and verified that the T-junction rendered better mass transfer performance because of the highest shear force. This conclusion has been confirmed by numerous experimental studies in the literature[75–77]. Instead of straight microchannel, other channel settings (spiral, serpentine, etc.) have also been proposed and tested, in view of intensifying the CO<sub>2</sub> absorption. This aspect will be further introduced in the later section 5.1.

In this section, some basic mass transfer models for CO<sub>2</sub> absorption in microchannel are introduced. The schematic for some mass transfer models are shown in Fig.3.



Figure 3 Schematic of mass transfer model. (a) Thin film mass transfer model. (b) Penetration mass transfer model. (c)Two film mass transfer model. (d) Unit-cell model. Adapted with permission from Ref. [78], copyright (2020) Elsevier

#### 2.3.1 Thin film model

Thin-film (or stagnant-film) model[79] (Fig.3a) assumes that a thin stagnant liquid film exists for stable gas molecular diffusion into the liquid phase. This model fits the condition that the gas and liquid phases are stable, or the diffusion through the film caused by concentration difference dominates the mass transfer. The basic overall mass transfer rate can be expressed as:

$$n_{0-1} = kA(C_0 - C_1) \tag{1}$$

Where  $n_{0-1}$  (kmol/h) is the molar flow rate , A (m<sup>2</sup>) is the gas-liquid contact area, k is the mass transfer coefficient,  $C_0$  and  $C_1$  (mol/L) are the solution concentration at the contact surface and the bulk solution, respectively. k could be described by the Fick's first law:

$$n_{0-1} = \frac{D_{diff}}{\delta} A(C_0 - C_1)$$
 (2)

Where  $\delta$  is the thin film thickness,  $D_{diff}$  is the diffusion coefficient. Although this model can characterize some features of the real mass transfer process in a simple way, the prediction or determination of  $\delta$  is sometimes difficult because it depends on the reactor geometry, two-phase flow pattern and physical properties (e.g., viscosity, density, etc.). Besides, significant error may occur for mass transfer prediction, especially when convection-based mass transfer cannot be neglected.

#### 2.3.2 Penetration model

Penetration model (Fig.3b)[79,80] are based on two assumptions: (1) a fixed hypothetical diffusion layer at steady-state mass transfer; (2) the diffusion-dominated mass transfer in the perpendicular direction to the interface whereas the convection-dominated mass transfer in the liquid flowing direction.

 $n_{0-1}$  for CO<sub>2</sub> absorption can be written as Eq. (3), and the liquid-phase mass transfer coefficient can be expressed using the Crussler formula[81]:

$$n_{0-1} = 2 \sqrt{\frac{D_{diff}}{\pi \tau}} A(C_0 - C_1)$$
(3)

$$k_L = 2 \sqrt{\frac{D_{co_2}}{\pi \tau}} \tag{4}$$

Where  $\tau$  is the gas-liquid contact time calculated by:

$$\tau = \frac{3600whL_c}{Q_G + Q_L} \tag{5}$$

 $Q_G$  and  $Q_L$  is the volumetric flow rate for the gas and liquid phase, respectively. *w*, *h*,  $L_c$  is width, height and length of channel respectively.  $D_{co_2}$  is the CO<sub>2</sub> diffusion coefficient in the chemical absorbent.

#### 2.3.3 Two-film model

The two-film model[82,83] (Fig.3c) assumes a gas stagnation layer (gas film) and a liquid stagnation layer (liquid film) on each side of the gas-liquid interface. CO<sub>2</sub> molecules are firstly transported to the two-phase interface and then dissolved in the liquid phase due to the partial pressure differences. Finally, the CO<sub>2</sub> required for chemical reaction is ready and begins to react with the liquid absorbent. The molar flux of volumetric mass transfer  $N_{co_2}a$  (kmol/m<sup>3</sup>h) can be calculated as Eq. (6) based on the mass conservation:

$$N_{co_2}a = \frac{n}{V_c}(y_{co_2}^{in} - y_{co_2}^{out})$$
(6)

Where  $V_c$  (m<sup>3</sup>) is reaction section volume of the micro reactor (such as microchannel) and  $N_{co_2}$  (kmol/m<sup>2</sup>.h) is the molar flux. For stable two-phase slug flow, the gas-liquid specific surface area (*a*) of a single long bubble can be calculated as:

$$a = \frac{\pi w^2 + [2\pi r + 4(w - 2r)](L_b - w)}{V_c}$$
(7)

Where  $L_b$  is the length of single bubble, *r* is the average radius of bubble cross-section.

 $N_{CO_2}a$  can also be expressed as the product of a mass transfer coefficient and the corresponding driving force[84], such as:

$$N_{co_2}a = K_G a p (y_{co_2}^G - y_{co_2}^*)$$
(8)

Where *p* (kPa) is absorption pressure,  $K_G$  (kmol/kPa.h.m<sup>2</sup>) is the overall gas phase-based mass transfer coefficient,  $y_{co_2}^G$  and  $y_{co_2}^*$  is the molar fraction of CO<sub>2</sub> in the gas phase and at two-phase interface, respectively.  $y_{co_2}^*$  is near-zero because of the instantaneous reaction between CO<sub>2</sub> molecular and absorbent molecular in the liquid phase[85]. As a result, Eq. (8) can be simplified as:

$$N_{co_2}a = K_G a p y_{co_2}^G \tag{9}$$

 $y_{co_2}^G$  is not a constant but varies along with the position of bubble. In practice, the average value of  $y_{co_2}^G$  between the inlet and outlet of the microchannel is often used:

$$y_{co_{2}}^{G} = \frac{y_{co_{2}}^{in} - y_{co_{2}}^{out}}{ln\left(\frac{y_{co_{2}}^{in}}{y_{co_{2}}^{out}}\right)}$$
(10)

 $K_G a$  can then be expressed by:

$$K_G a = \frac{N_{co_2} a}{p} \left( \frac{ln\left(\frac{y_{co_2}^{in}}{y_{co_2}^{out}}\right)}{y_{co_2}^{in} - y_{co_2}^{out}} \right)$$
(11)

 $N_{co_2}$  can also be expressed by liquid side mass-transfer coefficient in stable CO<sub>2</sub> absorption process, written as:

$$N_{co_2} = k_L (C_e - C) \tag{12}$$

Where  $k_L$  (m/s) is the liquid side mass-transfer coefficient, *C* is the CO<sub>2</sub> concentration in liquid equaling to 0 because of complete chemical reaction.  $C_e$  is the CO<sub>2</sub> equilibrium concentration at the interface, expressed as Eq. (13) according to Henry's law[86]:

$$C_e = p_{CO_2} / He \tag{13}$$

Where *He* (Pa.m<sup>3</sup>/mol) is Henry constant and decided by dissolution type and temperature.  $p_{CO_2}$  is the average CO<sub>2</sub> gas pressure in microchannel. The CO<sub>2</sub> molar quantity of whole bubble absorbed into the liquid in the main channel can be expressed as:

$$i * n_{gas} = \frac{p_{in}V_{in} - p_{out}V_{out}}{RT}$$
(14)

Where *R* is the gas constant, *T* is the experimental temperature and *i* is the number of bubbles in the microchannel.  $N_{co_2}$  and  $k_L$  can then be calculated as:

$$N_{co_2} = \frac{i * n_{gas}}{a} = \frac{(p_{in}V_{in} - p_{out}V_{out})}{aRT}$$
(15)

$$k_{L} = \frac{N_{co_{2}}}{C_{e}} = \frac{(p_{in}V_{in} - p_{out}V_{out})He}{aRTp_{CO_{2}}}$$
(16)

#### 2.3.4 Unit-cell model

The unit-cell model[78] (Fig.3d) is based on five main hypotheses: (1) no mass transfer between unitcells; (2) concentration equilibrium at gas-liquid interface; (3) mass transfer only from gas phase to liquid phase; (4) same velocity of gas phase and liquid phase; and (5) constant liquid side volume mass transfer coefficient along the microchannel. The  $CO_2$  concentration variation along the two phase motion direction (*z*-direction in Fig.3d) can be written as follows:

$$\frac{dC_{CO_2}}{dz} = \frac{k_L a_L}{U_{TP}} (C_e - C_{CO_2}(z))$$
(17)

Where  $U_{TP}$  is the two phase flow velocity calculated as:

$$U_{TP} = \frac{Q_G + Q_L}{S} = \frac{dz}{dt} \tag{18}$$

*S* (m<sup>2</sup>) is the microchannel cross-section area and *z* is the length from the supposed gas-liquid interface (*z*=0) to the calculation point. The  $k_L a$  along *z* direction can be expressed as:

$$k_L a(z) = \frac{U_{TP}}{z} ln(\frac{C_e - C_{CO_2}(z=0)}{C_e - C_{CO_2}(z)})$$
(19)

A key information for this model is the CO<sub>2</sub> concentration in liquid slug, which can be measured by many ways such as Raman spectra measurement[78]. More information about the CO<sub>2</sub> concentration measurement can be found in section 4.2 of this paper.

#### 2.3.5 Hydrodynamic model

Under the elevated system pressure, calculation methods based on the bubble volume may be no longer well suited because of the deviation from ideal gas assumption. Some simplified hydrodynamic models were proposed. For example, Jepsen[87] proposed the energy dissipation model as follows:

$$k_L a = \frac{\Delta p_F}{L_c} (U_G + U_L) \tag{20}$$

Where  $\Delta p_F$  is the inlet-outlet pressure drop caused by flow friction in microchannel. Yue et al.[88] studied the mass transfer process of the CO<sub>2</sub>/water system in the microchannel and proposed a simplified model with fewer parameters:

$$k_L a = 0.0863 \left(\frac{\Delta p_F}{L_c}\right)^{0.881}$$
(21)

Nevertheless, Guo et al.[89] pointed out that the average deviation between the predicted value and the experimental value could reach about 40%, the model correction is thereby needed. Yang et al.[90] proposed to include  $U_G$ ,  $U_L$  and E (the enhancement coefficient of chemical reaction rate on mass transfer) in the model:

$$k_L a = n_1 \left(\frac{\Delta P_F}{L_c}\right)^{n_2} (U_G + U_L)^{n_3} E^{n_4}$$
(22)

$$E = \frac{k_L}{k_L^0} \tag{23}$$

Where  $k_L^0$  is the physical mass transfer coefficient in the liquid phase, and  $n_1$ - $n_4$  are fitting constants by experimental data.

#### 2.3.6 Empirical correlations

Besides the above-mentioned physical models, some empirical correlations have also been proposed for fast estimation and prediction. These correlations usually have the form of liquid side Sherwood number ( $Sh_L = k_L d_h/D_{CO_2}$ ) in relation to other dimensionless numbers such as  $Re_L$ ,  $Re_G$ ,  $Sc_L$ , Ca, etc. For example, Yue et al.[37] investigated the microchannel based CO<sub>2</sub>-H<sub>2</sub>O absorption and proposed the correlation for slug flow (Eq. 24) and for slug-annular flow (Eq. 25), respectively.

$$Sh_{L} \cdot a \cdot d_{h} = 0.084Re_{G}^{0.213} \cdot Re_{L}^{0.937} \cdot Sc_{L}^{0.5}$$
<sup>(24)</sup>

$$Sh_L \cdot a \cdot d_h = 0.058Re_G^{0.344} \cdot Re_L^{0.912} \cdot Sc_L^{0.5}$$
(25)

For furtherly investigated the effects of physical parameters on mass transfer performance, Ji et al.[91] tested the mass transfer performance of CO<sub>2</sub>/DI water, CO<sub>2</sub>/ethanol and CO<sub>2</sub>/n-propanol in 40  $\mu$ m×240  $\mu$ m, 100  $\mu$ m×800  $\mu$ m and 100  $\mu$ m×2000  $\mu$ m rectangular microchannels within  $Re_L(1.1 - 13.07)$  and  $Re_G(0.87 - 61.9)$ . Eq. (26) was put forward for Taylor bubble flow and annular flow:

$$Sh_{L} \cdot a \cdot d_{h} = 0.22Re_{G}^{0.78} \cdot Re_{L}^{0.0535} \cdot Sc_{L} \cdot Ca_{L}^{0.7586}$$
(26)

For predicting the chemical reaction acompanied mass transfer performance, Niu et al.[92] investigated the CO<sub>2</sub>/N<sub>2</sub>-NHD system in a circular microchannel ( $d_h$ =1 mm). Eq. (27) was obtained to predict the mass transfer performance within the range of  $U_L$  (0.03 – 0.9 m/s) and  $U_G$  (0.03 – 9.98 m/s):

$$Sh_L \cdot a \cdot d_h = 0.116Re_6^{0.39} \cdot Re_L^{0.7} \cdot Sc_L^{0.5}$$
(27)

Li et al.[93] experimentally investigated the CO<sub>2</sub>/MEA solution mass transfer performance in a 400  $\mu$ m×600  $\mu$ m rectangular microchannel under Taylor flow and annular flow pattern (26.73 <  $Re_G$  < 467.79 and 5.35 <  $Re_L$  < 37.42). Two empirical correlations were proposed with a maximum error of 8.91% and 4.73% for small and large  $Re_G/Re_L$ , respectively.

$$Sh_L \cdot a \cdot d_h = 5.0107 Re_G^{2.9423} \cdot Re_L^{-1.5583} \cdot Sc_L^{0.5} \cdot Ca_L^{0.0743} \quad \frac{Re_G}{Re_L} < 1.7319$$
(28)

$$Sh_L \cdot a \cdot d_h = 19.3895 Re_G^{0.3872} \cdot Re_L^{0.8865} \cdot Sc_L^{0.5} \cdot Ca_L^{0.4176} \quad \frac{Re_G}{Re_L} \ge 1.7319$$
(29)

However, these prediction correlations are only based on physical parameters and for a specific microchannel geometry. Large discrepancy may occur when predicting  $CO_2$  chemical absorption in microchannel. For this issue, Yin et al.[94] introduced the *Da* number to characterize the effect of chemical reaction on mass transfer:

$$Sh_{L} \cdot a \cdot d_{h} = 0.81 \cdot Re_{G}^{0.78} \cdot Re_{L}^{0.41} \cdot Da^{0.35}$$
(30)

Where Da is the Damköhler number ( $Da = k_{ov} \cdot d_h^2/D$ ), and  $k_{ov}$  is the overall chemical reaction rate (1/s). Zhang et al.[95] investigated the CO<sub>2</sub> absorbed by sodium glycinate aqueous solution in microchannel and proposed to include the enhancement number *E* in the correlation:

$$Sh_L \cdot a \cdot d_h = 0.004 \cdot Re_G^{1.262} \cdot Re_L^{0.587} \cdot Sc_L^{0.5}E^{1.1955}$$
(31)

$$\mathbf{E} = \frac{\sqrt{k_{ov}D}}{k_I^0} \tag{32}$$

The above-mentioned correlations were proposed for smooth straight microchannels. For structured microchannels like non-straight channel and/or with obstacles inside, a corrective factor should be added to take the enhancement effect into account. For example, Chu et al.[96] proposed to add the enhancement number *E* for a serpentine microchannel (400  $\mu$ m×400  $\mu$ m):

$$Sh_L \cdot a \cdot d_h = 1.3425 Re_G^{1.0968} \cdot Re_L^{-0.4705} \cdot Sc_L^{0.5} \cdot Ca_L^{-0.0302} \cdot E^{0.7198}$$
(33)

Zhang et al.[59] proposed to add the structure factor  $\beta$  for their microchannel geometry with sudden expansion:

$$Sh_L \cdot a \cdot d_h = 0.084 Re_G^{1.064} \cdot Re_L^{0.422} \cdot Sc_L^{0.5} \cdot \beta^{0.191}$$
(34)

A summary of the studies over various microchannel geometries, used absorbents and absorption performances can be found in Table 3.

Absorbent	Microchannel structure	Size	Flow pattern	Operation condition	$\eta_{CO_2}$	<i>k</i> <sub>L</sub> (m/s)	<i>k<sub>L</sub>a</i> (1/s)	$\Delta p$ (kPa)	Ref.
NaHCO <sub>3</sub> /Na <sub>2</sub> CO <sub>3</sub>	Y-type rectangular	1000 μm×500 μm L <sub>c</sub> =48 mm	Slug	<i>U<sub>G</sub></i> : 0.7-13 m/s <i>U<sub>L</sub></i> : 0.09-1 m/s		14.4×10 <sup>-4</sup>	21	24	[37]
DI water	Y-type rectangular, square	d <sub>h</sub> =667, 400, 200 μm L <sub>c</sub> =48 mm	bubbly, slug, slug-annular, churn and annular	U <sub>G</sub> : 0.04-60 m/s U <sub>L</sub> : 0.02-1 m/s					[70]
PZ, MDEA	Cross T-type rectangular	d <sub>h</sub> =500, 1000, 2000 μm L <sub>c</sub> =180mm		U <sub>G</sub> : 2-5 m/s U <sub>L</sub> :0.02-0.05 m/s	98%			6.1	[97]
MEA	T-type rectangular	400 μm×600 μm L <sub>c</sub> =45 mm	Slug, annular	<i>Q<sub>G</sub></i> : 50-875 ml/h <i>Q</i> <sub>ℓ</sub> : 10-70 ml/h			0.25-54.94		[93]
DEA	Cross T-type circular	d <sub>h</sub> =762 μm	Slug	U <sub>G</sub> : 1.8-9.1 m/s U <sub>L</sub> : 0.04-0.4 m/s	Close to 100%		11-132	10.2	[98]
MDEA+PZ	Microporous tube-in-tube	d <sub>out</sub> =15 mm d <sub>in</sub> =15.5, 16, 16.5, 17 mm d <sub>micropore</sub> =5, 10, 20, 40, 80 µm	Annular	<i>Q<sub>G</sub></i> : 0-5.32 × 10³ ml/h <i>Q</i> <sub>L</sub> : 0-10⁵ ml/h	97%		1.7		[99]
Ammonia	Cross T-type square	$500 \ \mu m \times 500 \ \mu m$ $\mu m$ $L_c=60 \ mm$		<i>Q<sub>G</sub></i> : 2.9×10 <sup>4</sup> ml/h <i>Q<sub>L</sub></i> : 100-300 ml/h	96.45%				[100]
MEA	Cross T-type circular	d <sub>h</sub> =800 μm, L <sub>c</sub> =25 mm		<i>Q<sub>G</sub></i> : 0.6-5.4×10⁵ ml/h <i>Q</i> <sub>L</sub> : 0-2400 ml/h	Close to 100%		941.75		[76]
MDEA	Rectangular	1000 μm×600 μm L <sub>c</sub> =100 mm		<i>Q<sub>G</sub></i> : 15-90 ml/h <i>Q∟</i> : 20-50 ml/h					[101]
MDEA, MEA	T-type square	400 μm×400 μm L <sub>c</sub> =36 mm	Slug, long bubbly	<i>Q<sub>G</sub></i> : 20-160 ml/h <i>Q⊾</i> : 20-50 ml/h		8.5×10 <sup>-4</sup>	6		[102]
MEA, DEA, a-MDEA	Circular	d <sub>h</sub> =600 μm, L <sub>c</sub> =25 mm		$Q_{ m G}$ : 1.8, 3, 3.5 $ imes$ 10 <sup>5</sup> ml/h $Q_{L}$ : 0.6, 1.2, 1.8 $ imes$ 10 <sup>3</sup> ml/h	100%, 100% and 82%, respectively				[103]

Table 3 Summary of microchannel-based CO<sub>2</sub> absorption conditions and performances reported in different researches.

MEA, DEA, a-MDEA, methanol	Circular	d <sub>h</sub> =800 μm, L <sub>c</sub> =35 cm		<i>Q<sub>L</sub></i> : 30-270 ml/h	99%		3		[104]
DEA, DEEA	T-type square	400 μm×400 μm L <sub>c</sub> =30 mm	Slug, slug annular	<i>Q<sub>G</sub></i> : 10-650 ml/h <i>Q</i> ≀: 20-100 ml/h	Close to 100%	33.03× 10 <sup>-4</sup>	3.04-20.07		[61]
MEA, DEA	Cross T-type circular	d <sub>h</sub> =800 μm, L <sub>c</sub> =35 cm		<i>Q<sub>L</sub></i> : 18, 54, 90 ml/h	77.55% and 98.46%		1.91, 3.48		[105]
MEA+DEEA+[Apmim][BF <sub>4</sub> ]	T-type square	400 μm×400 μm L <sub>c</sub> =30 mm	Taylor-bubbly, Taylor and Taylor-annular	<i>Q<sub>G</sub></i> : 10-650 ml/h <i>Q</i> <sub>ℓ</sub> : 20-80 ml/h		22.1×10 <sup>-4</sup>	15		[75]
NaOH	Helical string microchannel	d <sub>h</sub> =500, 1000 μm		$Q_{G}$ : 0-13.8 ml/h $Q_{l}$ : 1-3 $ imes$ 10 <sup>4</sup> ml/h		2.51×10 <sup>-4</sup>			[106]
Silica nanofluids+SDS	Serpentine rectangular	800 μm×800 μm L <sub>c</sub> =120 mm	Taylor	<i>Q<sub>G</sub></i> : 40-700 ml/h <i>Q</i> ℓ: 40-80 ml/h	80%		6.7	0.59- 9.07	[107]
K <sub>2</sub> CO <sub>3</sub>	T-type square long serpentine	1000 μm×1000 μm L <sub>c</sub> =134 mm	Slug, Slug- bubbly	<i>Q<sub>G</sub></i> : 6-600 ml/h <i>Q</i> ≀: 60-180 ml/h		6.5× 10⁻⁴	1.4		[108]
MEA+SDS	T-type square	400 μm×400 μm L <sub>c</sub> =30 mm	Slug	<i>Q<sub>G</sub></i> : 20-280 ml/h <i>Q</i> ℓ: 20-40 ml/h		40× 10 <sup>-4</sup>	27		[109]
MEA+DEA/MEA+TEA	Cross T-type, circular	d <sub>h</sub> =600 μm L <sub>c</sub> =25 cm	Slug, annular	<i>Q</i> <sub>G</sub> : 1.8-9×10⁵ ml/h <i>Q</i> <sub>L</sub> : 20-40 ml/h	97.9% and 91.9%				[15]
MEA+[Bmim][BF <sub>4</sub> ]	T-type square	400 μm×400 μm L <sub>c</sub> =40 mm	Slug flow	U <sub>G</sub> : 0.139-0.694 m/s U <sub>L</sub> : 0.035-0.087 m/s		20× 10 <sup>-4</sup>	12		[94]
L-threonine, L-valine, L- alanine and glycine	T-type square long serpentine	400 μm×400 μm L <sub>c</sub> =108 mm	Slug-bubbly	<i>Q<sub>G</sub>:</i> 10-120 ml/h <i>Q∟</i> : 10-50 ml/h	Close to 100%		2.5		[95]
HMDA, AEEA, BDA, EG, ChCl	T-type circular	d <sub>h</sub> =2000 μm L <sub>c</sub> =5 cm		Q <sub>G</sub> : 3.6-9.6×10³ ml/h <i>Q<sub>L</sub>:</i> 15 ml/h	98%				[110]

## 3. Effects of operating conditions on the CO2 absorption in microchannel reactor

The operating conditions have a direct impact on the  $CO_2$  absorption performance of microchannelbased reactors. Therefore, understanding the separate influence of different factors is of great help to promote the overall  $CO_2$  absorption performance.

## 3.1 Temperature

The operation temperature affects the  $CO_2$  absorption in microchannel in different ways. At high temperature, the reaction rate constant becomes higher, enhancing the mass transfer. Meanwhile, the solution viscosity also decreases, generating thinner liquid film layer in slug/Taylor flow because of the reduced viscous drag[111]. Nevertheless, high temperature can also reduce the  $CO_2$  solubility in the solvent and favor the reverse (desorption) reaction.

Many researches have been performed to reveal the impact of operation temperature. Ye et al.[112] tested the CO<sub>2</sub>/MEA system ( $C_{CO2}$ =11.3 vol.%;  $C_{MEA}$ =30 wt.%) in T-junction microchannel reactors (700  $\mu$ m×300  $\mu$ m×60 mm, 700  $\mu$ m×500  $\mu$ m×60 mm, 700  $\mu$ m×700  $\mu$ m×60 mm) under the temperature of 298.15 to 318.15 K. Their results showed the positive effect of temperature rise, i.e., the maximum  $\eta_{co_2}$  at 318.15 K (95%) is about 5% higher than that at 298.15 K under the same  $U_G$  (Fig.4a). Niu et al.[97] tested the CO<sub>2</sub> absorption in PZ activated MDEA in circular channels ( $d_h$ =0.5, 1, 2 mm,  $L_c$ =180mm) under a wider range of temperature (303 K-350 K) and showed that the maximum  $\eta_{co_2}$  could be reached at 328 K at different  $U_G$  (Fig.4b). Aghel et al.[103] compared the  $\eta_{co_2}$  in three different absorbents (MEA, DEA and a-MDEA) in a circular microchannel ( $d_h$ =600  $\mu$ m,  $L_c$ =25 cm) at different temperatures (Fig.4c). At low temperature (<300 K), the CO<sub>2</sub> mass transfer flux showed a downward trend due to the dominated physical absorption (especially for a-MDEA). For MEA and DEA, a mass transfer peak due to chemical absorption appeared.

But generally speaking, the influence of operating temperature is relatively small compared to other factors, which will be introduced in the latter sub-sections.



Figure 4 Effect of temperature on the performance of CO<sub>2</sub> absorption in microchannel reactor. (a) CO<sub>2</sub>/MEA. Reproduced with permission from Ref.[112], copyright (2013) Elsevier. (b) CO<sub>2</sub>/PZ active MDEA. Reprinted with permission from Ref.[97], copyright (2009) American Chemical Society. (c) CO<sub>2</sub>/MEA, DEA and MDEA, Reproduced with permission from Ref. [103], copyright (2009) Elsevier.

#### 3.2 Liquid and gas flow rates

The gas and liquid flow rates (velocities) have a significant influence on the performance of  $CO_2$  absorption in microchannel reactor[103,113]. While their ratio (e.g.,  $U_G/U_L$ ) usually determines the two-phase flow pattern as introduced in section 2.1, here their main effect is discussed separately.

At high  $Q_L$ , the number of active sites per unit time for accepting and absorbing CO<sub>2</sub> increases, which will improve the  $N_{CO_2}a$  and  $\eta_{cO_2}$ . According to the unit cell theory, the global CO<sub>2</sub> absorption performance is largely decided by the amount of chemical absorbent in the unit bubble volume[114]. The increased  $Q_L$  would lengthen the liquid slug which also augments the CO<sub>2</sub> mass transfer flux. Kittiampon et al.[100] tested CO<sub>2</sub>/ammonia solution system in a T-type microchannel (500  $\mu$ m×500  $\mu$ m×60 mm) and showed 10% increase of  $K_G a$  with the increasing  $Q_L$  (0.12-0.3 ml/h) (Fig.5a). Aghel et al.[103] verified this trend within a larger  $Q_L$  range (0-0.04 L/min) using MEA, DEA or MDEA as the absorbent. Their results showed that the  $N_{CO_2}a$  firstly increased rapidly with the increasing  $Q_L$  but then reached a plateau (Fig.5b), due to the reduced gas-liquid contact time in the microchannel at high  $Q_L$ . Consequently, the  $Y_{CO_2}$  might be lowered at excessive  $Q_L$ , implying a low utilization rate of the chemical absorbent.



Figure 5 The effect of gas or liquid flow rate on  $CO_2$  absorption performance in microchannel. (a) Effect of  $Q_L$  on  $K_Ga$ . Reproduced with permission from Ref.[100], copyright (2017) Elsevier. (b) (c) Effect of  $Q_L$  on mass transfer flux and effect of  $Q_G$  on  $N_{CO_2}a$ , respectively. Reproduced with permission from Ref. [103], copyright (2019) Elsevier. (d) Effect of  $Q_G/Q_L$  on liquid side mass transfer coefficient. Reproduced with permission from Ref.[96], copyright (2019) Elsevier.

#### 3.3 CO<sub>2</sub> partial pressure and system pressure

The influences of  $CO_2$  partial pressure and system pressure have been discussed by several researchers[100,115–117]. High  $CO_2$  partial pressure facilitates the dissolution of  $CO_2$  molecular into the liquid phase. However, the insufficient absorbent molecules for excessive  $CO_2$  involved in the reaction would cause the  $CO_2$  accumulation in liquid phase. Consequently, the driving force for the

mass transfer will be reduced and the dominant liquid side film resistance increased, leading to the decreased mass transfer.

In order to verify this phenomenon, Zeng et al.[117] used the aqueous ammonia as absorbent in a packed column (400 mm in height and 100 mm in diameter) and showed the slight decreasing trend of  $K_G a$  with the increase of CO<sub>2</sub> partial pressure (Fig.6a). The negative impact of CO<sub>2</sub> partial pressure increase could be much more pronounced in microchannel reactors. The experimental study of Kittiampon et al.[100] showed 50% decrease of  $K_G a$  when CO<sub>2</sub> partial pressure increased from 50 kPa to 300 kPa (Fig.6b).

Meanwhile, high pressure CO<sub>2</sub> is often encountered in some industrial processes such as for the CO<sub>2</sub> removal from high pressure natural gas (2-7 MPa). Yao et al.[115] studied the effect of elevated system pressure (0.1- 4.0 MPa) on the CO<sub>2</sub> absorption by DEA solution in a parallel microchannel reactor. Their results showed clearly the negative impact of high system pressure on the  $k_L a$  (Fig.6d). Nevertheless, the results of Niu et al.[97] indicated that the  $\eta_{co_2}$  increased by 15% to 20% when the system pressure increased from 0.1 to 0.6 MPa (Fig.6c), mainly because of the prolonged reaction time caused by the compressed gas phase. Similar tendency was also reported by Yao et al.[115] with the higher system pressure (up to 4 MPa, Fig.6f). Besides, high system pressure could decrease the energy consumption for the CO<sub>2</sub> absorption process, especially at high  $Q_L$  (Fig.6e)[115]. The reasonable control of system pressure can effectively improve the CO<sub>2</sub> absorption performance with a low energy consumption.



Figure 6 Effect of CO<sub>2</sub> partial pressure and system pressure on the absorption performance. (a) The effect of CO<sub>2</sub> partial pressure on  $K_G a$ . Reprinted with permission from Ref.[117], copyright (2011) American Chemical Society. (b) The effect of elevated CO<sub>2</sub> partial pressure on the  $K_G a$ . Reproduced with permission from Ref. [100], copyright (2017) Elsevier. (c) The effect of system pressure on the  $\eta_{co_2}$ . Reprinted with permission from Ref. [97], copyright (2009) American Chemical Society. (d-f) The effect of elevated system pressure on overall volume mass transfer rate, two-phase pressure drop and  $\eta_{co_2}$ , respectively. Reproduced with permission from Ref. [115], copyright (2017) Elsevier.

#### 3.4 Absorbent concentration

The absorbent concentration directly influences the CO<sub>2</sub> chemical absorption performance. The study of Aghel et al.[103] clearly showed that increasing the concentration of solvents (MEA, DEA or MDEA)

would greatly augment the  $K_G a$  (Fig.7a). Kittiampon et al.[100] showed that in the low concentration range (4-10 wt.%), the  $K_G a$  increased monotonously with the increasing concentration of aqueous ammonia (Fig.7b). However, the higher absorbent concentration would augment the liquid viscosity, which may inhibit the CO<sub>2</sub> absorption. Ma et al.[75] found an optimal concentration value (0.6 mol/L) of [Apmim][BF<sub>4</sub>] that rendered the highest  $k_L$  (20.7×10<sup>-4</sup> m/s) (Fig.7c). Janati et al.[15] studied the CO<sub>2</sub> absorption using MEA+DEA or MEA+TEA in a T-junction microchannel ( $d_h$ =600  $\mu$ m;  $L_c$ =25 cm). For MEA+DEA solvent, the  $\eta_{co_2}$  was kept increasing with the increase of both amine concentration and volume percentage. For MEA+TEA solvent, these effects seemed much more sensitive and complicated, showing a parabolic trend, the optimal value being 21.5 wt% and 4 vol/vol%, respectively.

Nevertheless, the ammonia solution at high concentration will cause more serious corrosion problem to the container. For this reason, the concentration of traditional MEA (or most of amine blends) is controlled at  $\leq$ 30 wt% for most of the applications. In the meantime, the development of appropriate high-concentration amine absorbent blends by adding additives is becoming the focus of recent researches. Freeman et al.[118] studied the 8 M PZ in a wetted wall column and found that the  $N_{CO_2}a$  could be improved by a factor of 1.5-3.0 compared to 7M MEA. Li et al.[119,120] tested PZ+HMDA and PZ+DAB and showed that the  $\eta_{cO_2}$  could be improved by a factor of 1.5 to 2 compared to 30wt% MEA. Conway et al.[121] tested three types of higher-concentration amine blends (3M MEA+3M DMEA, 3M MEA+3M AMP, and 3M MEA+3M DEEA). The cyclic capacity (re-utilize efficiency of absorbent) could be increased by about 26%-11% compared to 5M MEA absorbent. Other advantages of high-concentration absorbent have also been recognized, such as low thermal degradation rate[122] and low oxidative degradation (4 times slower than 7M MEA)[118].



Figure 7. Influence of absorbent concentration on the  $CO_2$  absorption performance. (a) Effect of amine concentration (MEA, DEA and MDEA) on the  $K_Ga$ . Reproduced with permission from Ref. [103], copyright (2019) Elsevier. (b) Effect of aqueous ammonia concentration on  $K_Ga$ . Reproduced with permission from Ref. [100], copyright (2017) Elsevier. (c) Effect of [Apmim][BF4] concentration on the  $k_L$ . Reproduced with permission from Ref. [75], copyright (2021) Elsevier.

#### 3.5 Gas composition

In real carbon capture practice (e.g., power plant flue exhaust decarburization, biogas decarburization, etc.), the CO<sub>2</sub> to be absorbed is not pure but mixed with other gases, its concentration is thereby varied. Nitrogen with stable chemical properties is often used as the carrier gas in current laboratory experiments[100,103], the CO<sub>2</sub> concentration being set at 10 vol.% to simulate most of the practical cases. But for oxygen-enriched combustion of fossil energy, the CO<sub>2</sub> concentration could be much higher. Several studies have been dedicated to evaluate the effect of CO<sub>2</sub> concentration on the absorption performance in microchannel. Akkarawatkhoosith et al.[123] experimentally studied the chemical absorption of CO<sub>2</sub>-rich gas (40–60 vol.%) in a T-junction rectangular microchannel (500

 $\mu$ m×500  $\mu$ m×60 mm) using water as the solvent. It has been shown that the highest  $\eta_{co_2}$  could reach 70.9% with CO<sub>2</sub> concentration of 53.7%. In another study of microchannel-based biogas decarburization using MEA[124], 65% CH<sub>4</sub>– 35% CO<sub>2</sub> mixture was used to represent the biogas. Results showed 98.9% CH<sub>4</sub> purity and  $k_L a$  of 47-517 s<sup>-1</sup>, one or two orders of magnitude higher than traditional macroscopic wetted-wall columns (below 10 s<sup>-1</sup>). Aghel et al.[125] used synthesized gas (60% CH<sub>4</sub>, 40% CO<sub>2</sub>) to conduct CO<sub>2</sub> separation and biogas purification in a T-junction microchannel reactor. Their results indicated the augmentation of  $K_G a$  by a factor of 5-22 compared with the traditional packed towers.

Other multiple waste gases presenting in the CO<sub>2</sub> absorption process (e.g., SO<sub>2</sub>, NO<sub>2</sub> and other acid gases) can accelerate the degradation of MEA solution, thereby reducing the overall CO<sub>2</sub> absorption performance[126]. For alkaline absorbents, like NaOH, the presence of acid waste gases would also depress the CO<sub>2</sub> absorption because of the low screening capability[127]. At present, CO<sub>2</sub> separation before absorption is still the main method to improve the CO<sub>2</sub> capture performance in various industrial applications.

#### 3.6 Short summary

Aghel et al.[103] have experimentally investigated the contribution rate of different factors on  $CO_2$  absorption performance in the microchannel based reactor. For all three tested absorbents (MEA, DEA and a-MDEA), the influence of temperature is the smallest (Fig.8). For a-MDEA, the absorbent concentration had the largest influence on volumetric mass transfer while for the other two, the liquid-side mass transfer resistance seemed to be the limiting factor. In real practice, such dominant factor should be determined for certain  $CO_2$  absorption solvent and microchannel reactor geometry, so as to better control and improve the  $CO_2$  absorption performance.



*Figure 8 Sensitivity analysis for the effect of operating variables on the mass transfer flux. Reproduced with permission from Ref.*[103], *copyright (2019) Elsevier.* 

## 4. Interfacial & local parameters of CO2 absorption in microchannel

Gas-liquid interfacial and local parameters are essential for the comprehensive understanding of the transport process, allowing the development of the mass transfer theory/model on one hand, and the performance optimization of the  $CO_2$  absorption device on the other hand. In this section, the identification and characterization of such interfacial & local parameters in microchannel are presented. While the CFD simulation methods are frequently employed for this purpose, special focus in given on the experimental measuring techniques.

## 4.1 Liquid film thickness (LFT)

The LFT around the Taylor bubbles is in close relation to the mass transfer of gas-liquid two-phase flow in microchannel[128–131]. The mass transfer rate usually decreases with the increase of LFT due to the larger liquid-side mass transfer resistance[132] (Fig.9a).

Many studies have been devoted to the precise measurement of LFT and the development of empirical correlations based on some dimensionless numbers (*Re, Ca, We* or others), as summarized in Table 4. For example, Han & Shikazono[133] used laser focus displacement meter (LFDM) to measure the LFT at small *Ca* number condition (Fig.10c). Two semi-empirical correlations were put forward to predict the LFT under bubble steady motion and acceleration motion, respectively (cf. Table 4). By using the laser induced fluorescence (LIF) method, Bartkus & Kuznetsov[134] showed the deviation of the measured LFT from the Taylor LFT prediction model especially for the position away from the microchannel corner (Fig.10b). Cheng et al.[135] used the nano-particle tracking velocimetry technique (Mn-PTV) to measure the local oscillation of bubble LFT in square microchannels. Polystyrene fluorescent nanoparticles (20 nm) were used as the tracer particles and the measuring uncertainty was estimated to be smaller than 8%. Their result showed that the LFT exhibited periodic changes and with the decrease of channel dimension, the lag time of bubble movement was significantly shortened (Fig.10d).

It should be noted that most of the experiments and the proposed correlations do not consider the influence of mass transfer. Experiments and simulation studies combining the CO<sub>2</sub> mass transfer and dimensionless LFT are especially needed for a better understanding of CO<sub>2</sub> absorption in microchannel.



Figure 9 Investigation of LFT distribution around the gas-liquid interface in microchannel. (a) LFT vs. mass transfer performance of the Taylor bubbly flow on different position obtained from 3D simulation. Reproduced with permission from Ref.[132], copyright (2016) Elsevier. (b) LFT distribution along the microchannel width measured by LIF method. Reproduced with permission from Ref. [134], copyright (2016) EDP Sciences. (c) LFT distribution around the bubble acceleration mode measured by LFDM. Reproduced with permission from Ref. [133], copyright (2010) Elsevier. (d) LFT distribution around the oscillation bubble using the Mn-PTV method. Reproduced with permission from Ref. [135], copyright (2020) Elsevier.

Microchannel/capillary parameters	Fluids	Flow pattern/status	Investigation method and details	Semi/empirical formula for LFT prediction	Ref.
Glass tube: $d_h$ =2.26 mm	Air Water	Single slug	Bubble-tube measurement	$\frac{\delta_0}{d_h} = 0.5(Ca)^{0.5}$	[136]
Glass tube: $d_h$ =2; 3 mm	Air Water	Taylor	Visual measurement	$\frac{\delta_0}{d_h} = \frac{0.67Ca^{2/3}}{1 + 3.35Ca^{2/3}}$	[137]
Capillary tubes 1 mm< <i>d<sub>h</sub>&lt;</i> 2 mm	-	Taylor	Finite-difference simulation	$\frac{\delta_0}{d_h} = 0.36(1 - e^{-3.08Ca^{0.54}})$	[138]
Capillary tubes $d_h$ =0.84; 1.24; 1.56, 2.92 mm	Air Silicone oils; ethanol; alkanes	Single slug	Visual camera shooting (Capture frequency: 50 fps)	$\frac{\delta_0}{d_h} = \frac{1.34Ca^{2/3}}{1 + 3.35Ca^{2/3}}$	[139]
Square channel 1.56 mm×1.56 mm	Air Water	Taylor	Small platinum micro- probes method (Sample frequency up to 10 000 Hz)	$\frac{\delta_0}{d_h} = 0.72(1 - 0.7e^{-2.25Ca^{0.445}})$	[140]
Circular tubes $d_h$ =0.5; 0.7 and 1.0 mm	FC-40; Air Ethanol; water	Liquid-gas interface	LFDM Precision 99% (Resolution: 0.01 μm; Response time: 640 μs)	$\left(\frac{\delta_0}{d_h}\right)_{steady} = \frac{0.67Ca^{2/3}}{3.13Ca^{2/3} + 1 + 0.504Ca^{0.672}Re^{0.580} - 0.352We^{0.629}}$	[133]
Parallel channels: <i>h</i> =0.1; 0.3; 0.5 mm	Air Ethanol	Liquid-gas interface	Interferometer (Capture frequency: 5000 fps; Uncertainty: 0.534 µm) LFDM (Capture frequency: 1000 fps; Precision: 99%; Bocolution: 0.01 µm)	$\frac{\delta_0}{d_h} = \frac{0.67Ca^{2/3}}{3.13Ca^{2/3} + 1 + 0.504Ca^{0.672}Re^{0.589} - 0.352We^{0.629}}$ (Re <re critical)<="" td=""><td>[141]</td></re>	[141]
Circular tube 0 <ca<1.9< td=""><td>-</td><td>Horizontal</td><td>Bretherton model based numerical analysis</td><td><math display="block">\frac{\delta_0}{d_h} = \frac{1.38Ca^{2/3}}{1 + 3.732Ca^{2/3}}</math></td><td>[142]</td></ca<1.9<>	-	Horizontal	Bretherton model based numerical analysis	$\frac{\delta_0}{d_h} = \frac{1.38Ca^{2/3}}{1 + 3.732Ca^{2/3}}$	[142]
Circular tubes $d_h$ =1 mm	FC-40; air Ethanol; water	Slug Taylor	LFDM (Precision 99%; Resolution 0.01 μm; Response time 640 μs)	$\left(\frac{\delta_0}{d_h}\right)_{accel} = \frac{0.968Ca^{2/3}}{Bo^{0.414} + 4.838Ca^{2/3}}$	[143]

Table 4 Summary of main researches about the measurement of dimensionless liquid film thickness  $\delta_0/d_h$  for gas-liquid two phase flow in microchannel.

Rectangular microchannel	Nitrogen	Elongated	LIF		[134]
420 $\mu$ m×280 $\mu$ m, 720 $\mu$ m×1500 $\mu$ m	Water	bubble	(Resolution: 0.5 $\mu$ m)	-	
Circular tubes: $d_h$ =0.7; 1.0 and 1.3 mm	FC-40 Water; ethanol	Taylor-bubbly	Interferometer and LFDM (Precision: 99%; Resolution $0.01  \mu$ m)	$ \begin{pmatrix} \frac{\delta_0}{d_h} \end{pmatrix}_{decel} = \frac{0.67Ca^{2/3}}{3.36Ca^{2/3} - 0.352We^{0.640} + (1 + 0.534Ca^{0.672}Re^{0.593})(e^{-0.02Bo_{decel}^{1.45}})} $	[144]
T-type rectangular microchannel $100~\mu$ m $ imes$ 83 $\mu$ m	CO <sub>2</sub> Ethanol; methanol	Taylor-bubbly	Volume of Fluid (VOF) based 3-D simulation	- -	[132]
Circular tube $d_h$ =0.305; 0.715 and 1.305 mm	FC-40; air Water; ethanol	Bubbly	Arbitrary-Lagrangian– Eulerian based simulation	$\frac{\delta_0}{d_h} = \frac{1.34Ca^{2/3}}{3.13Ca^{2/3} + 1 + 0.504Ca^{0.672}Re^{0.589} - 0.305We^{0.664}}$	[145]
Square microchannel $d_h$ =510; 1020 $\mu$ m	Air Water	Slug	High-speed camera visualization (Precision: >93.2%; Resolution: 2.8 μm)	$rac{\delta_0}{d_h}=0.085Ca^{-0.137}$ (For corner film thickness)	[146]
Rectangular microchannel 3 mm×3 mm; 1.6 mm×1.6 mm; 0.5 mm×0.5 mm and 0.3 mm×0.3 mm	Air Water	Liquid-gas interface	Mn-PTV (Uncertainty < 8%; Resolution 0.02 μm; Capture frequency 500 fps)	-	[135]

## 4.2 CO<sub>2</sub> local concentration

The chemical reaction proceeds directionally according to the  $CO_2$  concentration gradient in the liquid, the mass transfer rate being non-uniform in different positions in microchannel. Knowing the local  $CO_2$  concentration distribution between the bubble border and the liquid slug is thereby much needed.

CFD simulation has been frequently used[147–149]. Soh et al.[147] studied the diffusion and the distribution of CO<sub>2</sub> molecules for a bubble moving in the silicone oil in a microchannel using the VOF model (Fig.10a). Li et al.[150] successfully simulated the CO<sub>2</sub> absorption in a tube-in-tube microchannel (Fig.10b), using the modified eddy-cell model coupled with VOF method. Dong et al.[151] simulated the CO<sub>2</sub> absorption by water, ethanol solution, 0.2 M MEA solution, and 0.2 M NaOH solution in a circular microchannel ( $d_h$ =500 µm) using the initial unit model. The CO<sub>2</sub> distribution in liquid phase around the bubble in CO<sub>2</sub>-water and CO<sub>2</sub>-NaOH system at the same reaction time were compared (Fig.10d), demonstrating the dominant role of chemical reaction on quick CO<sub>2</sub> mass transfer.



Figure 10 Investigation of concentration distribution of gas-liquid two-phase flow in microchannel. (a) Dissolved  $CO_2$  molecules distribution in silicone oil by CFD simulation. Reproduced with permission from Ref. [147], copyright (2017) Springer. (b) Simulation result of  $CO_2$  mass fraction distribution in a microporous tube-in-tube microchannel. Reproduced with permission from Ref. [150], copyright (2020) Elsevier. (c)  $CO_2$  concentration distribution in liquid phase around bubbles detected by pH-Sensitive LIF method. Reprinted with permission from Ref. [152], copyright (2012) American Chemical Society. (d)  $CO_2$  concentration in the liquid phase around the bubble ( $CO_2$ -H<sub>2</sub>O and  $CO_2$ -NaOH system). Reproduced with permission from Ref. [151], copyright (2020) Wiley. (e) 2D and 3D simulation of gas molecules diffusion in the liquid phase around a moving bubble in microchannel. Reproduced with permission from Ref. [153], copyright (2013) Elsevier. (f)  $CO_2$  concentration distribution in the liquid phase around the permission from Ref. [154], copyright (2022) Elsevier.

Regarding experimental techniques, Deleau et al.[78] used Raman spectroscopy and showed that the local CO<sub>2</sub> concentration along the capillary tube presented an upward trend but accompanied with a reduced growth rate. More recently, Deleau et al.[154] further explored the CO<sub>2</sub>-water segment flow mass transfer performance in a straight circular microchannel ( $d_h$ =250  $\mu$ m) under high pressure (10 MPa) by using pH sensitive colorimetric method. Their results showed that the  $k_La$  ranged from 1 to 13 s<sup>-1</sup> along the length direction (Fig.10f). Kuhn & Jensen[152] developed a pH-sensitive LIF method (5-(and-6)-carboxy SNARF-1 used as fluorescent dye) and showed that the mass transfer happened mainly at bubble caps in microchannel (Fig.10c). Nevertheless, only the CO<sub>2</sub> concentration distribution of a 2D

plane could be obtained, due to the limitation of excitation chip laser. Ichiyanagi et al.[155] implemented the 'confocal micro-PIV +LIF' method to investigate the CO<sub>2</sub> concentration distribution in spiral and meandering microchannels (400  $\mu$ m×400  $\mu$ m), illustrating clearly the dominating mass transfer at the bubble caps. Dietrich et al.[153] studied the oxygen-water mass transfer process in a straight circular microchannel ( $d_h$ =750  $\mu$ m) using oxygen-sensitive dye and a monochromatic CCD high-speed camera (Fig.10e). This colorimetric technique could be a promising tool for local CO<sub>2</sub> concentration measurement in microchannel.

#### 4.3 Local liquid velocity

Various approaches have been employed to capture the liquid velocity field of Taylor/bubbly flow in microchannel (especially near the gas-liquid interface), most of them demonstrating the existence of symmetrical[151,156] or asymmetrical[114] local fluid microcirculations in the liquid slug. By using the VOF model, Yang et al.[148] simulated the air/water slug flow in a straight circular microchannel ( $d_h$ =200  $\mu$ m). Validated hydrodynamics coupled with chemical reaction pushed mass transfer term was used as solver to capture the circulatory patterns in the segment flow (Fig.11a). Dong et al.[151] showed the symmetrical liquid velocity profile between two CO<sub>2</sub> bubbles around a water slug in a microchannel ( $d_h$ =500  $\mu$ m) by using the unit bubble model and neglecting the gravity effect and the bubble volume change (Fig.11b). Yin et al.[57] also used the VOF model to simulate the liquid velocity field of CO<sub>2</sub>-MEA/[Bmim][BF<sub>4</sub>] aqueous in a microchannel (400  $\mu$ m×800  $\mu$ m×30.4 mm), showing the strongly perturbed velocity profiles due to interior micro-baffle structures (Fig.11c). Similar phenomenon has also been reported for concave heart-shaped groove microchannel (Fig.11d)[157].



Figure 11 Flow velocity field around Taylor bubbles in microchannel. (a) Circulatory patterns in the segment flow by VOF model. Reproduced with permission from Ref. [148], copyright (2017) Elsevier. (b) Simulated velocity and streamline distribution in the micro channel for the CO<sub>2</sub>-water system. Reproduced with permission from Ref. [151], copyright (2020) Wiley. (c) Liquid velocity distribution around the bubble in baffled microchannel by the VOF method. Reproduced with permission from Ref. [57], copyright (2019) Elsevier. (d) Liquid velocity distribution in groove etched microchannel by the VOF method. Reproduced with permission from Ref. [157], copyright (2021) Elsevier. (e)(h) Symmetrical relative velocity field in the liquid slug between two Taylor bubbles in straight channel (flow direction: left to right) and asymmetrical relative velocity field in the liquid slug between two Taylor bubbles in a meandering channel (flow direction: left to right) captured by micro-PIV method, respectively. Adapt with permission from Ref. [114], copyright (2012) Elsevier. (f) Experimental PIV time-averaged velocity fields for slug liquid between two bubbles. Reproduced with permission from Ref. [156], copyright (2018) Elsevier. (g) Liquid velocity vectors around a bubble measured by PTV technique. Reproduced with permission from Ref. [161], copyright (2021) Elsevier

Experimentally, Zaloha et al.[114] used micro PIV method and showed that the liquid microcirculation rate gradually increased with the shorter liquid slug at higher  $U_G$  and  $U_L$  (Fig.11e). Butler et al.[156] used both the PIV and PLIF-I techniques to investigate the O<sub>2</sub>-water mass transfer process in a circular channel ( $d_h$ =3 mm). Both the velocity and O<sub>2</sub> concentration fields were measured, showing great consistency with each other (Fig.11f). Recently, the PTV method has also been applied to measure the velocity field[158–160]. Azadi & Nobes[161] explored the liquid flow velocity around a slug bubble, especially in the thin film and at the corners, in a square mini-channel (3 mm×3 mm) (Fig.11g). Particularly for meandering microchannel, some researchers reported the velocity asymmetry and micro-circulations in the bend position (Fig.11h), implying a possible way to enhance the mass transfer using this geometry[114]. This point will be further discussed in the next section of the paper.

# 5. Intensification measures for $CO_2$ chemical absorption in microchannel reactors

The application of microchannel-based reactors for  $CO_2$  chemical absorption can greatly reduce the size of the equipment, but some limitations still exist such as the lowered throughput and the higher energy consumption to overcome the pressure drop. Further intensifying this process is still the focus of researches, as classified and reviewed in this section. Detailed information about these studies may be found in Table 5.

#### 5.1 Setting the global microchannel geometry

Instead of the common straight channel, serpentine and spiral shapes are proposed and used to strengthen the CO<sub>2</sub> absorption and to save the space in the length direction for easier stacking. These curved channel geometries are expected to disturb the moving boundaries of bubbles (interfaces) by the dean vortex effect[162,163], thereby promoting the mass transfer. Yang et al.[164,165] showed the significantly increased mass transfer rate owing to the enhanced liquid mixing in a curved microchannel (Fig.12a). Zhou et al.[40] designed and experimentally tested a serpentine rectangular microchannel (600  $\mu$ m×600  $\mu$ m) for CO<sub>2</sub> absorption by MEA (Taylor-bubbly flow, Fig.12b), showing 40% increase of the  $K_G a$  compared to that of straight channel. Similar improvement effect has also been reported by Yao et al. [166]. Seo et al. [167] investigated the chemical  $CO_2$  absorption by nickel nanoparticles (NiNPs) catalyzed MEA absorbent in a serpentine microchannel (Fig.12c). Their results showed that compared to the straight channel, the average CO<sub>2</sub> absorption rate could be improved by 34% with the single effect of NiNPs catalyst, and by 54% with the combined effect of catalyst and serpentine geometry. Tan et al.[168] reported that the  $k_L$  of CO<sub>2</sub>/N<sub>2</sub>-NaOH solution segmented flow could be doubled  $(2-7 \times 10^{-4} \text{ m/s})$  in a curve-shaped microchannel compared to that in the straight channel (1-3×10<sup>-4</sup> m/s). Higher  $k_{l}$  could be achieved by decreasing the radius of the U-turns because of the strengthened gas-liquid disturbance.

Kuhn & Jensen[152] compared the  $k_L a$  of CO<sub>2</sub>/alkaline solution between a spiral and a meandering microchannel reactor (Fig.12d) and found that the spiral geometry performed much better owing to the decreased residence time and increased gas volume transport fraction. MacInnes et al.[169] tested

a spiral minichannel rotating reactor (1.5 mm×4 mm) for CO<sub>2</sub>/N<sub>2</sub> absorption by MEA solution, the  $\eta_{CO_2}$ >90% could be reached. Pang et al.[108] experimentally investigated the CO<sub>2</sub> absorption in potassium carbonate solution using a serpentine microchannel (1 mm×1 mm) with 6 elbows. Their results indicated much higher  $k_L$  (0.5-2.5×10<sup>-4</sup> m/s) than that of straight microchannel (0.07-13×10<sup>-5</sup> m/s)[170] and the maximum pressure drop was smaller than 3.49 kPa. All these researches have demonstrated that setting the global microchannel geometry is an effective and practical way for the design of microchannel-based CO<sub>2</sub> absorbers.



Figure 11 Setting of global microchannel geometry to enhance the  $CO_2$  absorption performance. (a) Continuous 90° meandering microchannel. Reproduced with permission from Ref. [165], copyright (2017) Elsevier. (b) Serpentine microchannel with T-type junction. Reprinted with permission from Ref. [40], copyright (2020) American Chemical Society. (c) Serpentine microchannel with hedge-type mixing structure. Adapted with permission from Ref. [167], copyright (2020) Elsevier. (d) Spiral- (left) and meandering- (right) microchannel reactor used for the  $CO_2$ /alkaline solution two-phase flow experiments study. Reprinted with permission from Ref. [152], copyright (2021) American Chemical Society.

#### 5.2 Structuration of channel walls

The structuration of channel walls by installing flow disturbance elements (micro grooves[35,66,171], micro protrusion obstacle[57,172], sudden expansion/shrink, etc.) could induce the local eddy flow of liquid phase and deform the gas-liquid interface, both effects enhance the mass transfer. Chen et al.[35] tested a straight microchannel ( $800 \mu m \times 400 \mu m \times 35.2 mm$ ) etched with heart-shaped grooves (Fig.13a) and found that the  $k_L a$  for CO<sub>2</sub>-[Bmim][BF<sub>4</sub>] aqueous solution could be increased by 150%-210% compared with the smooth microchannel. Similar enhancement rate for CO<sub>2</sub>-NaOH solution has been reported by Cantu-Perez et al.[66] owing to the grooves (100  $\mu$ m) etched in a falling film microreactor constituting of 16 parallel rectangular mini-channels (1200  $\mu$ m×400  $\mu$ m×66.4 mm) (Fig.13b).

For micro protrusion obstacle, the most commonly used structures include rectangular, triangular and cylindrical shapes having different flow disturbing capacities [173]. Among them, the rectangular protrusion obstacle has been reported to be the most effective for single-phase flow in microchannel [174], thereby also frequently applied for gas-liquid flows. Yin et al. [175] fabricated a rectangular microchannel (400  $\mu$ m×800  $\mu$ m) with staggered rectangular obstacles (Fig.13c) and investigated the effect of geometrical parameters (length, arrangement, etc.) on the CO<sub>2</sub>/water mass

transfer performance. Their results showed that the maximum  $k_L a$  could be increased by a factor of 2.8 owing to the greatly shredded liquid-gas boundary layer by obstacle elements.



Figure 13 Wall structuration for microchannel gas-liquid reactors. (a) Straight microchannel etched with heart-shaped grooves. Reproduced with permission from Ref. [35], copyright (2021) Elsevier. (b) Parallel mini-channels reactor with rectangular grooves. Reproduced with permission from Ref. [66], copyright (2012) Elsevier. (c) Microchannel with protrusion baffle structure. Reproduced with permission from Ref. [175], copyright (2020) Elsevier. (d) Microchannel accompanied with fan and triangular shaped sudden expansion structure. Reproduced with permission from Ref. [176], copyright (2015) Elsevier. (e) Microchannel accompanied with arced rectangular shape sudden expansions structure. Reproduced with permission from Ref. [59], copyright (2021) Elsevier. (f) Combined sudden expansion structure with rectangular obstacle in the middle. Reproduced with permission from Ref. [58], copyright (2022) Elsevier.

Some researchers have demonstrated that sudden expansion/ shrink structured microchannel (Fig. 13d) has the unique advantage in inducing two-phase mixing, i.e., the oscillation of gas bubble and surrounding liquid[176–178]. Zhang et al.[59] fabricated the arced sudden expansion structured microchannel reactor to test the CO<sub>2</sub> absorption performance in [Bmim][BF<sub>4</sub>] aqueous solution. Shrink and expand slug flow was captured (Fig.13e), leading to an improvement of  $k_L a$  by a factor of 1.2-2.4 compared with the smooth microchannel with a total pressure drop smaller than 5 kPa. Adding micro-obstacles in the middle of the microchannel has also been proposed to generate the bubble breakup. Yin et al.[58] combined the sudden expansions layout with rectangular obstacle (600  $\mu$ m×300  $\mu$ m) and observed complete and partial bubble breakups (Fig.13f). The  $k_L a$  could be augmented by a factor of 2.74 with the maximum pressure drop smaller than 12 kPa, acceptable for most of the industrial applications[179]. Further deisgn and shape/topology optimization the the obstacle geometry for the microchannel for intensified mass transfer process with reasonable pressure drop could be an interesting topic for future researches.

Applying external operating conditions has also been considered, including ultrasound[41,180], electric field[172,181], and special working modes such as high system pressure[115] or mechanical oscillating.

The ultrasound imposed on two-phase flow could induce the regular oscillation of both bubbles (volume oscillation, shape oscillation and high frequency transient oscillation[182], Fig.14a) and liquid slugs in the microchannel, drastically increasing the gas-liquid interfacial area and creating more local vortices and liquid streaming near the two-phase interface[183,184]. Dong et al.[41,184] used high-power ultrasonic (100 W; 20 kHz) to intensify the CO<sub>2</sub>–DI water slug flow mass transfer in a serpentine minichannel (1 mm×1 mm). Their results demonstrated that the  $k_La$  could be enhanced by a factor of 3.3–5.7 with the increase of ultrasonic power from 0 to 70 W. Akbari et al.[185] employed the 1.7 MHz ultrasound waves generated by piezoelectric transducer to accelerate the CO<sub>2</sub> absorption by NaOH solution (Fig.14c), leading to the increased  $k_La$  and a by 21% and 22%, respectively. Among different two-phase flow patterns, the bubbly flow has shown to be the most sensitive to ultrasound waves thus has the most significant enhancement by oscillatory effect. Zhang et al.[186] investigated the effect of ultrasonic wave frequency (20, 28, 40 kHz) and surfactant (sodium dodecyl sulfate) on CO<sub>2</sub> absorption performance in rectangular channels (0.5-2.0 mm in width). The determined optimum condition was at low frequency (20 kHz), high acoustic power (0.14 W/mL) and high surfactant concentration (Fig.14d), resulting in the enhancement of  $k_La$  by a factor of 3 to 22.

The external electric field has been used for osmotic flow to enhance the liquid mixing in the microchannel[172]. Hoseini et al.[187] investigated the effect of external electric field (133, 200, or 266 kV/m) on CO<sub>2</sub> absorption by water/Fe<sub>3</sub>O<sub>4</sub> solution in a small channel ( $d_h$ =6 mm). 16% increase of  $k_L$  could be achieved with 266 kV/m electric field.

Lliuta & Larachi[188] explored the effect of mechanical oscillation (rotation, up-down and rocking oscillation) on CO<sub>2</sub> absorption in MEA solution in packed-bed columns (Fig.14e). The heaving motion (up-down) has shown to result in the best CO<sub>2</sub> absorption performance. Such external oscillatory operation could be inspiring for microchannel-based CO<sub>2</sub> absorption devices.

While the above-mentioned methods show great potential on improving the  $CO_2$  absorption performance, they are rarely applied to real industrial cases due to the additional power input and high equipment investment. For example, the power input for conventional ultrasonic chemical reactor is about 0.05-0.6 W/mL[189–191], meaning that at least 50 KW of additional input power is required for unit volume (1 m<sup>3</sup>) of absorption liquid[184] which is hardly affordable in real industry applications.



Figure 14 Special operating modes for intensifying the CO<sub>2</sub> chemical absorption performance. (a) Oscillations caused by different ultrasound frequency. Reproduced with permission from Ref. [182], copyright (2020) Elsevier. (b) High-power ultrasonic accompanied serpentine mini-channel to strengthen the CO<sub>2</sub> absorption. Adapted with permission from Ref. [180], copyright (2019) American Chemical Society. (c) Ultrasound accompanied T-junction microchannel CO<sub>2</sub> absorption system. Reproduced with permission from Ref. [185], copyright (2017) Elsevier. (d) Oscillation of bubble surface under different acoustic power. Reproduced with permission from Ref. [186], copyright (2021) Elsevier. (e) Schematic of mechanical oscillation mode. Adapted with permission from Ref. [188], copyright (2018) Elsevier.

#### 5.4 Integration of microchannels

The microchannel integration involves the use of a multitude of channels instead of single microchannel in the device, which is considered as an essential step for the scaling-up for industrylevel application. If properly conceited and controlled, the integration of microchannels is equivalent to repeating settings for a single microchannel with high CO<sub>2</sub> absorption performance, but greatly increased throughput (amount of CO<sub>2</sub> processed per unit time). Based on the basic type of straight microchannel, the most commonly used integration pattern at present is through the parallelization (numbering-up), and the problem of connection has been particularly addressed. Tree-like arborescent structure has been frequently utilized to equally distribute and collect the fluids[192,193]. For example, Yue et al.[194] investigated the CO<sub>2</sub>-water mass transfer performance in a 16 parallel microchannel contactor equipped with dichotomic-tree gas and liquid distributors (Fig.15b). It has been observed that the multi-channel concept could be beneficial for maintaining the slug flow pattern at high  $Q_G$  but disordered two-phase flow pattern would lead to the deterioration of mass transfer performance. Yao et al. [115] fabricated a tree-like reactor (Fig. 15c) with 16 main rectangular microchannels (300  $\mu$ m×600  $\mu$ m) and 8 sub microchannels (300  $\mu$ m×800  $\mu$ m) to perform the CO<sub>2</sub> absorption in DEA solution under elevated system pressures (4 MPa). The  $k_L a$  ranging from 1.59 to 90 s<sup>-1</sup> could be obtained mainly due to the increased contacting (residence) time of two-phase flow in the reactor. Zhu et al.[195] designed 4-parallel channel microreactor to carry out the CO<sub>2</sub> absorption (Fig.15a) and showed that the average  $k_L a$  of the multi-channel collection was 0.3-3.5 s<sup>-1</sup>, 5%-10% lower than single microchannel. However, the two-phase flow fluctuation caused by the increase of  $Q_L$  and  $Q_G$  could be eliminated.

Mesh collection is another way to integrate the microchannels[196,197]. Constantinou & Gavriilidis[198] tested and compared the CO<sub>2</sub> absorption performance in 2M NaOH solution by three mesh-structured reactors (pore size: circle 25  $\mu$ m in diameter; rectangular 25  $\mu$ m×75  $\mu$ m; and square 35  $\mu$ m×35  $\mu$ m, 100  $\mu$ m in thickness) (Fig.15d). The increased two phase contact area due to mesh structure would not only help stabilize the gas/liquid interface but also augment the CO<sub>2</sub> absorption performance. Reducing the pore size (5-25  $\mu$ m) and increasing the open area could furtherly increase the  $\eta_{co_2}$  by 20%-25% compared to the previous reactor (Fig.15e)[199].

Some researchers also put forward the structure integration with nature-inspired geometries. For example, Zhang et al.[200] designed a honeycomb-like fractal type reactor ( $d_h$ =3 mm ) to investigate the CO<sub>2</sub>- MEA absorption performance (Fig.15f). Compared to a serpentine tubular reactor, both the  $\eta_{co_2}$  and the CO<sub>2</sub> absorption rate could be increased by about 30%. Recently, the use of triply periodic minimal surface (TPMS) geometries as the CO<sub>2</sub> absorber has been reported owing to its higher *a* and smooth surface[201,202], offering an interesting potential for the design of macro-sized CO<sub>2</sub> absorbers. For example, Singh et al.[201] designed a Schwarz-D cell based CO<sub>2</sub> absorber (Fig.15g) consisting of two separated domains, one for CO<sub>2</sub> and absorbent mixed flow and the other for the cooling fluid for reaction heat management. Sithamparam et al.[203] numerically studied the gyroid-based CO<sub>2</sub> absorber. Despite that this absorber design relies on membrane permeation, it shows a great potential for the development of macro-sized CO<sub>2</sub> absorbers with high efficiency.



Figure 15 Different types of microchannel integration for CO<sub>2</sub> chemical absorption. (a) 4-parallel microchannel-based CO2 absorption reactor with tree-like distributor. Reproduced with permission from Ref. [195], copyright (2020) Elsevier. (b) 16-parallel microchannel CO<sub>2</sub> absorber with dichotomic tree-like gas and liquid inlet distributors. Reproduced with permission from Ref. [194], copyright (2010) Wiley. (c) Tree-like microchannel based CO<sub>2</sub> absorption device. Reproduced with permission from Ref. [195], copyright (2017) Elsevier. (d) CO<sub>2</sub> absorber with microchannel mesh structure. Adapted with permission from Ref. [198], copyright (2010) American Chemical Society. (e) Mesh contactor with SEM picture of the circle silicon nitride mesh. Adapted with permission from Ref. [199], copyright (2012) Elsevier. (f) Honeycomb fractal-type CO<sub>2</sub> absorber. Adapted with permission from Ref. [200], copyright (2018) Elsevier. (g)TPMS structure based CO<sub>2</sub> absorber. Reproduced with permission from Ref. [201], copyright (2022) Elsevier. (h) gyroid structure based CO<sub>2</sub> membrane absorber. Reproduced with permission from Ref. [203], copyright (2021) Elsevier.

Table 5 Summary of intensification measures to improve the CO<sub>2</sub> absorption performance in microchannel-based reactors

Working fluids	Microchannel geometry and dimension	Two-phase flow pattern	Intensification measures	Performance enhancement wrt. reference channel	Energy consumption/pressure drop	Ref.
DEA+CO <sub>2</sub> NaOH+CO <sub>2</sub>	Flat microchannel with a 1 $\mu$ m thickness silicon nitride mesh	Co-current and counter-current flow	Silicon nitride mesh contactor	30% CO <sub>2</sub> absorbed for contact time less than 0.5 s		[199]
DI water+CO <sub>2</sub>	Rectangular serpentine microchannel (1 mm×1 mm)	Slug-bubbly	Serpentine geometry + high- power ultrasonic	$K_L a$ increased by 5.7 times	Power input: 100W	[41]
DEA+CO <sub>2</sub>	16 parallel rectangular microchannel (600 $\mu$ m×300 $\mu$ m)	Taylor-bubbly	Multiple channel collection + serpentine geometry + high system pressure	$k_L^0$ increased by 1-2 orders of magnitude	Maximum Δ <i>p</i> : 0.45 MPa ( <i>Re</i> <sub>L</sub> =1500, <i>Q</i> <sub>GS</sub> =0.1 L/min)	[115]
MDEA+CO <sub>2</sub>	Circular serpentine microchannel $(d_h$ =800 $\mu$ m)	Bubbly	Serpentine geometry + ultrasonic	$k_La$ 10 times higher than conventional reactor	Power input: 100W	[180]
MEA/[Bmim][BF <sub>4</sub> ] aqueous solution+CO <sub>2</sub>	Rectangular straight microchannel (800 $\mu$ m×400 $\mu$ m)	Taylor bubbly	Micro baffle obstacles	$k_L a$ increased by 50%	Maximum Δ <i>p</i> : 2.5 kPa	[57]
MEA+CO <sub>2</sub> +nickel nanoparticles	Rectangular serpentine microchannel (100 $\mu$ m×50 $\mu$ m)	Bubbly	Serpentine geometry + NiNPs catalyst	$\eta_{co_2}$ increased by 54%		[167]
Sodium glycinate aqueous solution+CO <sub>2</sub>	4 parallel rectangular microchannels $(400~\mu{ m m} imes400~\mu{ m m})$	Taylor bubbly	multiple channel collection	$\eta_{co_2}$ reached 80%-100%, average $k_L a_v$ reached 3-4 m-1	$\Delta p$ ranging from 0.63 to 6.7 kPa	[195]
DI water+CO <sub>2</sub>	Rectangular straight microchannel (800 $\mu$ m×800 $\mu$ m, 600 $\mu$ m×600 $\mu$ m and 400 $\mu$ m×400 $\mu$ m )	Slug	Micro rectangular baffle obstacles	$k_L a$ increased by up to 180%	Maximum Δ $p$ : 6 kPa ( $U_G$ =0.25 m/s, $U_l$ =0.069 m/s)	[175]
[Bmim][BF <sub>4</sub> ] aqueous solution+CO <sub>2</sub>	Rectangular straight microchannel (400 $\mu$ m×400 $\mu$ m)	Bubbly	Continuous sudden expansion units inside the microchannel	$k_L a$ increased by 240%	Total $\Delta p$ smaller than 4.5 kPa ( $Q_L$ =30 ml/h, $Q_G$ =180 ml/h)	[59]
Potassium carbonate solution+CO <sub>2</sub>	Rectangular serpentine microchannel $(1000 \ \mu m  imes 1000 \ \mu m)$	Slug bubbly	Special serpentine geometry+SiO <sub>2</sub> particles	$k_L a$ increased by 30%	Δ <i>p</i> ranging from 0.26 to 3.49 kPa	[108]
[Bmim][BF <sub>4</sub> ] aqueous solution+CO <sub>2</sub>	Rectangular straight microchannel (800 μm×400 μm)	Taylor	Heart-shaped grooves inside the microchannel	$k_L a$ increased by 220%	$\Delta p$ ranging from 1.5 to 3.7 kPa	[35]
MEA+CO <sub>2</sub>	Serpentine rectangular microchannel (600μm×600μm)	Taylor	Special serpentine geometry	$k_L a$ increased by 120%		[40]

## 6. Conclusion and prospects

This work provides an extensive review on the chemical absorption of  $CO_2$  using microfluidic devices. The main findings may be summarized as follows.

- Microfluidic-based CO<sub>2</sub> chemical absorption technology has shown to exhibit various advantages compared to large-size conventional equipment, e.g., at least 1 or 2 orders of magnitude higher liquid-side volumetric mass transfer coefficient and interfacial area.
- The absorption of CO<sub>2</sub> in slug/bubbly two-phase flow pattern in microchannel using high-performance absorbents is an effective way to perform the CO<sub>2</sub> capture. The mass transfer between the gas bubble and the liquid phase determines the absorption performance in microchannel.
- Two-film model and unit-cell model are generally used for the mass transfer modeling. Besides, some dimensionless number-based empirical correlations are used for a fast estimation of mass transfer performance. Better predication may be achieved by introducing chemical reaction and structure factors into the correlations.
- Gas and liquid flowrates and the solvent concentration have important impact on the absorption while the influence of working temperature is relatively minor. The elevated system pressure may have a negative impact on the  $K_G a$ , but can improve the overall  $\eta_{co_2}$  and decrease the pressure drop (energy dissipation).
- Measuring local & interfacial parameters by using the non-intrusive optical-based techniques (e.g., LFDM, LIF, PIV, etc.) is very meaningful for understanding the mechanisms of transport phenomena and for the development of new models. The local and transient characteristics obtained can also provide useful insights for the structural optimization of the microchannel.
- Different measures were proposed to enhance the CO<sub>2</sub> mass transfer in microchannelbased reactor, at the cost of additional energy consumption. Compared with external energy input, the structural modification method seems to be more economic. Furthermore, the combination of two or several intensification measures could be a promising strategy.
- The CO<sub>2</sub> processing capacity can be multiplied by adopting proper integration of microchannels, such as parallelization, meshed network, lattice structure, etc. Nevertheless, the uneven distribution of gas-liquid flow remains as an obstacle to overcome for such scaling-up strategy.'

These findings well illustrate both the potential and the current limitations of the microfluidic-based CO<sub>2</sub> chemical absorption technology. Great efforts are still needed to overcome the scientific and technological barriers for the widespread application of microchannel-based CO<sub>2</sub> capture technology in different industry sectors. Some of the remaining challenging issues include but not limited to:

• To develop new and efficient absorbents with low regeneration energy consumption, low corrosiveness and low solvent loss during cyclic operations.

- To improve and develop CO<sub>2</sub> mass transfer models in microchannel based on the accurate and transient information of two-phase flow dynamics and CO<sub>2</sub> concentration field in microchannel, obtained by advanced numerical and measuring techniques.
- To better understand the reaction mechanisms/kinetics and the corresponding heat transfer characteristics, which is largely insufficient at this stage.
- To search for the effective structuration of microchannel geometry based on the advanced modeling, optimization algorithms, and experimental verification, taking the fluid flow and coupled heat and mass transfer into account.
- To design and develop macro-sized, locally microfluidic-based devices possessing both the high CO<sub>2</sub> absorption performance and high throughput of CO<sub>2</sub> treatment, and their realization by advanced additive manufacturing method.
- Scaling-up of microfluidic-based CO<sub>2</sub> absorbers and their field testing with postcombustion gases[204,205] under realistic operation conditions (e.g., high temperature and pressure, gas mixture, etc.). The cost estimation and economic analysis will help showcase the commercialization potential and the competitiveness of such technology for the carbon capture duties in different industrial sectors.

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