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1 A porous medium approach to the 3D modelling of an entire rotating packed bed
2 for post-combustion carbon capture

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6 **Abstract:** Rotating packed bed (RPB) technology shows great potential for post-
7 combustion capture. However, the capture process inside the full RPB is difficult to
8 simulate, due to the complexity of the process and the neglect of the CO₂ capture in the
9 outer cavity zone. In this paper, a full 3D CFD model, including the packing and the
10 inner and outer cavity zones, has been established employing the Eulerian porous
11 medium method coupled with various sub-models. The CO₂ capture performance in the
12 packing and outer cavity zones has been quantitatively analyzed under different
13 operating conditions. The simulation results show good agreement with the
14 experimental data, and the contribution of the outer cavity zone to the CO₂ capture of
15 the RPB is in the range of 28%~42%. This work provides a new approach to efficiently
16 simulate the mass transfer process in the RPB.

17 **Keywords:** rotating packed bed, Eulerian method, 3D porous media model, CO₂
18 absorption, outer cavity zone

19

20 Nomenclatures

a_S	specific area of the packing materials, $\text{m}^2 \cdot \text{m}^{-3}$	$S_{h,i}$	heat of chemical reaction, $\text{W} \cdot \text{m}^{-3}$
A_e	effective interfacial area, $\text{m}^2 \cdot \text{m}^{-3}$	\vec{S}_{GL}	interfacial force between the gas and liquid phases, $\text{N} \cdot \text{m}^{-3}$
C	concentration, $\text{kmol} \cdot \text{m}^{-3}$	t	time, s
C_{pG}	specific heat of gas phase, $\text{J} \cdot \text{kg}^{-1} \cdot \text{K}^{-1}$	T_i	temperature of phase i , K
C_D	drag coefficient	u	flow rate per unit area, $\text{m} \cdot \text{s}^{-1}$
D	diffusivity, $\text{m}^2 \cdot \text{s}^{-1}$	\vec{u}_i	velocity of phase i , $\text{m} \cdot \text{s}^{-1}$
E	enhancement factor	V	volume, m^3
E_i	total energy, J	We	Weber number
f, f^*	drag function	x	radial length of a cell, m
f_e	ratio of wetted packing area to the total packing surface area	y_{i,CO_2} , y_{o,CO_2} , y_{op,CO_2}	CO_2 mole fraction in the gas inlet and outlet and on the outer packing surface, respectively
$\vec{F}_{disp,i}$	dispersion forces of phase i , $\text{N} \cdot \text{m}^{-3}$	Y	mass fraction of species
$\vec{F}_{drag,i}$	interaction force between the phase i and solids of the packing material, $\text{N} \cdot \text{m}^{-3}$	<i>Greek</i>	
g	gravitational force, $\text{m} \cdot \text{s}^{-2}$	α	phase saturation
h	height of the packing, m	γ	packing void fraction (porosity)
h_j	enthalpy of the species, $\text{J} \cdot \text{mol}^{-1}$	β	dynamic contact angle, $^\circ$
h_{GL}	heat transfer coefficient between the gas and liquid phases, $\text{W} \cdot \text{m}^{-2} \cdot \text{K}^{-1}$	ε	volume fraction
H	Henry constant	δ	diffusion layer thickness, m
Ha	Hatta number	ω	angular velocity, $\text{rad} \cdot \text{s}^{-1}$
\vec{j}	mass diffusion flux of species, $\text{kg} \cdot \text{m}^{-2} \cdot \text{s}^{-1}$	κ_G	thermal conductivity of the gas phase, $\text{W} \cdot \text{m}^{-1} \cdot \text{K}^{-1}$
k_i	thermal conductivity, $\text{W} \cdot \text{m}^{-1} \cdot \text{K}^{-1}$	θ	angle of flow direction slop to the bed axis, $^\circ$
k_i	mass transfer coefficient of phase i , $\text{m} \cdot \text{s}^{-1}$	ρ_i	density of phase i , $\text{kg} \cdot \text{m}^{-3}$
k_2	reaction rate constant, $\text{m}^3 \cdot \text{mol}^{-1} \cdot \text{s}^{-1}$	$\Delta\rho_{GL}$	absolute value of the density difference between liquid and gas phases, $\text{kg} \cdot \text{m}^{-3}$
K_L	overall mass transfer coefficient, $\text{m} \cdot \text{s}^{-1}$	μ	dynamic viscosity, $\text{kg} \cdot \text{m}^{-1} \cdot \text{s}^{-1}$

K_{GL}	momentum exchange coefficient between the gas and liquid phases	μ_e	effective dynamic viscosity, $\text{kg} \cdot \text{m}^{-1} \cdot \text{s}^{-1}$
$K_L a_e$	overall volumetric mass transfer coefficient, s^{-1}	τ	particulate relaxation time, s
M	molar mass, $\text{g} \cdot \text{mol}^{-1}$	$\bar{\tau}$	stress tensor, $\text{N} \cdot \text{m}^{-2}$
N_{CO_2}	transferred CO_2 between the gas and liquid, $\text{kg} \cdot \text{m}^{-1} \cdot \text{s}^{-1}$	$\bar{\tau}_{eff,ij}$	effective shear tensor
Nu	Nusselt number	λ_{RT}	Rayleigh-Taylor instability wavelength
P	pressure, Pa	σ	surface tension, $\text{N} \cdot \text{m}^{-1}$
P_c	capillary pressure, Pa	ν	kinematic viscosity, $\text{m}^2 \cdot \text{s}^{-1}$
Pr	Prandtl number	<i>Subscripts</i>	
q	liquid initial velocity number	$cell$	mesh cell
Q	volume flow rate, $\text{m}^3 \cdot \text{s}^{-1}$	CO_2	carbon dioxide
$Q_{h,GL}$	transferred heat between the gas and liquid phases, $\text{W} \cdot \text{m}^{-3}$	G	gas phase
r	radius or radial distance, m	i	=G, L
r_i	inner packing radius, m	j	species
r_o	outer packing radius, m	L	liquid phase
r_{CO_2}	CO_2 reaction rate, $\text{kg} \cdot \text{m}^{-3} \cdot \text{s}^{-1}$	MEA	monoethanolamine
R	gas constant	O	outer cavity zone
R_{ij}	net rate of production of the species j by the reaction, $\text{mol} \cdot \text{m}^{-3} \cdot \text{s}^{-1}$	OW	computational cell next to the casing wall
Re	Reynolds number	P	packing zone
Re'	relative Reynolds number	PW	computational cell next to the packing wall
$S_{m,i}$	mass transfer rate between gas and liquid, $\text{kg} \cdot \text{m}^{-3} \cdot \text{s}^{-1}$	S	solids phase for the packing materials

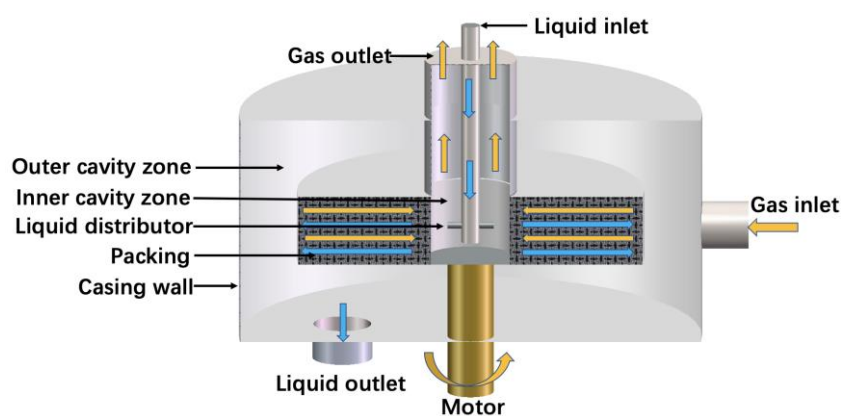
21 1. Introduction

22 CO_2 emissions, as a kind of greenhouse gas, has become an urgent issue in the 21st
23 century (Khan, 2017). In response to this important issue, the EU has a commitment for
24 a net-zero greenhouse gas emissions target by 2050 (Garba, 2012), which aims to reach
25 a balance between the amount of greenhouse gas emissions produced and the amount
26 captured. Particularly, the total power plants contribute a huge amount of CO_2 , which
27 could contribute to 30% of the total CO_2 emissions. Therefore, reducing CO_2 emissions
28 released by the power stations has become an emergent activity.

29 Currently, the amine-based post-combustion CO₂ capture (PCC) is the most common
30 and commercial way to capture CO₂ using the conventional packed bed (CPB), which
31 results in the CPB being large and costly. In addition, the high amine solvent flow rate
32 demanded by the large CPB would consume large heat energy for the rich solvent
33 regeneration. On the other hand, the rotating packed bed (RPB), one of the most
34 important applications in the area of process intensification, has the potential to enhance
35 the CO₂ capture performance and reduce the packed bed size by applying a high
36 centrifugal force (100-1000 times gravity) (Liu et al., 2019). However, the RPB is on
37 the early stage of the technological development for PCC, and the CO₂ capture
38 processes, including the hydrodynamics, heat and mass transfer, are still unclear inside
39 a RPB. Thus, it requires extensive research and design optimization.

40 In general, the RPB may be divided into three parts - inner cavity, packing and outer
41 cavity zones, see Figure 1. At the beginning, the liquid jets out from the liquid
42 distributor and passes through the inner cavity zone entering the packing region, then
43 the liquid contacts and reacts with the opposing gas flow on its way outwards through
44 the packing region. During this process, the liquid breaks into tiny droplets, or generates
45 thin films on the packing surface due to the strong interaction with the rotating mesh
46 packing, which can enhance the interfacial area and micromixing (Yang et al., 2015b)
47 between gas and liquid. In addition, the liquid surface renewal rate in the packed bed is
48 improved, which also leads to a large increase in the CO₂ capture. In addition, the liquid
49 leaves the packing with a large tangential velocity and enters the outer cavity zone in
50 the form of liquid droplets. Finally, the liquid droplets hit the cavity casing wall and the
51 liquid phase flows out from the liquid outlet tube under the influence of the gravitational
52 force. It should be noted that before the liquid leaves the outlet tube, the liquid has been
53 reacting with the gas phase all the time in the outer cavity zone. In addition, the space
54 ratio of the outer cavity zone to the packing region is large (Guo et al., 2019). As a
55 result, it can be concluded that the CO₂ capture in the RPB occurs not only in the
56 packing zone, but also in the outer cavity zone.

57 However, most investigations only focus on the packing region, the contribution of the
58 outer cavity zone for the CO₂ capture is rarely studied. In the experimental
59 investigations, it has been reported that the effective interfacial area of the outer cavity
60 zone could be up to 30% of the entire effective area (Guo et al., 2014; Sang et al.,
61 2017a). Further, experiments showed that the contribution of the outer cavity zone to
62 the mass transfer was determined to take up approximately 13–25% of the total mass
63 transfer in the entire RPB (Sang et al., 2019; Yang et al., 2011). Therefore, the CO₂
64 absorption in the outer cavity zone cannot be ignored.



65

66 **Figure 1.** Schematic diagram of a typical RPB for CO₂ capture.

67 In addition, the Volume of Fraction (VOF) method has widely used to computationally
68 study the CO₂ capture in the RPBs. For instance, the micromixing and mass transfer
69 have been studied in a 2D packing region along with the Volume of Fraction (VOF)
70 method and user defined functions (UDF) (Guo et al., 2016; Yang et al., 2016). The
71 advantage of employing the VOF method is that it can reasonably, clearly and
72 accurately capture the interface between the gas and liquid, including the evolution of
73 the liquid droplets in the RPB. Due to the multiscale nature of the liquid flow in an RPB,
74 the computational mesh should be fine enough to resolve the finest droplet and liquid
75 films, typically in millimeter and even micron meter scales, in addition to the packings,
76 within a meter scale RPB. When modelling in 3D this will require a prohibitively large
77 number of computational cells and computational time. This is the reason why most of

78 the CFD simulations using CFD methods are for small RPB models and mostly in 2D.
79 If the packing in an RPB is treated as a porous medium using the Eulerian model, where
80 the liquid flow is modelled as a continuum, there is no need to resolve the packing or
81 the liquid droplets. Instead, the interfacial area and the momentum transfer between the
82 phases are modelled using experimental correlations. Clearly, this method could not
83 produce a clear picture of the real discrete liquid droplet flow and its accuracy relies on
84 correlations employed for the resistance among the phases and the interfacial area.
85 Nevertheless, the Eulerian method could significantly reduce the simulation time and
86 with appropriate validations, it can potentially be utilized to address the significant
87 difficulties of modelling 3D and large-scale RPBs (Lu et al., 2019; Zhang et al., 2022a).
88 However, the Eulerian porous medium approach has so far seldom been employed for
89 the RPB because the momentum transfer between the phases has to be closed by closure
90 equations. In other words, a suite of mathematical formulations is essential for
91 accurately expressing the flow characteristics in the RPB, such as the sub-models for
92 the forces for the phases and the effective interfacial area between the phases (Fourati
93 et al., 2013; Iliuta et al., 2014). Since the characteristics of the multiphase flow are very
94 different when passing through the rotating packing and in the static cavity space, sub-
95 models for the forces and the interfacial area should be separately introduced for the
96 packing region and the outer cavity zone, which is quite complex. For the packing
97 region, not only the porous resistance should be considered due to the wire mesh
98 packing, but also the dispersion force should be taken into account due to the strong
99 interaction between the liquid and the packing (Jiang et al., 2002; Xu et al., 2019; Zhang
100 et al., 2022b). In addition, it is complex to predict the contact area between gas and
101 liquid due to the non-uniform liquid distribution in the packing zone. The above force
102 and the effective mass transfer area models for the non-uniform multiphase flow in the
103 packing zone have been developed and validated in our previous paper (Zhang et al.,
104 2022a). However, for the outer cavity zone, the liquid phase exists in the form of liquid
105 droplets in the cavity space and liquid film on the casing wall. As a result, different

106 contact area correlations for the liquid droplets and film should be proposed. In addition,
107 there is no existing force models for the outer cavity zone via employing the Eulerian
108 method. Therefore, the appropriate model and correlation for the outer cavity zone
109 should be developed for accurately analysing the hydrodynamics in this zone.

110 In addition to the momentum transfer between phases, the heat and mass transfer should
111 be modelled in the RPB. Many semi-empirical correlations for predicting the mass
112 transfer coefficients have been proposed in the literature based on different applications
113 and packing systems of the RPBs (Munjal et al., 1989), however, these correlations are
114 too general to be confidently employed for the CO₂-MEA absorption system employed
115 (Lu et al., 2019). A method that has been commonly used to model the chemically-
116 enhanced mass transfer in the CPB, is to use an enhancement factor coupled with a
117 mass transfer theory, such as the two-film, penetration, or surface renewal theories.
118 Although penetration and surface renewal theories are more accurate in calculating the
119 mass transfer by introducing the contact time and surface renewal frequency, these two
120 parameters are difficult to measure in the RPB, especially under unsteady-state
121 conditions. Furthermore, the two-film reaction-enhanced mass transfer model has
122 already shown sufficient accuracy for expressing the CO₂ mass transfer among the
123 phases in many CPB investigations (Dashliborun et al., 2019; Kim et al., 2017; Pham
124 et al., 2015), which is introduced into the RPB model as presented in Sections 2.4 and
125 2.5.

126 The thermodynamics in the RPB should also be carefully considered because the liquid
127 temperature and the chemical reaction rate could affect each other in this CO₂
128 chemisorption system. The heat released by the chemical reaction increases the liquid
129 temperature; meanwhile, some of the heat would be carried away by the gas flow due
130 to the temperature difference between the phases so that the gas temperature also
131 increases. The increasing phase temperature would speed up the reaction rate until the
132 whole CO₂ absorption system reaches a balance. In particular, the efficiency of the heat

133 transfer between the phases is determined by the diameter of the liquid droplet. The
134 smaller the droplet diameter, the more heat transfer takes place. The predicted heat and
135 mass transfer processes can be made accurate only when the liquid droplet diameters
136 in the packing and outer cavity zones are set properly.

137 In our previous work, the CO₂ capture performance in the RPB was explored in a 2D
138 packing region by the Eulerian porous medium method (Lu et al., 2019). However, the
139 predicted liquid outlet temperature and the overall volumetric mass transfer rate ($K_L a_e$),
140 which was defined in Section 3.3, were lower than the experimental data because the
141 outer cavity zone was not taken into consideration at the time and the liquid droplet
142 diameter was set using the default value in the software. In this paper, for accurately
143 studying the CO₂ capture process within a RPB, a comprehensive 3D RPB model,
144 including the packing, inner and outer cavity zones, was built based on a practical pilot-
145 scale RPB coupled with various sub-models. The $K_L a_e$ and liquid outlet temperature
146 under various operating conditions were analysed. Furthermore, the ratio of the CO₂
147 capture performance in the packing zone and outer cavity zone was quantitatively
148 investigated. Therefore, this paper provides, for the first time, a new feasible approach
149 to effectively and accurately predict the mass transfer within the entire RPB.

150 **2. CFD simulations**

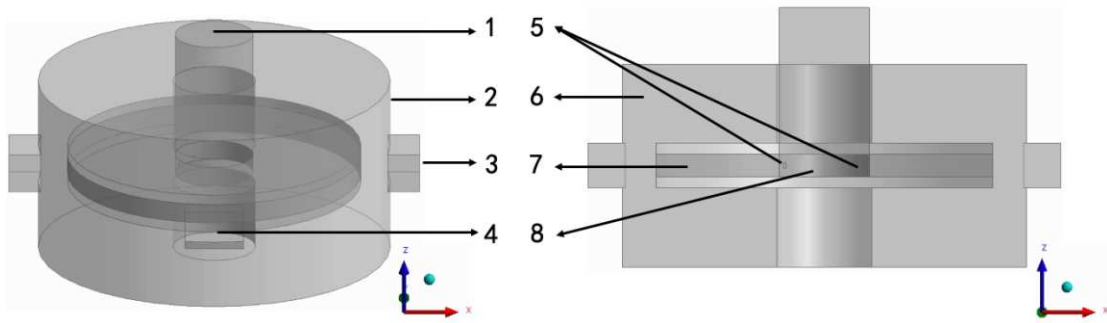
151 In this section, a 3D model has been built that incorporates the porous resistance,
152 dispersion force, two-film reaction enhancement mass transfer and heat transfer
153 models in order to achieve an accurate prediction of the flow dynamics, mass transfer
154 and heat transfer in the whole RPB. Since the resistance and dispersion models
155 developed for the packing zone have been illustrated in our previous paper (Zhang et
156 al., 2022a), only some of the most important models and equations, such as the
157 governing equations, mass and heat transfer models, and the correlations of the
158 effective interfacial area and liquid droplet diameter, are presented in this paper.

159 **2.1 Geometry of the RPB**

160 In order to establish and validate the 3D RPB model, the detailed information of the
161 experimental rig and the quality of the experimental results are required. Among all the
162 published experimental works on RPBs, the Kolawole experimental data (Kolawole,
163 2019) has been selected as the comparison data. The reasons are: (i) the relative detailed
164 operating conditions and dimensions of the RPB have been offered, especially the size
165 of the packing and outer cavity zones. (ii) The CO₂ capture performance with different
166 operational conditions is analyzed by various indexes, including the overall mass
167 transfer coefficient, the liquid and gas phase temperatures at the outlets, etc., which
168 could help to validate the simulation results. And (iii) the experimental data has been
169 presented in the published paper (Lee et al., 2017) and previously utilized by Lu et al.
170 (2019) for verifying their models, thus meaning that the experimental data is reliable.
171 However, no experimental data related to the flow characteristics (i.e. liquid holdup)
172 has been shown in the thesis (Kolawole, 2019), thus, the flow dynamics obtained by
173 the experiment and simulation are not compared in this work.

174 The 3D RPB geometry has been reproduced in Figure 2 according to the experimental
175 rig (Kolawole, 2019). The inner diameter, outer diameter and height of the wire mesh
176 packing are 80, 300 and 20 mm, respectively, and it is made from stainless steel with
177 an expanded mesh grade 707. The void fraction and specific area of the packing are
178 0.801 and 663 m²/m³, respectively. The diameter and the estimated height of the RPB
179 casing are 360 and 180 mm, respectively and the space ratio of the outer cavity zone to
180 the packing zone is about 11. There are two gas inlet tubes and one liquid inlet tube
181 with diameters of 60 and 14 mm, respectively. In order to make the mesh in the outer
182 cavity tetrahedral and further reduce the mesh number, the shape of the gas inlet tubes
183 and one liquid inlet tube is simplified to be a rectangle but with the same flow cross-
184 sectional area. In addition, a gas outlet tube is outside the liquid inlet tube with a
185 diameter being 40 mm. A 2-arm liquid distributor is used, and its length and diameter
186 are 22.4 and 7 mm, respectively. In Figure 2, only two liquid nozzle outlet holes are
187 presented. In order to save computational time, the liquid flows passing through the

188 liquid inlet tube and 2-arm liquid distributor has not been modelled. Initially, a liquid
 189 pressure outlet boundary was built at the bottom of the RPB to ensure that the liquid
 190 phase flows out from the RPB. However, after the simulation, we found that a large
 191 amount of the gas phase would flow out from this boundary at the same time. Thus, an
 192 elimination zone is employed at the bottom of the RPB to remove the liquid phase but
 193 to retain the gas phase, and the appropriate source equations for this zone can be found
 194 in (Zhang et al., 2022a).



195

196 **Figure 2.** Schematic diagram of the 3D RPB (1, gas outlet; 2, case; 3, gas inlet; 4,
 197 liquid elimination zone; 5, liquid inlet; 6, outer cavity zone; 7, packing zone; 8, inner
 198 cavity zone).

199 **2.2 Governing fluid flow equations**

200 The main assumptions of the RPB model are as follows:

- 201 (i) The wire mesh packed bed is a homogenous porous medium.
- 202 (ii) The gas is incompressible.
- 203 (iii) The pressure field is shared by the gas and liquid phases.
- 204 (iv) The liquid phase exists in the form of the film and droplets in the packing
 205 and outer cavity zone.
- 206 (v) The mass transfer only happens in the packing and outer cavity zones
 207 due to the small interfacial area and short contact time in the inner cavity
 208 zone.
- 209 (vi) The heat transfer only occurs between the gas and liquid phases and the

210 casing wall is adiabatic.

211 The continuity equation utilized to calculate the overall mass conservation is given as
 212 follows:

$$213 \quad \frac{\partial}{\partial t} (\varepsilon_i \rho_i) - \nabla \cdot (\varepsilon_i \rho_i \vec{u}_i) = S_{m,i} \quad (1)$$

214 where ρ_i is the density of the i th phase ($i = G$ for gas or L for liquid), t is the time, \vec{u}_i
 215 is the phase velocity, $S_{m,i}$ is the mass transfer rate between phases, and ε_i is the phase
 216 fraction, which is defined as follows:

$$217 \quad \varepsilon_i = \alpha_i \gamma = \frac{V_i}{V_G + V_L} \gamma = \frac{V_i}{V_G + V_L + V_S} \quad (2)$$

218 where α_i is the phase saturation ($\alpha_i = \frac{V_i}{V_G + V_L}$), V_i is the volume of the i th phase, and γ
 219 and V_S are the porosity and solid volume of the packing, respectively, which are 1 and
 220 0 for the outer cavity zone.

221 The momentum conservation equations for the packing and outer cavity zones are
 222 different since the drag, capillary and mechanical dispersion forces only exist in the
 223 packing region. The governing momentum equations for the whole RPB are as follows:

$$224 \quad \frac{\partial}{\partial t} (\varepsilon_L \rho_G \vec{u}_L) + \nabla \cdot (\varepsilon_L \rho_L \vec{u}_L \vec{u}_L) = -\varepsilon_L \nabla P + \varepsilon_L \nabla P_c + \nabla \cdot (\varepsilon \bar{\tau}_L) + \varepsilon_L \rho_L \vec{g} - \vec{F}_{drag,L} + \vec{S}_{GL} + \vec{F}_{disp,L} \quad (3)$$

$$225 \quad \frac{\partial}{\partial t} (\varepsilon_G \rho_G \vec{u}_G) + \nabla \cdot (\varepsilon_G \rho_G \vec{u}_G \vec{u}_G) = -\varepsilon_G \nabla P + \nabla \cdot (\varepsilon \bar{\tau}_G) - \vec{F}_{drag,G} + \varepsilon_G \rho_G \vec{g} - \vec{S}_{GL} + \vec{F}_{disp,G} \quad (4)$$

226 where P is the pressure, P_c is the capillary pressure, which is produced by the difference
 227 in the pressures across the fluid interface and only included in the liquid phase
 228 momentum equation, model details please refer to (Zhang et al., 2022a), $\bar{\tau}_i$ is the stress
 229 tensor, $\vec{F}_{drag,i}$ is the drag force between the phase and packing, \vec{S}_{GL} is the interfacial
 230 force between the gas and liquid, and $\vec{F}_{disp,i}$ is the mechanical dispersion force.

231 The species transport equation and energy equation are shown as follows:

$$232 \quad \frac{\partial(\varepsilon_i \rho_i \bar{u}_i Y_{ij})}{\partial t} + \nabla \cdot (\varepsilon_i \rho_i \bar{u}_i Y_{ij}) = -\nabla \cdot (\varepsilon_i \vec{J}_{ij}) + \varepsilon_i R_{ij} \quad (5)$$

$$233 \quad \frac{\partial(\varepsilon_i \rho_i E_i)}{\partial t} + \nabla \cdot (\varepsilon_i \bar{u}_i (\rho_i E_i + P)) = \varepsilon_i \nabla P \left(k_i \nabla T_i - \sum h_j \vec{J}_{ij} + (\bar{\tau}_{eff,ij} \cdot \bar{u}_i) \right) + \varepsilon_i Q_{h,i} + \varepsilon_i S_{h,i} \quad (6)$$

234 where Y_{ij} is the mass fraction of the species j in i th phase, such as the CO_2 in the gas
 235 phase or MEA in the liquid phase (j = species), R_{ij} is the production of the species j by
 236 the reaction, such as MEACOO^- ($\text{HOC}_2\text{H}_4\text{NHCOO}^-$), \vec{J}_{ij} is the mass diffusion flux, E_i
 237 is the total energy, k_i is the thermal conductivity, h_j is the enthalpy of the species,
 238 $\bar{\tau}_{eff,ij}$ is the effective shear tensor, $Q_{h,i}$ is the transferred heat between the phases, see
 239 Section 2.6, and $S_{h,i}$ is the heat of chemical reaction, which may be obtained by $S_{h,i} =$
 240 $-\sum_j h_j R_{ij}$ (Mardani and Mahalegi, 2019).

241 **2.3 Effective interfacial area and forces**

242 Since the effective gas-liquid interfacial area and forces are quite different between the
 243 packing zone and outer cavity zone, the effective interfacial area correlation and force
 244 models for these two zones should be separately developed. In the packing region,
 245 models for the interfacial, drag and dispersion (including capillary and mechanical
 246 dispersion) forces and the effective interfacial area for the non-uniform multiphase flow
 247 in the RPB have been introduced in the previous work (Zhang et al., 2022a). It is worth
 248 mentioning that the angle between the flow direction and the bed axis (θ) included in
 249 the porous resistance model is set as 10° , which is determined by a commonly used
 250 specification of the wire meshes with the wire mesh diameter and the centre distance
 251 between the wire mesh being 0.6 and 3.5 mm, respectively. Also, this value has been
 252 employed in the work of Zhang et al. (2022c). Furthermore, when the dynamic contact
 253 angle (β) in the effective interfacial area model is set as 12° , the modelled fractional
 254 effective interfacial area for the cases simulated in Section 3 is in a reasonable range of
 255 0.29-0.52 (Luo et al., 2017; Yang et al., 2011) and the CO_2 capture coefficient matches

256 the experimental data well. In this section, only the effective interfacial area and force
 257 model for the outer cavity zone are discussed.

258 2.3.1 Effective interfacial area model

259 At present, no correlation of the effective interfacial area has been introduced for the
 260 outer cavity zone of the RPBs in the published papers. For a spherical bubble or droplet,
 261 the algebraic interfacial area concentration models are derived from the surface area to
 262 volume ratio ($A_p = \frac{\pi d_p^2}{\frac{1}{6}\pi d_p^3} = \frac{6}{d_p}$). When using the Eulerian multiphase model, a
 263 commonly used equation, as given in Eq. 7, could be utilized to estimate the effective
 264 interfacial area, which has been built in Fluent ia-symmetric model.

$$265 \quad A_{e,o} = \frac{6\varepsilon_G\varepsilon_L}{d_{p,o}} \quad (7)$$

266 where $A_{e,o}$ is the effective interfacial area in the outer cavity zone, $d_{p,o}$ is the average
 267 diameter of the liquid droplets in the outer cavity zone. The ia-symmetric model not
 268 only considers the gas and liquid volume fraction, but also takes into account the liquid
 269 diameter. In the experimental work of Sang et al. (2017b), they concluded that the liquid
 270 exists in the outer cavity zone mainly in the form of droplets and a correlation of the
 271 average droplet diameter in the outer cavity zone has been proposed as follows (Sang
 272 et al., 2017b):

$$273 \quad d_{p,o} = 0.042We^{-0.272}Re^{0.068}\left(\frac{u_0}{\omega r_o}\right)^{0.098}r_o \quad (8)$$

$$274 \quad We = \frac{\rho\omega^2r_o^3}{\sigma}; Re = \frac{\rho\omega r_o^2}{\mu}; q = \frac{u_0}{\omega r_o}; u_0 = \frac{Q_L}{2\pi r_i h} \quad (9)$$

275 where We is the Weber number, Re is the Reynolds number with outer packing radius
 276 as the characteristic linear dimension, ω is the angular velocity, Q_L is the liquid
 277 volumetric flow rate, σ is the liquid surface tension, μ is the liquid viscosity, and r_o , r_i
 278 and h are the outer radius, inner radius and height of the packing, respectively.

279 Because the liquid on the casing wall exists in the form of the liquid film instead of the
 280 liquid droplet, Eq. (7)-(9), which are developed for the liquid droplets in the outer cavity
 281 space, cannot be used for estimating the contact area between the gas and the liquid
 282 film near the casing wall. If the casing wall surface is fully covered by the liquid film,
 283 then the effective interfacial area in the cell next to the casing wall ($a_{e,OW}$) is estimated
 284 as follows:

$$285 \quad A_{e,OW} = \frac{A_{cell,OW}}{V_{cell,OW}} = \frac{1}{\Delta x_{OW}} \quad (10)$$

286 where $A_{cell,OW}$ and $V_{cell,OW}$ are the wall area surface and volume in the computational
 287 cell next to the casing wall, respectively, and Δx_{OW} is the radial length of the cell
 288 normal to the casing wall. For the case studied in this paper, the radial length Δx_{OW} is
 289 0.005 m, thus, the effective interfacial area near the casing wall is no more than 200
 290 m^2/m^3 . According to Equation (10), it appears that the maximum effective interfacial
 291 area ($A_{e,OW}$) depends on Δx_{OW} . Nevertheless, it should be noted that the unit of the
 292 interfacial area ($A_{e,OW}$) is m^2/m^3 , and therefore it is more suitable to describe $A_{e,OW}$
 293 as the interfacial area concentration. The real physical interfacial area (m^2) could be
 294 obtained by $A_{OW} = A_{e,OW}V_{cell,OW} = A_{cell,OW}$, which is independent on the first-layer
 295 mesh size at the wall.

296 In addition, within the packing bed, the method given by (Lu et al., 2019) has been
 297 employed to calculate the effective interfacial area in the cell next to the packing wall
 298 as follows:

$$299 \quad A_{e,PW} = \frac{A_{PW}f_e + a_s f_e V_{cell,PW}}{V_{cell,PW}} = \left(\frac{1}{\Delta x_{PW}} + a_s \right) f_e \quad (11)$$

300 where $A_{cell,PW}$ and $V_{cell,PW}$ are the wall surface area and volume of the cell next to the
 301 packing wall, respectively, Δx_{PW} is the radial length of the cell normal to the packing
 302 wall, a_s is the specific area of the packing, f_e is the fractional effective interfacial area
 303 in the packing, which is the ratio of the wet area to the total packing area ($\frac{A_{e,P}}{a_s}$), and f_e
 304 of the packing walls is considered to be the same as that in the packing region.

305 2.3.2 Interfacial force model

306 The interfacial force is the only force for the liquid and gas phases in the outer cavity
 307 zone. It is due to the momentum exchange between the gas phase and liquid phase and
 308 this force is usually presented as an exchange coefficient in Ansys Fluent. There are
 309 many built-in models to calculate the interfacial exchange coefficient and a model,
 310 named universal drag laws (Ishii and Zuber, 1979), has been designed for the bubble-
 311 liquid and droplet-gas flows that could be utilized for modelling the interfacial
 312 exchange coefficient in the outer cavity zone, namely

$$313 \quad K_{GL} = \frac{\varepsilon_G \varepsilon_L \rho_L f}{\tau_L} \quad (12)$$

$$314 \quad \tau_L = \frac{\rho_L d_p^2}{18 \mu_G} \quad (13)$$

$$315 \quad f = \frac{C_D Re}{24} \quad (14)$$

$$316 \quad Re' = \frac{\rho_G |\vec{u}_G - \vec{u}_L| d_p}{\mu_e} \quad (15)$$

$$317 \quad C_D = \begin{cases} \frac{24}{Re'} & Re' < 1 \\ \frac{24}{Re'} (1 + 0.1 Re'^{0.75}) & 1 \leq Re' \leq 1000 \\ \frac{2}{3} \left(\frac{d_p}{\lambda_{RT}} \right) \left\{ \frac{1 + 17.67 f^{*6/7}}{18.67 f^*} \right\}^2 & Re' > 1000 \end{cases} \quad (16)$$

$$318 \quad f^* = (1 - \varepsilon_L)^3; \mu_e = \frac{\mu_G}{(1 - \varepsilon_L)^{2.5}}; \lambda_{RT} = \left(\frac{\sigma}{g \Delta \rho_{GL}} \right)^{0.5} \quad (17)$$

319 where K_{GL} is the interfacial exchange coefficient, f and f^* are the drag functions, τ_L is
 320 the particulate relaxation time, C_D is the drag coefficient, Re' is the relative Reynolds
 321 number, μ_e is the effective viscosity, λ_{RT} is the Rayleigh-Taylor instability wavelength,
 322 g is the gravity, and $\Delta \rho_{GL}$ is the absolute value of the density difference between liquid
 323 and gas phases.

324 2.4 Mass transfer rate model

325 The two-film theory has by far been the most popular and useful theory for dealing with
326 the CO₂ mass transfer among the phases (Gbadago et al., 2020; Kim et al., 2016, 2017;
327 Pham et al., 2015). Generally, based on the two-film model and the Henry law, the
328 overall mass transfer coefficient and enhancement factor are applied to model the CO₂
329 mass transfer between the gas phase and MEA solution, which can be expressed as
330 follows:

$$331 \quad N_{CO_2} = K_L A_e (C_{CO_2}^* - C_{CO_2}) \quad (18)$$

$$332 \quad \frac{1}{K_L} = \frac{RT}{H_{CO_2-MEA} k_G} + \frac{1}{E k_L} \quad (19)$$

333 where N_{CO_2} is the CO₂ mass transferred through the gas-liquid interface, K_L is the
334 overall mass transfer coefficient, k_G and k_L are the mass transfer coefficients in the gas
335 and liquid phases, respectively, R is the gas constant, T is the temperature, H_{CO_2-MEA} is
336 the Henry constant, E is the enhancement factor, which is defined as the ratio of the
337 absorption rate with and without chemical reaction (Sebastia-Saez et al., 2015), and
338 $C_{CO_2}^*$ and C_{CO_2} are the CO₂ concentrations on the surface of the liquid and in the liquid
339 bulk flow, respectively. In particular, H_{CO_2-MEA} , E and $C_{CO_2}^*$ have been adequately
340 illustrated in many works (Borhani et al., 2018; Lu et al., 2019; Pham et al., 2015), so
341 that these factors are expressed in Table 1 along with some other parameters.

342 Since the CO₂ mass transfer resistance between the gas and liquid is dominated by the
343 liquid side, the CO₂ mass transfer resistance in the gas side is neglected. Therefore, Eq.
344 (19) may be simplified as the following equation:

$$345 \quad \frac{1}{K_L} = \frac{1}{E k_L} \quad (20)$$

346 According to the film theory expression, the mass transfer coefficient is given as
347 follows:

$$348 \quad k_L = \frac{D_{L,CO_2}}{\delta} \quad (21)$$

349 where D_{L,CO_2} is the diffusivity of the CO₂ in the liquid phase, which could be calculated

350 by the N₂O analogy method (Liu et al., 2006) and given in Table 1, and δ is the diffusion
 351 layer thickness for the mass transfer.

352 Guo et al. (1997) and Munjal et al. (1989) proposed the correlations for predicting the
 353 film thickness for the RPBs, which are shown in Eq. (22) and (23), respectively,

$$354 \quad \delta = 4.20 \times 10^8 \frac{Q_L}{2\pi r h} \frac{\nu_L}{\omega^2 r} \quad (22)$$

$$355 \quad \delta = \left(3 \left(\frac{Q_L}{2\pi r} \right) \frac{\nu_L}{r \omega^2} \right)^{\frac{1}{3}} \quad (23)$$

356 where r is the radial distance, ν_L is the kinematic viscosity, and ω is the rotational
 357 speed.

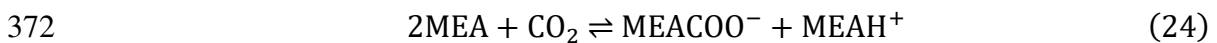
358 For the operating condition in the simulations, the enhancement factor (E) is
 359 approximately equal to the Hatta number (Ha) when the reaction is in the fast reaction
 360 regime ($5 \leq Ha \ll E_i$) due to the high MEA concentration or the small CO₂ partial
 361 pressure given by Eq. (T-7) (Jiru and Eimer, 2013; Ying and Eimer, 2013). As a result,

$$362 \quad K_L = Ek_L \approx H a k_L = \sqrt{\frac{k_2 D_{L,CO_2} C_{L,MEA}}{(k_L)^2}} k_L = \sqrt{k_2 D_{L,CO_2} C_{L,MEA}}, \text{ which means that the}$$

363 overall mass transfer rate is almost independent of the film layer thickness. In addition,
 364 this conclusion is consistent with that drawn by Lu et al. (2019). Therefore, either of
 365 these equations could be utilized in this paper and Eq. (22) is selected in this paper since
 366 this equation is derived from the wire mesh (Guo et al., 1997), which is the experimental
 367 packing material used in the reference paper (Kolawole, 2019).

368 **2.5 Chemical reaction rate**

369 The CO₂-MEA absorption system can be expressed by the zwitterion mechanism
 370 (Danckwerts, 1979), and the overall reaction between CO₂ and MEA is described as
 371 follows (Borhani et al., 2018):



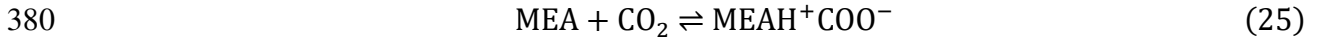
373 The above reaction could be separated into two-step reactions. According to the
 374 zwitterion mechanism, an intermediate product - the zwitterion ion (MEAH⁺COO⁻) is
 375 generated in the CO₂-MEA absorption process (Ebadi Amooghin et al., 2018). Then,

Table 1 Some of the correlations utilized in the current study.

Equation name	Equations	Eq. No.	Reference
Diffusivity of the MEA	$D_{L,MEA} = \exp\left(-13.275 - \frac{2198.3}{T} - 0.078142C_{L,MEA}\right)$	T-1	(Snijder et al., 1993)
Diffusivity of CO ₂ in the MEA solution	$D_{L,CO_2} = D_{L,N_2O} \frac{D_{w,CO_2}}{D_{w,N_2O}}$	T-2	(Liu et al., 2006)
	$D_{w,CO_2} = 2.35 \times 10^{-6} \exp\left(-\frac{2119}{T}\right)$	T-3	
	$D_{w,N_2O} = 5.07 \times 10^{-6} \exp\left(-\frac{2371}{T}\right)$	T-4	
	$D_{L,N_2O} = 5.07 + 0.865C_{MEA} + 0.278C_{MEA}^2 \exp\left(-\frac{2371 - 93.4C_{MEA}}{T}\right)$	T-5	
Enhancement factor	$E = 1 + ((E_i - 1)^{-1.35} + (E_1 - 1)^{-1.35})^{-\frac{1}{1.35}}$	T-6	(Wellek et al., 1978)
	$E_i = 1 + \frac{D_{L,MEA}C_{L,MEA}}{2D_{L,CO_2}C_{L,CO_2}}$	T-7	(Sebastia-Saez et al., 2015)
	$E_1 = \frac{Ha}{\tanh(Ha)}$	T-8	
	$Ha = \sqrt{\frac{k_2 D_{L,CO_2} C_{L,MEA}}{(k_L)^2}}$	T-9	
CO ₂ saturation concentration	$C_{L,CO_2}^* = M_{CO_2} \frac{P_{CO_2}}{H_{CO_2-L}}$	T-10	(Penttilä et al., 2011)
Henry constant of the CO ₂	$H_{CO_2-L} = H_{N_2O-L} \left(\frac{H_{CO_2-H_2O}}{H_{N_2O-H_2O}} \right)$	T-11	(Penttilä et al., 2011)

	$H_{CO_2-H_2O} = \exp\left(145.369 - \frac{8172.355}{T} - 19.303\right)$	T-12	
	$H_{N_2O-H_2O} = \exp\left(158.245 - \frac{9048.596}{T} - 20.86 \ln T - 0.00252\right)$	T-13	
	$H_{N_2O-MEA} = \exp\left(-9172.5 + \frac{39.598}{T}\right)$	T-14	
	$H_{N_2O-L} = H_{N_2O-H_2O}C_{L,H_2O} + H_{N_2O-MEA}C_{L,MEA} + 3524641.533(C_{L,H_2O}C_{L,MEA})^2 \left(1 - \frac{T}{324.718}\right) \exp(-13.219C_{L,MEA})$	T-15	
Viscosity of the MEA solution	$\mu_{0.3} = 0.3083 - 0.00262T + 7.4882 \times 10^{-6}T^2 - 7.17293 \times 10^9T^3$ $\mu_{0.9} = 4.37711 - 0.03776T + 1.08945 \times 10^{-4}T^2 - 1.05031 \times 10^7T^3$	T-16	(Amundsen et al., 2009)
Surface tension of the MEA solution	$\sigma_{H_2O} = 0.18548\left(1 - \frac{T}{647.13}\right)^{(2.717+3.554(\frac{T}{647.13})+2.047(\frac{T}{647.13})^2)}$	T-17	(Vázquez et al., 1997)
	$\sigma_{MEA} = 0.09945\left(1 - \frac{T}{614.45}\right)^{(1.067)}$	T-18	
	$\sigma_L = \sigma_{MEA} + \frac{2.129(1 - y_{MEA})}{(1 + (2.129 - (1 - y_{MEA})))}(\sigma_{H_2O} - \sigma_{MEA})$	T-19	

378 this zwitterion transfers into carbamate (MEACOO⁻) via deprotonation by a base (MEA)
 379 (Moftakhari Sharifzadeh et al., 2016), and the two-step reactions are as follows:



382 Reaction (25) has a finite reaction rate and could be performed in Ansys Fluent through source
 383 term UDFs. Reaction (25) (i.e. carbamate formation) is the controlling step and presents the
 384 whole reaction kinetics (Sebastia-Saez et al., 2014; Sebastia-Saez et al., 2015). It can be
 385 regarded as a second-order irreversible reaction, and the reaction rate is expressed as follows
 386 (Luo et al., 2012):

$$387 \quad r_{\text{CO}_2} = -k_2[\text{MEA}][\text{CO}_2] \quad (27)$$

388 where k_2 is the reaction rate constant. Different k_2 equations have been proposed for the CO₂-
 389 MEA absorption system based on various experimental conditions (Hikita et al., 1977; Luo et
 390 al., 2015; Versteeg and van Swaaij, 1988; Ying and Eimer, 2013).

391 Among them, the reaction rate reported by Versteeg et al. (1996) has been validated by Ying
 392 and Eimer (2013) and used in many studies (Dashliborun et al., 2019; Ying and Eimer, 2013),
 393 which is given as follows:

$$394 \quad k_2 = 4.4 \times 10^{11} \exp\left(-\frac{5400}{T}\right) \quad (28)$$

395 **2.6 Heat transfer rate**

396 The amount of the transferred heat between the gas and liquid phases is a function of the
 397 temperature difference and the interfacial area:

$$398 \quad Q_{h,GL} = h_{GL}A_{GL}(T_L - T_G) \quad (29)$$

399 where $Q_{h,GL}$ is the transferred heat between the two phases, and h_{GL} is the heat transfer rate.

400 A commonly used model- the Hughmark model (Hughmark, 1967) was utilized to calculate
 401 the heat transfer rate, namely

$$402 \quad h_{GL} = \frac{\kappa_G Nu_L}{d_p} \quad (30)$$

$$Nu_L = \begin{cases} 2.0 + 0.6Re_L^{\frac{1}{2}}Pr_G^{\frac{1}{3}} & 0 \leq Re_L < 776.06, 0 \leq Pr_G < 250 \\ 2.0 + 0.27Re_L^{0.62}Pr_G^{\frac{1}{3}} & 776.06 \leq Re_L, 0 \leq Pr_G < 250 \end{cases} \quad (31)$$

$$Pr_G = \frac{C_{pG}\mu_G}{\kappa_G} \quad (32)$$

where κ_G is the thermal conductivity of the gas phase, Nu_L is the Nusselt number of the liquid phase, Pr_G is the Prandtl number of the gas phase, and C_{pG} is the specific heat. The heat transfer in the RPB is determined by the heat transfer rate, which is reversely proportional to the liquid droplet diameter according to Eq. (30). The larger the liquid droplet diameter, the lower is the heat transfer rate, which means that more heat could be retained in the liquid phase. As a result, the liquid temperature changes and the CO₂ capture performance is changed. Therefore, the liquid droplet diameter is a critical parameter for the CO₂ absorption, and this parameter in the packing region and outer cavity zone should be carefully modelled. In particular, the diameter of the liquid droplets in the outer cavity zone (Sang et al., 2017b) are shown in Eq. (8) and the diameter in the packing region has been regressed as two correlations based on the same set of the experimental data (Zhang, 1996), namely (Guo et al., 1997; Yi et al., 2009):

$$d_{p,P} = 0.7284 \left(\frac{\sigma}{\omega^2 r \rho} \right)^{0.5} \quad (33)$$

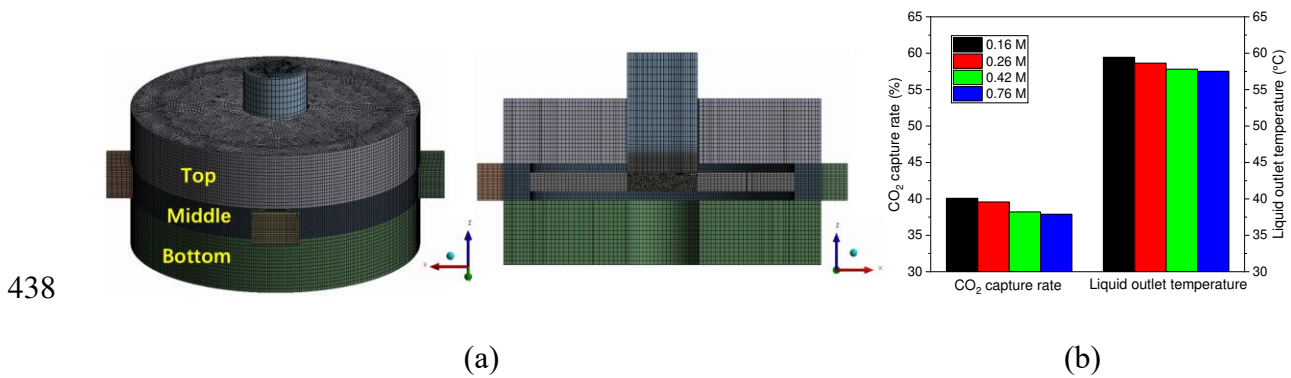
$$d_{p,P} = 12.84 \left(\frac{\sigma}{\omega^2 r \rho} \right)^{0.630} u^{0.201} \quad (34)$$

where r is the radial coordinate of the packing from the centre, and u is the liquid flow rate per unit area, which is only included in Eq. (34). Since the liquid diameter is influenced by the liquid flow rate u , Eq. (34) could more accurately predict the diameter of the liquid droplets in the packing region. Thus, Eq. (34) has been utilized in this work.

2.7 CFD model setup

Figure 3(a) shows the outside and inside mesh layouts of the 3D pilot-scale RPB model generated by the ANSYS Mesh. In addition to the packing region, the flow characteristics change evidently in the middle of the outer cavity zone and the inner cavity zone, the meshes

427 in these areas are finer compared with the top and bottom cavity zones. To reduce the number
 428 of cells and improve the calculation accuracy (Zhang et al., 2022b), the structured hexahedral
 429 grids were generated in the whole RPB except in the inner cavity zone. Various numbers of the
 430 grid cell were tested with total cells of 0.16 M, 0.26 M, 0.42 M and 0.76 M in order to obtain
 431 a mesh independent solution, which is shown in Figure 3(b). According to Figure 3(b), until a
 432 grid of 0.42 M cells, the CO₂ capture rate and liquid outlet temperature reached stable.
 433 the grid of 0.42 M cells, including 0.05 M cells in the packing zone and 0.28 M cells in the
 434 outer cavity zone, was applied. The minimum and largest mesh volumes are 1.4×10^{-10} and
 435 $3.3 \times 10^{-7} \text{ m}^3$, respectively, and the average skewness and element quality are 0.11 and 0.85,
 436 respectively. It should be noted that the mesh number used in this study is adequate only for
 437 this case study. A solid mesh study may be required for other applications.



439 **Figure 3.** (a) Schematic of the mesh in the 3D model and (b) predicted CO₂ capture rate and
 440 liquid outlet temperature using different grids.

441 The 3D transient simulations have been performed using the ANSYS Fluent 2021 R1 in a
 442 double precision mode based on the High Performance Computing cluster in the University of
 443 Sheffield. Various UDFs have been developed for defining the properties of the MEA solution,
 444 implementing the extra forces in the momentum equations, and calculating the transferred CO₂
 445 in the mass equations, etc. The Phase Coupled SIMPLE method was applied, and the pressure
 446 equations were discretized by the second-order scheme. The pressure-based method and the
 447 absolute velocity formulation have been utilized. The time step was set as 5×10^{-4} s, and a
 448 maximum of 35 iterations was employed per time step, and the convergence tolerance was

449 1×10^{-5} . The simulation case can be assumed to be the pseudo steady state when the CO₂
450 concentration at the gas outlet and the liquid outlet temperature were reduced to within 1% in
451 ten seconds, and the governing equations' residuals were less than 5×10^{-5} .

452 For the RPB model, the realizable k- ϵ turbulence model is more suitable for implementation in
453 the RPB than the standard k- ϵ model, and the reasons are illustrated in our previous paper
454 (Zhang et al., 2022a). In addition, this turbulence model has been frequently used for the fluid
455 flow simulations in RPBs (Wang et al., 2020; Wu et al., 2018; Yang et al., 2010), thus, this
456 model has been employed in this study.

457 The properties of the gas and liquid phases are shown in (Xie, 2019). The gas phase consists
458 of CO₂ and air, and the liquid phase is composed of CO₂, MEA (HOC₂H₄NH₂), MEAH⁺
459 (HOC₂H₄NH₃⁺), MEACOO⁻ (HOC₂H₄NHCOO⁻) and water. The correlations of the surface
460 tension and viscosity for the MEA solution are present in Table 1 (Amundsen et al., 2009;
461 Vázquez et al., 1997). In addition, the liquid density, thermal conductivity and specific heat of
462 the two phases are evaluated by the mixing law based on every species in each phase. The
463 operating conditions are shown in Table 2.

464 **Table 2.** The operational conditions of the experiments (Kolawole, 2019).

MEA concentration (wt%)	Rotational speed (rpm)	Gas flow rate (kg/h)	L/G mass ratio
30, 50, 70	600, 850, 1150	42	1.3-3.5

465 In the experiment conducted by Kolawole (2019), all MEA solutions were preloaded to 0.1 mol
466 CO₂/mol MEA before the MEA solution was fed into the RPB. The same MEA loading is
467 employed in the CFD model. The gas flow rate is fixed at 42 m³/h with a temperature of 40 °C
468 according to the experimental settings and the gas outlet boundary is set as the pressure-outlet
469 with a zero gauge pressure. In terms of the reverse flow at the gas outlet boundary, the CO₂
470 fraction on the inner packing surface is used as the CO₂ fraction in the gas outlet, which has
471 been performed iteratively. In addition, different L/G mass ratios, e.g. L/G = 2.7, 3.1, 3.5 for
472 30% MEA, are tested in the model. Accordingly, the liquid inlet velocity ranges from 0.20 to
473 0.54 m/s with a temperature fixed at 40 °C and the liquid disappears after entering the liquid

474 elimination zone. The gravity force is set as 9.8 m/s^2 and in the -Y direction. The rotating speeds
475 employed in the experiment, including 600, 850 and 1150 rpm are used in the simulations. The
476 sliding model has been performed to achieve the motion of the packing. The wall boundaries
477 have been set as no slip and adiabatic.

478 **3. Results and discussion**

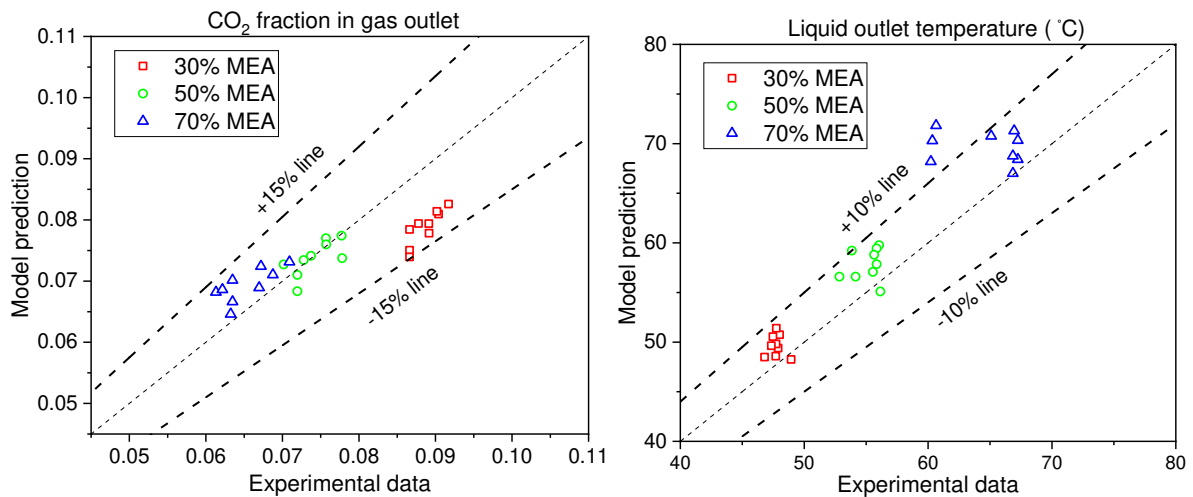
479 In order to validate the CFD model developed, 27 simulation cases have been compared with
480 various experimental cases, and all the major variables have been critically analyzed. After
481 that, the effects of various operational parameters on the mass transfer process have been
482 investigated. In addition, the ratio of the CO_2 captured in the outer cavity zone to that in the
483 full-scale RPB has been investigated.

484 **3.1 Validation and the distribution of variables**

485 For validating the developed 3D RPB model, the obtained CO_2 mole fraction in the gas outlet
486 and the liquid temperature at the liquid outlet, which could reflect the mass and heat transfer
487 performance, are compared with the experimental results in the Kolawole thesis (Kolawole,
488 2019). The full operating conditions are listed in Table 2. Although the flow dynamics may be
489 compared via the liquid holdup, unfortunately, this parameter has not been measured in the
490 experimental investigation.

491 Figure 4 presented a comparison of the CO_2 fractions in the gas outlet and the liquid outlet
492 temperatures obtained from the experiments and simulations under various operating
493 conditions, including the MEA concentration, rotational speed and the L/G mass ratio. From
494 Figure 4(a), it is observed that all relative deviations of the CO_2 mole fraction are within 15%.
495 And the CO_2 fraction for the 50% MEA solution has a better agreement with the experimental
496 data, whose relative deviations are less than 6%. According to Figure 4(b), most of the relative
497 deviations of the liquid outlet temperature are less than 10%. Only a few points for the 70%
498 MEA solution with a lower rotational speed are larger than 10%. The difference between the
499 simulations and experimental results may mainly result from the difficulties and uncertainties
500 in measuring the variables since the experiments have been conducted in a relatively small

501 reactor (Lu et al., 2019). In addition, the employed empirical correlations may not be suitable
502 for this RPB model since these correlations were derived from different RPB sizes, operating
503 conditions, etc. Also, the phase properties may not be able to be accurately evaluated as the
504 temperature and CO₂ loading increase. Furthermore, the employed mesh grid with only 0.42
505 M cells could be a possible reason for the errors between the simulations and experiment
506 results. It can be found that the average absolute relative deviations in the CO₂ fraction and
507 liquid outlet temperature are smaller than 7% and 6%, respectively, thus indicating that the
508 effective interfacial area, mass and heat transfer models could accurately describe the
509 thermodynamics and mass transfer processes within the RPB. Furthermore, the same sub-
510 models, such as the hydrodynamics, including the interfacial, drag, capillary pressure and
511 mechanical dispersion force models, have been verified in our previous paper (Zhang et al.,
512 2022a) using a smaller RPB model. Therefore, it can be concluded that the developed 3D RPB
513 model gives a good match to the experimental results, and it can be employed to investigate
514 the CO₂ capture process within this RPB. It should be noted, although the overall CFD results
515 agree with the experimental data for this RPB, further validation may be required when
516 applying the sub-models to other RPBs, in particular when different packings are employed.



517
518 **Figure 4.** Comparison of the (a) CO₂ mole fraction in the gas outlet; (b) liquid outlet
519 temperature (Kolawole, 2019).

520 In addition, the distributions of some important variables are presented in order to analyze the

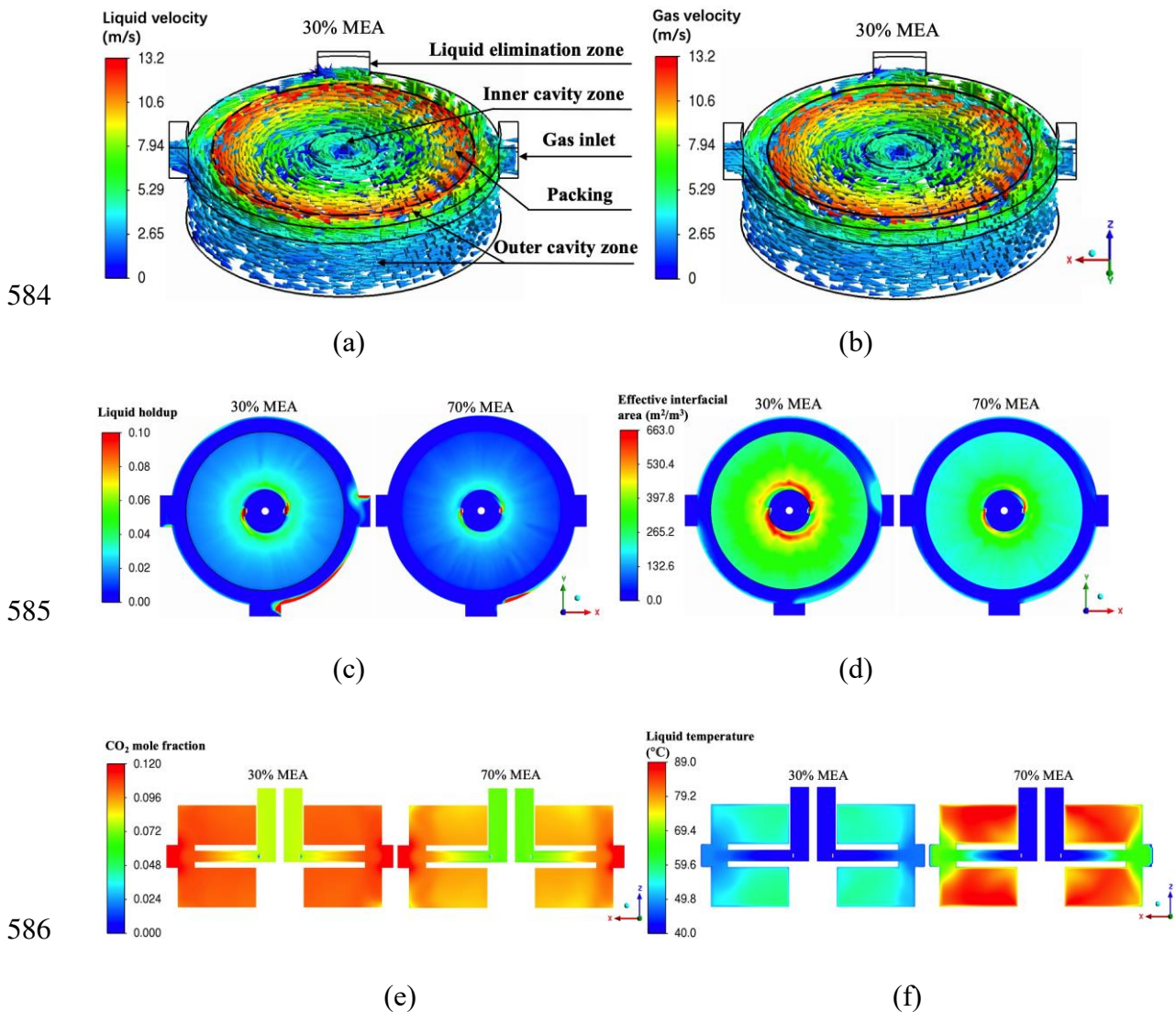
521 hydrodynamics, thermodynamics, and reaction-enhancement mass transfer within the RPB.
522 Figure 5 presents the predicted variables for the 30% and 70% MEA solutions. Also, the cases
523 are performed with the rotational speed being 850 rpm and the L/G ratios being 3.1 and 1.5 for
524 30% and 70% MEA solutions, respectively. Since the vectors of the gas and liquid velocities
525 are similar for the 30% and 70% MEA solutions, only the vectors for the 30% MEA solution
526 in half of the RPB cut from the central plane is presented in Figures 5(a) and 5(b) in order to
527 observe them clearly. Figure 5(a) shows that the liquid flows tangentially followed the
528 rotational bed due to the porous resistance and its velocity gradually increases on the way
529 outwards through the packing. Near the outer packing surface, the liquid velocity reaches a
530 maximum and then the liquid velocity suddenly reduces in the outer cavity zone due to the
531 interfacial force between the gas and liquid phases. In addition, the liquid also rotates with a
532 small velocity in the bottom outer cavity zone. From Figure 5(b), it is observed that the gas
533 phase that radially flows from the gas inlet, begins to rotate after entering the outer cavity zone
534 due to the effect of the rotational packing. Its motion is in rapid synchronization with the
535 rotating packing in the outer packing zone and it reacts with the MEA solution when passing
536 through the packing region. It is worth mentioning that although the vectors of the phase
537 velocity in Figures 5(a) and 5(b) are very similar, the radial flow directions of the gas and liquid
538 phases are opposite.

539 According to Figure 5(c), it is observed that the liquid holdup gradually reduces along the radial
540 direction in the packing zone and this is due to the increasing flow space and the increasing
541 liquid radial velocity due to the centrifugal force, which also can be seen in Figure 6(a).
542 Although the liquid holdup data is not available in the Kolawole's thesis, this phenomenon has
543 been accurately predicted by other simulation and experimental investigators (Burns et al.,
544 2000; Lu et al., 2018). After flowing out from the packing region, the liquid radial velocity
545 suddenly increases without the restriction of the porous resistance, thus causing the liquid
546 fraction to become quite small in the outer cavity zone. Subsequently, the liquid droplets collide
547 on the casing wall and flow downwards under the influence of the gravitational force. Thus,
548 the liquid with a higher volume fraction may be observed on the casing wall surface. Also, it

549 can be found that the liquid holdup for the 30% MEA solution is clearly larger than that for the
550 70% MEA solutions as the ratio of the liquid flow rate for 30% to 70% MEA is 2.1. However,
551 the ratio of the liquid holdup in the packing regions is about 1.6, which is smaller than the ratio
552 of the liquid flow rate of 2.1. The first reason is that the liquid viscosity for the 70% MEA is
553 larger, which means that the 70% MEA solution is more likely to adhere to the wire mesh
554 surface (Xie et al., 2019). Furthermore, the liquid radial velocity increases and the liquid
555 residence time reduces when more of the liquid phase enters the packing zone, which allows
556 the 30% MEA solution to faster escape into the packing. These factors lead to a higher liquid
557 holdup for the 30% MEA solution and thus the ratio of the liquid holdup is not exactly equal
558 to the ratio of the liquid flow rate. As a result, the effective interfacial area for the 30% MEA
559 solution is larger, see Figure 5(c), since more liquid is attached to the wire mesh packing surface,
560 or it is split into numerous small droplets in the packing zone (Liu et al., 2019; Yang et al.,
561 2015a). This means that the contact area between the gas and liquid phases is larger, thus
562 indicating that the 30% MEA solution has more chance to interact with the CO₂.

563 Although the liquid holdup and contact area are larger for the 30% MEA solution resulting
564 from the larger liquid flow rate, the CO₂ fraction in the gas outlet is higher by comparing the
565 two illustrations in Figure 5(e). This is because the CO₂ capture performance is not only related
566 to the liquid holdup and contact area, but also it is affected by some other parameters, such as
567 the MEA concentration, liquid temperature and residence time. Clearly the MEA concentration
568 is the dominant factor in determining the CO₂ capture process. For the CO₂-MEA absorption
569 system, the heat will be released when the CO₂ is captured in the MEA solution, thus leading
570 to an increasing liquid temperature. Simultaneously, some heat is taken away by the opposing
571 gas flow via the heat transfer and the rest of the heat remains in the liquid phase. This could be
572 the reason why the liquid temperature for the 70% MEA solution is higher than that for the 30%
573 MEA solution, see Figure 5(f), because more active MEA participates in the reaction and
574 releases more heat to the already smaller flow rate of the liquid phase (Vaewhongs et al., 2020).
575 This phenomenon is more clearer in the top and bottom outer cavity zones, where the liquid
576 fraction is quite low and the liquid temperature could be even up to 89 °C. In return, the

577 chemical reaction is faster when the phase temperature is higher. In addition, the phase
 578 temperature could also influence the physical properties, including the viscosity and density,
 579 which further has an impact on the flow dynamics, such as the liquid holdup and residence
 580 time. Thus, this indicates that the liquid holdup, CO₂ capture rate, and liquid temperature could
 581 affect each other in this system. The predicted variables can be made accurate and stable only
 582 when all the parameters related to the flow dynamics, mass and heat transfer are set properly
 583 and these processes reach a balance within the whole RPB.



587 **Figure 5.** Vectors of the (a) liquid velocity; (b) gas velocity; contours of the (c) liquid holdup;
 588 (d) effective interfacial area on the planes $z = 0.01$; (e) CO₂ fraction in the gas phase; and (f)
 589 liquid temperature on the planes $y=0$ for 30% and 70 % MEA.

590 3.2 Mass transfer in different zones in the RPB

591 As we know, the CO₂ absorption within a RPB mainly occurs in two zones: packing and outer
592 cavity zones. Based on the newly developed 3D full-scale RPB model, where the CO₂ capture
593 process in the packing and outer cavity zones is separately analyzed by using the CFD method.

594 Taking the 50% MEA solution with a L/G ratio of 2.1 in Section 3.1 as an example, Figures
595 6(a), 6(b) and 6(c) show the circumferentially averaged liquid holdup and effective interfacial
596 area (per unit, m²/m³), liquid velocity, as well as the CO₂ mole fraction in the gas phase and
597 the liquid temperature along the radial direction, respectively. For $r_i \leq r \leq r_o$, these
598 parameters are only circumferentially averaged in the packing region rather than including the
599 top and bottom cavity zones. When flowing into the inner packing zone, with a radial velocity
600 only, the liquid violently collides with the rotational packing and it is quickly dispersed, thus
601 generating numerous very small tiny droplets, or forming thin films on the wire mesh surface.
602 Due to the small flow space in the inner periphery of the packing, a large effective interfacial
603 area could be observed in Figure 6(a), which is defined as the “end-effect zone” - an area where
604 excellent micromixing occurs due to the strong interaction and liquid dispersion (Cortes Garcia
605 et al., 2017; Esmacili et al., 2022). Although the phase surface renewal is fast and the effective
606 interfacial area is large in the end-effect zone, a sharp decrease in the CO₂ fraction is not
607 observed in Figure 6(c) due to the small local CO₂ fraction in the gas phase flow and the low
608 liquid temperature.

609 Soon after entering the bulk of the packing zone, the liquid starts to synchronize with the
610 rotating packing, and its tangential velocity almost coincides with the packing rotational
611 velocity in Figure 6(b). Consequently, the liquid dispersion is relatively weak, thus the effective
612 interfacial area decreases along with the radial position in Figure 6(a) (Zhang et al., 2017).
613 Since smaller droplets and thinner film are formed in this region, the liquid radial velocity
614 gradually decreases due to the larger interfacial and drag forces, which is shown in Figure 6(b).
615 In this packing region, most of the CO₂ is captured and a large amount of heat is released, and
616 this is due to the large interfacial surface area and the enhanced surface renewal of the phases

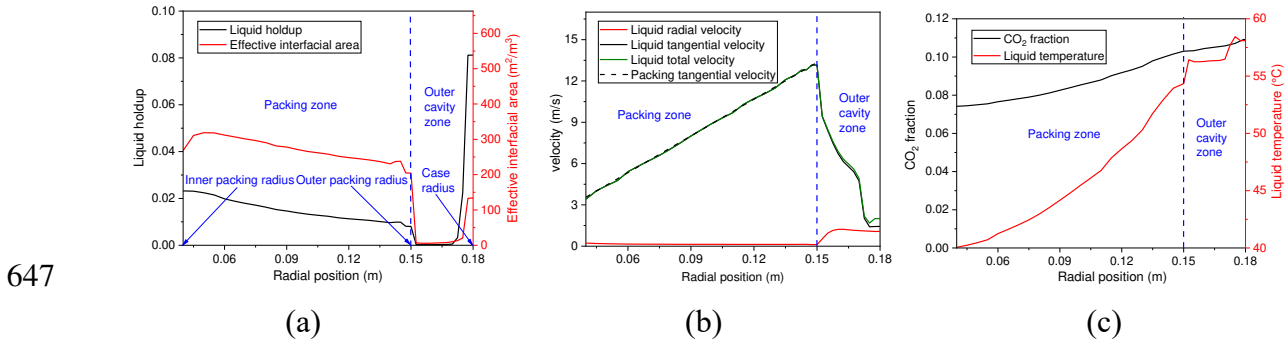
617 (Wang et al., 2021). Therefore, the CO₂ fraction in the gas phase reduces and the liquid
618 temperature increases on their way through the packing as shown in Figure 6(c).

619 After flowing out from the outer edge of the packing, the liquid droplets with large velocity
620 pass through the cavity space, causing a sudden decrease in the effective interfacial area and
621 liquid holdup in Figure 6(a). Without the interaction with the packing, the liquid flow direction
622 is almost unchanged in the outer cavity space, thus causing the liquid radial velocity component
623 to increase and the tangential velocity component to decrease as shown in Figure 6(b). As a
624 result, the difference between the gas and liquid radial velocity increases, which leads to an
625 increase in the interfacial force between the two phases and a decrease in the total liquid
626 velocity. In addition, an increase in the liquid temperature can be observed in Figure 6(c) due
627 to the high liquid temperature in the top and bottom regions of the outer cavity zone (see Figure
628 5(f)). Then the liquid droplets collide on the inner casing wall, generating the liquid film and a
629 large number of splashing droplets (Sang et al., 2017b). These phenomena have been modelled
630 well with a steep increase in the liquid holdup and effective interfacial area, see Figure 6(a).
631 Figure 6(c) shows that the magnitude of the decreased CO₂ fraction is small in the outer cavity
632 zone due to the low average effective interfacial area.

633 In general, the outer cavity zone's volume is much larger than the packing region. For instance,
634 the volume of the outer cavity zone to the packing region is about 11 for the present employed
635 RPB model. However, most of the CO₂ is captured in the packing region instead of the outer
636 cavity zone, which means that the larger the volume of the packing occupied in the fixed full-
637 scale RPB, the more effective is the CO₂ absorption.

638 Based on the validation cases under various operating conditions shown in Figure 4, it is found
639 that the outer cavity zone takes up 25%~40% of the total effective interfacial area and the effect
640 of the outer cavity zone on the mass transfer could range from 28% to 42% for this case study.
641 From the wide range of the contribution of the mass transfer and effective interfacial area in
642 the outer cavity zone, it is known that the CO₂ capture process in the packing and outer cavity
643 zones are quite different under various operating conditions. With the aim of providing some

644 new, useful and important suggestions for RPB design and scaling up, the contribution of the
 645 CO₂ removal and effective interfacial area in different zones has been investigated, which is
 646 presented in the following section.



648 **Figure 6.** The circumferentially averaged (a) liquid holdup and effective interfacial area; (b)
 649 liquid and rotating packing velocities and (c) CO₂ mole fraction in the gas phase and liquid
 650 temperature along the radial direction.

651 3.3 Effect of the operating parameters

652 Among the three MEA concentrations investigated, the relative deviation of the CO₂ fraction
 653 in the gas outlet for the 50% MEA solution is small, as discussed in Section 3.1. Thus, the 50%
 654 MEA solution is selected as the baseline case for discussion, and its operating conditions are
 655 listed in Table 3.

656 **Table 3.** The operational conditions for the baseline case.

MEA concentration (%)	L/G ratio	Rotational speed (rpm)	Liquid inlet temperature (°C)	Gas flow rate (kg/s)
50	2.1	850	40	42

657 For evaluating the CO₂ absorption performance in the full-scale RPB, the overall volumetric
 658 mass transfer coefficient ($K_L a_e$), a parameter that may determine the rate at which a gaseous
 659 compound (CO₂) can transfer from the gas phase to the MEA solution, is introduced along with
 660 the liquid temperature at the liquid outlet. The equation for $K_L a_e$ is shown in Eq. (35).
 661 $K_L a_e$ becomes larger when the CO₂ fraction in the gas outlet (y_{CO_2out}) is lower, indicating a
 662 better CO₂ capture performance.

663

$$K_L a_e = \frac{Q_G}{\pi(r_o^2 - r_i^2)Z} \ln\left(\frac{y_{CO_2in}}{y_{CO_2out}}\right) \quad (35)$$

664 The ratios of the captured CO₂ (r_c) and the effective interfacial area (r_e) in the outer cavity zone
 665 to those in the whole RPB are examined in order to analyze the mass transfer in different zones.
 666 The expression for r_c and r_e are given as follows:

$$667 \quad r_c = \frac{y_{i,CO_2} - y_{op,CO_2}}{y_{i,CO_2} - y_{o,CO_2}} \quad (36)$$

$$668 \quad r_e = \frac{\int_o a_e dV}{\int_p a_e dV + \int_o a_e dV} \quad (37)$$

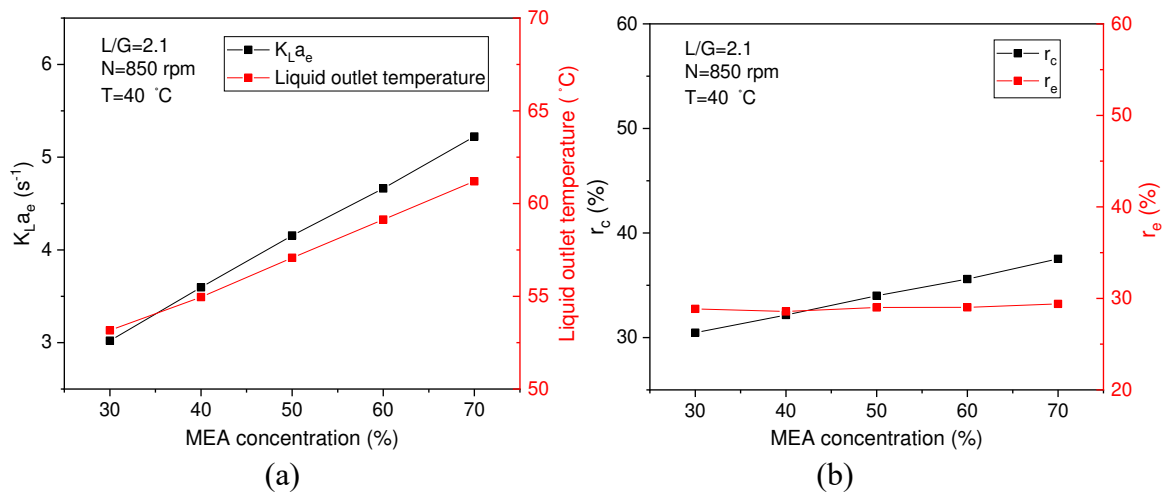
669 (i) Effect of the MEA concentration

670 It is known that the CO₂ capture efficiency increases when using the solution with a higher
 671 MEA concentration. In terms of the flow dynamics, the liquid phase can lead to early flooding
 672 or abnormal distribution in the packing of a CPB due to its large viscosity. However, one of the
 673 advantages of applying the RPB is that a higher MEA concentration solution with larger
 674 viscosity can fluently pass through the packing due to the higher gravitational environment.

675 Figure 7(a) illustrates the influence of the MEA concentration on the CO₂ absorption and
 676 thermodynamics in the RPB in terms of $K_L a_e$ and liquid temperature at the liquid outlet. In this
 677 case study, the MEA concentration varies from 30 to 70% while keeping all the other operating
 678 conditions consistent with the base case. It can be observed that the magnitude of the increased
 679 $K_L a_e$ and liquid outlet temperature significantly becomes larger when using a higher MEA
 680 concentration solution since the increased active MEA available in the solution could remove
 681 more CO₂ from the gas phase and more reaction heat would be generated at the same time
 682 (Qing et al., 2011). Although the solution with a higher MEA concentration favors for the CO₂
 683 absorption, the greater trend for corrosion, degradation and foaming should be carefully
 684 considered before the PCC industrial application.

685 Figure 7(b) illustrates the ratios of the CO₂ capture and effective interfacial area in the outer
 686 cavity zone to the whole RPB under the same operating conditions as Figure 7(a). On one hand,
 687 the effective interfacial area mainly depends on the liquid flow rate and rotational speed. On
 688 other hand, the increasing temperature in Figure 7(a) affects the phase properties, such as the
 689 density and viscosity. As a results, the r_e changes only slightly in Figure 7(b).

690 On considering Figure 7(b), the liquid temperature approximately reaches the maximum after
 691 entering the outer cavity zone, which means that the average liquid temperature is relatively
 692 high in the outer cavity zone compared with that in the packing zone. The higher liquid
 693 temperature benefits the reaction rate. Therefore, the proportion of the CO₂ removed in the
 694 outer cavity zone (r_c) increases as the MEA concentration increases. From the above analysis,
 695 it is indicated that the higher MEA concentration could enhance the CO₂ capture in the full
 696 RPB, and more proportions of CO₂ are captured in the outer cavity zone.



697
 698 **Figure 7.** The effect of the MEA concentration on (a) $K_L a_e$ and liquid outlet temperature and
 699 (b) ratio of the contribution of CO₂ capture and effective interfacial area in the outer cavity
 700 zone.

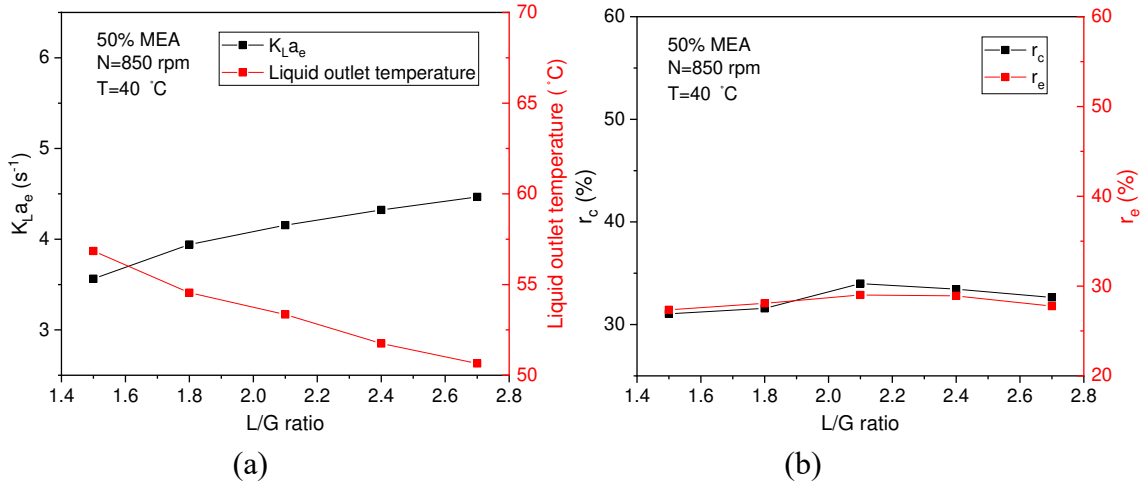
701 **(ii) Effect of the liquid flow rate**

702 For post-combustion capture using the CPB, it is reported that the liquid outlet temperature and
 703 CO₂ capture level increase when the L/G ratio increases due to the larger amount of free amine
 704 molecules and the higher effective interfacial area (Sønderby et al., 2013). However, a reduced
 705 liquid outlet temperature is observed in the RPB from the experimental work (Kolawole, 2019).
 706 Therefore, it is worth exploring the difference in the CO₂ capture processes under various L/G
 707 ratios within the CPB and RPB.

708 Figure 8(a) presents the predicted impact of the L/G ratio on $K_L a_e$ and liquid outlet temperature
 709 with the L/G ratio ranging from 1.5 to 2.7 while keeping the gas flow rate unchanged. As the
 710 L/G ratio increases, more CO₂ is captured resulting from the larger amount of free amine

711 molecules and also the mass transfer resistance is reduced due to the enhanced gas-liquid
712 mixing (Luo et al., 2021) and liquid film refreshing. In addition, the increasing L/G ratio leads
713 to a higher effective interfacial area. These factors increase $K_L a_e$, but this increase tends to
714 slow down as L/G increases. Although increasing the number of tiny droplets that could be
715 generated, the increase in the effective interfacial area is limited at the higher range of the liquid
716 flow rate (Wu et al., 2017). Similarly, although more heat is generated as the L/G ratio increases,
717 the heat generation is not high enough to increase the liquid temperature due to the large
718 sensible heat of the liquid phase. In addition, this may explain the decreasing liquid outlet
719 temperature in Figure 8(a), which is consistent with the Kolawole's experimental results
720 (Kolawole, 2019). This is different from that typically observed in a CPB because the liquid
721 flow rate in a CPB is the dominant factor that could significantly increase the interfacial area
722 leading to the significantly increased mass transfer and heat generation. From the above
723 analysis, the increased liquid flow rate, effective interfacial area and the decreased liquid
724 temperature together cause $K_L a_e$ to increase more slowly (Kuntz and Aroonwilas, 2009).

725 Figure 8(b) shows the ratios of the CO₂ capture and effective interfacial area in the outer cavity
726 zone to the whole RPB for L/G ratio from 1.5 to 2.1. The r_c and r_e slightly change with
727 changing L/G ratios, but it is noted that a critical point is observed in Figure 8(b). As the L/G
728 ratio increases from 1.5 to 2.1, more liquid concentrates in the outer cavity zone and the
729 temperature in this zone is relatively higher. As a result, both r_c and r_e increase. While the L/G
730 ratio continues to increase, the liquid turbulence is dominant in the packing region and the
731 liquid temperature continues to decrease in the outer cavity zone. As a result, the CO₂ that is
732 captured in the packing increases, thus leading to a reduced value of r_c and r_e .



733

734 **Figure 8.** The effect of the L/G ratio on (a) $K_L a_e$ and liquid outlet temperature and (b) ratios
 735 of the contribution of CO₂ capture and effective interfacial area in the outer cavity zone.

736 **(iii) Effect of the rotational speed**

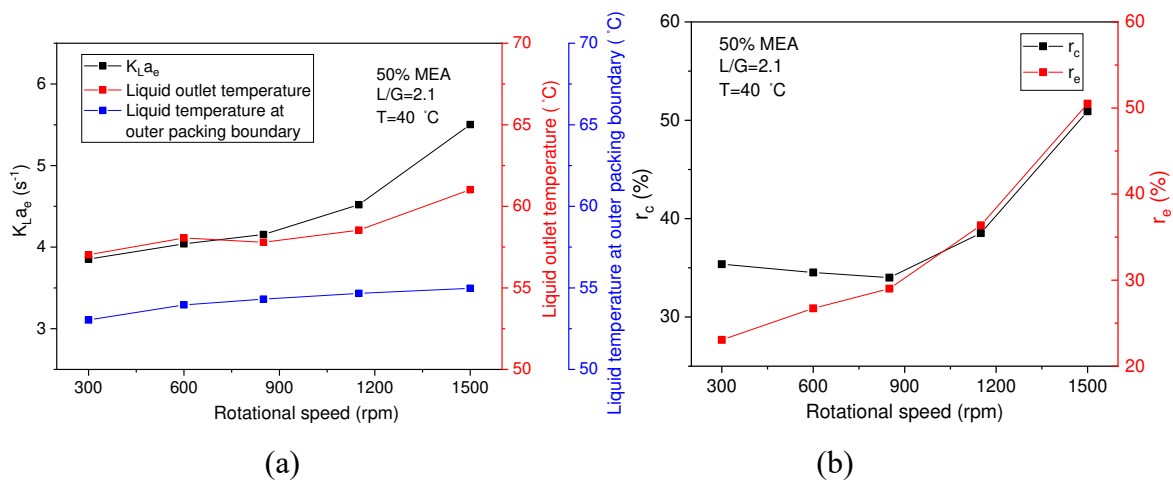
737 In our previous investigation (Zhang et al., 2022a), it was reported that the rotational speed has
 738 a considerable impact on the liquid holdup and liquid distribution, thus it will further affect the
 739 CO₂ capture in the RPB absorber. In addition, the rotational speed could directly affect the
 740 effective interfacial area and the reaction time between the liquid and gas phases. Thus, it is a
 741 key parameter for the PCC in a RPB.

742 Figure 9(a) presents the impact of the rotational speed on the $K_L a_e$, the liquid outlet
 743 temperature of the entire RPB and the liquid temperature at the outer packing boundary. Figure
 744 9(b) illustrates the ratios of the CO₂ captured and effective interfacial area in the outer cavity
 745 zone to the whole RPB under various rotational speeds. As the rotational speed increases, the
 746 liquid holdup in the packing region reduces due to the stronger centrifugal force. However,
 747 more tiny liquid droplets are formed among the packing region, which results in an improved
 748 effective interfacial area (Zhang et al., 2022a), although the magnitude of the improvement is
 749 very limited. On the other hand, the liquid fraction in the outer cavity zone becomes larger
 750 because the liquid accumulation in the outer cavity zone relies on the gravitational force to
 751 remove out of the RPB. In addition, more liquid is more likely to attach and cover the casing
 752 wall when it flows out from the packing with a higher rotational speed, which leads to a
 753 significant increase in the effective interfacial area in the outer cavity zone. Thus, the r_e in

754 Figure 9(b) gradually increases as the rotational speed increases. And the increasing effective
 755 interfacial area in both the packing and outer cavity zones makes the $K_L a_e$ increase in Figure
 756 9(a).

757 From Figure 9(a), the liquid temperature flowing out the packing region (blue line) increases
 758 with the rotational speed increasing and it is always lower than the liquid outlet temperature
 759 (red line). However, the liquid outlet temperature reduces when the rotational speed increases
 760 from 600 to 850 rpm. The possible reason is that in the outer cavity zone, the liquid fraction
 761 increases more significantly compared with the increase in the effective interfacial area when
 762 the rotational speed increases from 600 to 850 rpm. And the reaction heat generated in this
 763 zone at 850 rpm cannot significantly increase the temperature of the liquid with a large fraction.
 764 Therefore, the liquid outlet temperature at 850 rpm slightly reduces from the value at 600 rpm.

765 In addition, r_e increases as the rotational speed increases, thus, r_c is expected to increase
 766 accordingly. However, r_c reduces in the range of 300 to 850 rpm as observed in Figure 9(b).
 767 The possible reason is that the micromixing performance in the packing region improves
 768 significantly at the lower rotational speed (Ouyang et al., 2019). As a result, the packing region
 769 captures more CO_2 compared with the outer cavity zone when the rotational speed increases
 770 from 300 to 850 rpm. Overall, the $K_L a_e$ and r_e increase with the increase in the rotational speed,
 771 and r_c decreases first and then increases.



772

773 **Figure 9.** The effect of the rotational speed on the (a) $K_L a_e$ and liquid temperatures at liquid
774 outlet and outer packing boundary and (b) ratios of the contribution of CO₂ capture and
775 effective interfacial area in the outer cavity zone.

776 **4. Conclusions**

777 This paper has established a full 3D RPB model using the Eulerian porous medium approach,
778 and the physical and chemical processes occurring in the packing and outer cavity zone of a
779 pilot scale RPB have been modelled. The overall CFD results agree with the experimental data.
780 Nonetheless, further validation may be required when applying the sub-models to other RPBs,
781 in particular when different packings are employed. The main findings of this paper are as
782 follows: (i) a new completed 3D Eulerian porous medium RPB model was established based
783 on a pilot scale RPB model. By using the Eulerian porous medium modelling method, the CO₂
784 absorption performance within a whole RPB could be effectively investigated due to the
785 significantly reduced computational cost; (ii) the force, effective interfacial area, heat transfer,
786 mass transfer models were coupled with the Eulerian RPB model, thus, the hydrodynamics,
787 thermodynamics and mass transfer processes could be thoroughly analyzed; (iii) the effective
788 interfacial area, force and even liquid droplet diameter models were introduced separately for
789 the packing region and the outer cavity zone for accurately predicting the CO₂ capture
790 performance inside the full RPB, and the CO₂ absorption processes in these zones were
791 quantitatively evaluated under various operating conditions by the CFD method for the first
792 time.

793 The predicted simulation results are in good agreement with the experimental data by
794 comparing the CO₂ fraction in the gas outlet and liquid outlet temperature. In addition, the
795 end-effect zone is observed near the inner packing region where a large gas-liquid contact area
796 and strong micromixing occur due to the liquid dispersion and strong interaction between the
797 liquid and packing. However, the amount of the CO₂ transferred between the phases is small
798 in the end-effect zone due to the small local CO₂ fraction in the gas phase and the low liquid
799 temperature.

800 The outer cavity zone has an effect on the CO₂ capture inside the RPB. The contributions of
801 the outer cavity zone to the effective interfacial area and CO₂ capture are respectively in the
802 ranges of 25%~40% and 28%~42% in the RPB investigated in this paper. However, of course,
803 these values may be different for different designs. Also, the simulation results show that the
804 CO₂ captured in the outer cavity zone becomes more as the MEA concentration increases,
805 however, the MEA concentrations have little effect on the ratio of the effective interfacial area
806 in the outer cavity zone to the whole RPB. The contributions of the outer cavity zone to the
807 effective interfacial area and CO₂ capture change slightly with the L/G ratio increasing. With
808 the increase in the rotational speed, the ratio of the effective interfacial area in the outer cavity
809 zone to the whole RPB increases, however, the ratio of the CO₂ capture in the outer cavity
810 zone to the whole RPB first decreases slightly and then increases significantly.

811 The RPB model developed in this paper can successfully and effectively predict the CO₂
812 capture process in the whole RPB, and this demonstrates the substantial potential of the model,
813 with further validation, to be used for process optimization and design of the large-scale RPB
814 for industrial PCC. There are two limitations of the CFD model proposed in this study. The
815 first is that using the porous media approach, the details of the fluid flows are not resolved, and
816 therefore the characteristics of the formation of liquid droplets/films are unable to be revealed.
817 Secondly, the accuracy of the model is highly dependent on the applicability of the sub-models
818 employed and therefore, careful validation of the model should be considered, especially when
819 a very different packing is employed for the RPB.

820 **Acknowledgement**

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822 **Reference**

- 823 Amundsen, T.G., Øi, L.E., Eimer, D.A., 2009. Density and viscosity of
824 monoethanolamine+water+carbon dioxide from (25 to 80) C. *Journal of Chemical &*
825 *Engineering Data* 54, 3096-3100.
- 826 Borhani, T.N., Oko, E., Wang, M., 2018. Process modelling and analysis of intensified CO₂
827 capture using monoethanolamine (MEA) in rotating packed bed absorber. *Journal of Cleaner*

828 Production 204, 1124-1142.

829 Cortes Garcia, G.E., van der Schaaf, J., Kiss, A.A., 2017. A review on process intensification
830 in HiGee distillation. *J. Chem. Technol. Biot* 92, 1136-1156.

831 Danckwerts, P., 1979. The reaction of CO₂ with ethanolamines. *Chem. Eng. Sci.* 34, 443-446.

832 Dashliborun, A.M., Larachi, F., Taghavi, S.M., 2019. Gas-liquid mass-transfer behavior of
833 packed-bed scrubbers for floating/offshore CO₂ capture. *Chem. Eng. J.* 377.

834 Ebadi Amooghini, A., Moftakhari Sharifzadeh, M.M., Zamani Pedram, M., 2018. Rigorous
835 modeling of gas permeation behavior in facilitated transport membranes (FTMs); evaluation
836 of carrier saturation effects and double-reaction mechanism. *Greenh. Gases: Sci. Technol* 8,
837 429-443.

838 Esmacili, A., Tamuzi, A., Borhani, T.N., Xiang, Y., Shao, L., 2022. Modeling of carbon dioxide
839 absorption by solution of piperazine and methyldiethanolamine in a rotating packed bed. *Chem.*
840 *Eng. Sci.* 248.

841 Fourati, M., Roig, V., Raynal, L., 2013. Liquid dispersion in packed columns: Experiments and
842 numerical modeling. *Chem. Eng. Sci.* 100, 266-278.

843 Garba, M.U., 2012. Prediction of ash deposition for biomass combustion and coal/biomass co-
844 combustion. University of Leeds.

845 Gbadago, D.Q., Oh, H.T., Oh, D.H., Lee, C.H., Oh, M., 2020. CFD simulation of a packed bed
846 industrial absorber with interbed liquid distributors. *Int. J. Greenh. Gas Con.* 95.

847 Guo, F., Zheng, C., Guo, K., Feng, Y., Gardner, N.C., 1997. Hydrodynamics and mass transfer
848 in cross-flow rotating packed bed. *Chem. Eng. Sci.* 52, 3853-3859.

849 Guo, J., Jiao, W., Qi, G., Yuan, Z., Liu, Y., 2019. Applications of high-gravity technologies in
850 gas purifications: A review. *Chin. J. Chem. Eng.* 27, 1361-1373.

851 Guo, K., Zhang, Z., Luo, H., Dang, J., Qian, Z., 2014. An innovative approach of the effective
852 mass transfer area in the rotating packed bed. *Ind. Eng. Chem. Res.* 53, 4052-4058.

853 Guo, T.Y., Shi, X., Chu, G.W., Xiang, Y., Wen, L.X., Chen, J.F., 2016. Computational fluid
854 dynamics analysis of the micromixing efficiency in a rotating-packed-bed reactor. *Ind. Eng.*
855 *Chem. Res.* 55, 4856-4866.

856 Hikita, H., Asai, S., Ishikawa, H., Honda, M., 1977. The kinetics of reactions of carbon dioxide
857 with monoethanolamine, diethanolamine and triethanolamine by a rapid mixing method. *the*
858 *chemical Engineering Journal* 13, 7-12.

859 Hughmark, G.A., 1967. Mass and heat transfer from rigid spheres. *AIChE J.* 13, 1219– 1221.

860 Iliuta, I., Larachi, F., Fourati, M., Raynal, L., Roig, V., 2014. Flooding limit in countercurrent
861 gas-liquid structured packed beds-Prediction from a linear stability analysis of an Eulerian two-
862 fluid model. *Chem. Eng. Sci.* 120, 49-58.

863 Ishii, M., Zuber, N., 1979. Drag coefficient and relative velocity in bubbly, droplet or
864 particulate flows. *AIChE J.* 25, 843-855.

865 Jiang, Y., Khadilkar, M., Al-Dahhan, M., Dudukovic, M., 2002. CFD of multiphase flow in
866 packed-bed reactors: I. k-Fluid modeling issues. *AIChE J.* 48, 701-715.

867 Jiru, Y., Eimer, D.A., 2013. A study of mass transfer kinetics of carbon dioxide in
868 (monoethanolamine + water) by stirred cell. *Energy Procedia* 37, 2180-2187.

869 Khan, M.Z.A., 2017. Causes and consequences of greenhouse effect & its catastrophic
870 problems for Earth. *Inter. J. Sustainability Management and Information Technologies* 3, 34.

871 Kim, J., Pham, D.A., Lim, Y.I., 2016. Gas-liquid multiphase computational fluid dynamics
872 (CFD) of amine absorption column with structured-packing for CO₂ capture. *Comput. Chem.*
873 *Eng.* 88, 39-49.

874 Kim, J., Pham, D.A., Lim, Y.I., 2017. Effect of gravity center position on amine absorber with
875 structured packing under offshore operation: Computational fluid dynamics approach. *Chem.*
876 *Eng. Res. Des.* 121, 99-112.

877 Kolawole, T.O., 2019. Intensified post-combustion carbon capture using a pilot scale rotating
878 packed bed and monoethanolamine solutions. Newcastle University.

879 Kuntz, J., Aroonwilas, A., 2009. Mass-transfer efficiency of a spray column for CO₂ capture
880 by MEA. *Energy Procedia* 1, 205-209.

881 Lee, J., Kolawole, T., Attidekou, P., 2017. Carbon capture from a simulated flue gas using a
882 rotating packed bed adsorber and monoethanolamine (MEA). *Energy Procedia* 114, 1834-1840.

883 Liu, G.B., Yu, K.T., Yuan, X.G., Liu, C.J., Guo, Q.C., 2006. Simulations of chemical absorption
884 in pilot-scale and industrial-scale packed columns by computational mass transfer. *Chem. Eng.*
885 *Sci.* 61, 6511-6529.

886 Liu, Y.Z., Luo, Y., Chu, G.W., Liu, W., Shao, L., Chen, J.F., 2019. Liquid holdup and wetting
887 efficiency in a rotating trickle-bed reactor. *AIChE J.* 65.

888 Lu, X., Xie, P., Ingham, D.B., Ma, L., Pourkashanian, M., 2019. Modelling of CO₂ absorption
889 in a rotating packed bed using an Eulerian porous media approach. *Chem. Eng. Sci.* 199, 302-
890 318.

891 Lu, X., Xie, P., Ingham, D.B., Ma, L., Pourkashanian, M., 2018. A porous media model for

892 CFD simulations of gas-liquid two-phase flow in rotating packed beds. *Chem. Eng. Sci.* 189,
893 123–134

894 Luo, X., Hartono, A., Hussain, S., F. Svendsen, H., 2015. Mass transfer and kinetics of carbon
895 dioxide absorption into loaded aqueous monoethanolamine solutions. *Chem. Eng. Sci.* 123, 57-
896 69.

897 Luo, X., Hartono, A., Svendsen, H.F., 2012. Comparative kinetics of carbon dioxide absorption
898 in unloaded aqueous monoethanolamine solutions using wetted wall and string of discs
899 columns. *Chem. Eng. Sci.* 82, 31-43.

900 Luo, X., Wang, M., Lee, J., Hendry, J., 2021. Dynamic modelling based on surface renewal
901 theory, model validation and process analysis of rotating packed bed absorber for carbon
902 capture. *Appl. Energy* 301.

903 Luo, Y., Luo, J.Z., Chu, G.W., Zhao, Z.Q., Arowo, M., Chen, J.F., 2017. Investigation of
904 effective interfacial area in a rotating packed bed with structured stainless steel wire mesh
905 packing. *Chem. Eng. Sci.* 170, 347-354.

906 Mardani, A., Mahalegi, H. K. M., 2019. Hydrogen enrichment of methane and syngas for
907 MILD combustion. *Int. J. Hydrog. Energy* 44, 9423-9437.

908 Moftakhari Sharifzadeh, M.M., Ebadi Amooghin, A., Zamani Pedram, M., Omidkhah, M.,
909 2016. Time-dependent mathematical modeling of binary gas mixture in facilitated transport
910 membranes (FTMs): A real condition for single-reaction mechanism. *J. Ind. and Eng. Chem.*
911 39, 48-65.

912 Munjal, S., Duduković, M.P., Ramachandran, P., 1989. Mass-transfer in rotating packed beds-
913 I. Development of gas-liquid and liquid-solid mass-transfer correlations. *Chem. Eng. Sci.* 44,
914 2245-2256.

915 Ouyang, Y., Xiang, Y., Gao, X.Y., Zou, H.K., Chu, G.W., Agarwal, R.K., Chen, J.F., 2019.
916 Micromixing efficiency optimization of the premixer of a rotating packed bed by CFD. *Chem.*
917 *Eng. Process.* 142.

918 Penttilä, A., Dell’Era, C., Uusi-Kyyny, P., Alopaeus, V., 2011. The Henry's law constant of N₂O
919 and CO₂ in aqueous binary and ternary amine solutions (MEA, DEA, DIPA, MDEA, and AMP).
920 *Fluid Phase Equilib.* 311, 59-66.

921 Pham, D.A., Lim, Y.I., Jee, H., Ahn, E., Jung, Y., 2015. Porous media Eulerian computational
922 fluid dynamics (CFD) model of amine absorber with structured-packing for CO₂ removal.
923 *Chem. Eng. Sci.* 132, 259-270.

924 Qing, Z., Yincheng, G., Zhenqi, N., 2011. Experimental studies on removal capacity of carbon
925 dioxide by a packed reactor and a spray column using aqueous ammonia. *Energy Procedia* 4,
926 519-524.

927 Sang, L., Luo, Y., Chu, G.W., Liu, Y.Z., Liu, X.Z., Chen, J.F., 2017a. Modeling and
928 experimental studies of mass transfer in the cavity zone of a rotating packed bed. *Chem. Eng.*
929 *Sci.* 170, 355-364.

930 Sang, L., Luo, Y., Chu, G.W., Zhang, J.P., Xiang, Y., Chen, J.F., 2017b. Liquid flow pattern
931 transition, droplet diameter and size distribution in the cavity zone of a rotating packed bed: A
932 visual study. *Chem. Eng. Sci.* 158, 429-438.

933 Sang, L., Luo, Y., Chu, G.W., Sun, B.C., Zhang, L.L., Chen, J.F., 2019. A three-zone mass
934 transfer model for a rotating packed bed. *AIChE J.* 65.

935 Sebastia-Saez, D., Gu, S., Ranganathan, P., 2014. Volume of Fluid modeling of the reactive
936 mass transfer of CO₂ into aqueous amine solutions in structured packed elements at micro-
937 scale. *Energy Procedia* 63, 1229-1242.

938 Sebastia-Saez, D., Gu, S., Ranganathan, P., Papadikis, K., 2015. Micro-scale CFD modeling of
939 reactive mass transfer in falling liquid films within structured packing materials. *Int. J. Greenh.*
940 *Gas Con.* 33, 40-50.

941 Snijder, E.D., te Riele, M.J., Versteeg, G.F., Van Swaaij, W., 1993. Diffusion coefficients of
942 several aqueous alkanolamine solutions. *J. Chem. Eng. Data* 38, 475-480.

943 Sønderby, T.L., Carlsen, K.B., Fosbøl, P.L., Kiørboe, L.G., von Solms, N., 2013. A new pilot
944 absorber for CO₂ capture from flue gases: Measuring and modelling capture with MEA solution.
945 *Int. J. Greenh. Gas Con.* 12, 181-192.

946 Vaewhongs, P., Photein, K., Nimchareon, R., Limlertchareonwanit, T., Minakanishtha, K.,
947 Maneeintr, K., Charinpanitkul, T., 2020. Gas-phase mass transfer coefficient of CO₂ in different
948 alkanolamine solutions within packed-bed absorption column. *AIP Conf. Proc.* 2279, 90001.

949 Vázquez, G., Alvarez, E., Navaza, J.M., Rendo, R., Romero, E., 1997. Surface tension of binary
950 mixtures of water + monoethanolamine and water + 2-amino-2-methyl-1-propanol and tertiary
951 mixtures of these amines with water from 25 C to 50 C. *J. Chem. Eng. Data* 42, 57-59.

952 Versteeg, G., Van Dijck, L., van Swaaij, W.P.M., 1996. On the kinetics between CO₂ and
953 alkanolamines both in aqueous and non-aqueous solutions. An overview. *Chem. Eng. Commun.*
954 144, 113-158.

955 Versteeg, G., van Swaaij, W.P.M., 1988. On the kinetics between CO₂ and alkanolamines both

956 in aqueous and non-aqueous solutions-I. Primary and secondary amines. *Chem. Eng. Sci.* 43,
957 573-585.

958 Wang, J.Q., Ouyang, Y., Li, W.L., Esmaeili, A., Xiang, Y., Chen, J.F., 2020. CFD analysis of
959 gas flow characteristics in a rotating packed bed with randomly arranged spherical packing.
960 *Chem. Eng. J.* 385.

961 Wang, Y., Li, Y.B., Su, M.J., Chu, G.W., Sun, B.C., Luo, Y., 2021. Liquid droplet dispersion in
962 a rotating packed bed: Experimental and numerical studies. *Chem. Eng. Sci.*, 240

963 Wellek, R., Brunson, R., Law, F., 1978. Enhancement factors for gas-absorption with second-
964 order irreversible chemical reaction. *Can. J. Chem. Eng.* 56, 181-186.

965 Wu, W., Luo, Y., Chu, G.W., Liu, Y., Zou, H.K., Chen, J.F., 2018. Gas flow in a multiliquid-
966 inlet rotating packed bed: three-dimensional numerical simulation and internal optimization.
967 *Ind. Eng. Chem. Res.* 57, 2031-2040.

968 Wu, X., He, M., Yu, Y., Qin, Z., Zhang, Z., 2017. Overall mass transfer coefficient of CO₂
969 absorption in a diameter-varying spray tower. *Energy Procedia* 114, 1665-1670.

970 Xie, P., 2019. Hydrodynamics and mass transfer of rotating packed beds for CO₂ capture.
971 University of Sheffield.

972 Xie, P., Lu, X., Ding, H., Yang, X., Ingham, D., Ma, L., Pourkashanian, M., 2019. A mesoscale
973 3D CFD analysis of the liquid flow in a rotating packed bed. *Chem. Eng. Sci.* 199, 528-545.

974 Xu, Y.C., Li, Y.B., Liu, Y.Z., Luo, Y., Chu, G.W., Zhang, L.L., Chen, J.F., 2019. Liquid jet
975 impaction on the single-layer stainless steel wire mesh in a rotating packed bed reactor. *AIChE*
976 *J.* 65.

977 Yang, K., Chu, G., Zou, H., Sun, B., Shao, L., Chen, J.F., 2011. Determination of the effective
978 interfacial area in rotating packed bed. *Chem. Eng. J.* 168, 1377-1382.

979 Yang, W., Wang, Y., Chen, J., Fei, W., 2010. Computational fluid dynamic simulation of fluid
980 flow in a rotating packed bed. *Chem. Eng. J.* 156, 582-587.

981 Yang, Y., Xiang, Y., Chu, G., Zou, H., Luo, Y., Arowo, M., Chen, J.F., 2015a. A noninvasive
982 X-ray technique for determination of liquid holdup in a rotating packed bed. *Chem. Eng. Sci.*
983 138, 244-255.

984 Yang, Y., Xiang, Y., Chu, G., Zou, H., Sun, B., Arowo, M., Chen, J.F., 2016. CFD modeling of
985 gas-liquid mass transfer process in a rotating packed bed. *Chem. Eng. J.* 294, 111-121.

986 Yang, Y., Xiang, Y., Li, Y., Chu, G., Zou, H., Arowo, M., Chen, J., 2015b. 3D CFD modelling
987 and optimization of single-phase flow in rotating packed beds. *Can. J. Chem. Eng.* 93, 1138-

988 1148.

989 Yi, F., Zou, H.K., Chu, G.W., Shao, L., Chen, J.F., 2009. Modeling and experimental studies
990 on absorption of CO₂ by Benfield solution in rotating packed bed. *Chem. Eng. J.* 145, 377-384.

991 Ying, J., Eimer, D.A., 2013. Determination and measurements of mass transfer kinetics of CO₂
992 in concentrated aqueous monoethanolamine solutions by a stirred cell. *Ind. Eng. Chem. Res.*
993 52, 2548-2559.

994 Zhang, G., Ingham, D., Ma, L., Pourkashanian, M., 2022a. Modelling of 3D liquid dispersion
995 in a rotating packed bed using an Eulerian porous medium approach. *Chem. Eng. Sci.* 250.

996 Zhang, J., 1996. Experiment and modelling of liquid flow and mass transfer in rotating packed
997 bed. Beijing University of Chemical Technology.

998 Zhang, J.P., Luo, Y., Chu, G.W., Sang, L., Liu, Y., Zhang, L.L., Chen, J.F., 2017. A hydrophobic
999 wire mesh for better liquid dispersion in air. *Chem. Eng. Sci.* 170, 204-212.

1000 Zhang, W., Xie, P., Li, Y., Teng, L., Zhu, J., 2022b. 3D CFD simulation of the liquid flow in a
1001 rotating packed bed with structured wire mesh packing. *Chem. Eng. J.* 427.

1002 Zhang, W., Xie, P., Li, Y., Zhu, J., 2022c. Modeling of gas-liquid flow in a rotating packed bed
1003 using an Eulerian multi-fluid approach. *AIChE J.* 68.