

1 Process Intensification for Post-combustion CO₂ capture 2 with Chemical Absorption: A critical review

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8 Abstract

9 The concentration of CO₂ in the atmosphere is increasing rapidly. CO₂ emissions
10 may have an impact on global climate change. Effective CO₂ emission abatement
11 strategies such as carbon capture and storage (CCS) are required to combat this
12 trend. Compared with pre-combustion carbon capture and oxy-fuel carbon capture
13 approaches, post-combustion CO₂ capture (PCC) using solvent process is one of the
14 most mature carbon capture technologies. There are two main barriers for the PCC
15 process using solvent to be commercially deployed: (a) high capital cost; (b) high
16 thermal efficiency penalty due to solvent regeneration. Applying Process
17 intensification (PI) technology into PCC with solvent process has the potential to
18 significantly reduce capital costs compared with conventional technology using
19 packed columns. This paper intends to evaluate different PI technologies for their
20 suitability in PCC process. The study shows that rotating packed bed (RPB)
21 absorber/stripper has attracted much interest due to its high mass transfer capability.
22 Currently experimental studies on CO₂ capture using RPB are based on standalone
23 absorber or stripper. Therefore a schematic process flow diagram of intensified PCC
24 process is proposed so as to motivate other researches for possible optimal design,
25 operation and control. To intensify heat transfer in reboiler, spinning disc technology
26 is recommended. To replace cross heat exchanger in conventional PCC (with
27 packed column) process, printed circuit heat exchanger will be preferred. Solvent
28 selection for conventional PCC process has been studied extensively. However, it
29 needs more studies for solvent selection in intensified PCC process. The authors
30 also predicted research challenges in intensified PCC process and potential new
31 breakthrough from different aspects.

32 **Keywords:** *Post-combustion CO₂ capture, Chemical Absorption, Rotating Packed*
33 *Bed (RPB), Process Intensification (PI), Solvents, Intensified Heat Exchanger*

34 1. Introduction

35 1.1 CO₂ Emissions and Climate Change

36 Global energy demand is expected to continue to rise due to the increasing world
37 population, in addition to economic development of nations such as Brazil, Russia

38 India and China. Dependence on renewable energy alone such as solar, wind and
39 tidal power to meet the projected demand is not feasible due to their intermittent
40 nature. Therefore, fossil fuel remains the very attractive options in meeting future
41 energy demands. But power generation using fossil fuel is estimated to be the
42 largest source of worldwide carbon emissions [1].

43 Combustion of fossil fuels (e.g. petroleum, coal and natural gas) accounts for the
44 majority of CO₂ emissions. Globally most fossil-fuelled electricity production is from
45 coal (63%), followed by natural gas (29%) and oil (9%) [2]. For instance about 85.5%
46 of its coal (produced and imported) is used for electricity generation in the UK in
47 2011[3]. Albo *et al.* [4] stated that among the greenhouse gases, CO₂ contributes to
48 more than 60% of global warming. Statistics from World Metrological Organisation
49 (WMO) showed that the amount of CO₂ in the atmosphere reached 393.1 ppm in
50 2012. The WMO report also showed that the amount of CO₂ in the atmosphere has
51 increased on average by 2 ppm per year for the past 10 years, this increased
52 atmospheric concentration of CO₂ affects the radiative balance of the Earth [5].

53 1.2 CCS Technologies

54 Intergovernmental panel on climate change (IPCC) [6] has set ambitious goal to
55 reduce CO₂ emission by 50% in 2050 as compared to the level of 1990 so as to
56 overcome the foreseen environmental challenges. In order to achieve the required
57 emission reductions in the most cost-effective manner, carbon capture and storage
58 (CCS) will need to contribute around one-fifth of total reductions in emissions by
59 2050 [7].

60 CCS consists of three basic stages: (a) separation of CO₂; (b) CO₂ transportation
61 and (c) CO₂ storage. There are three major approaches for CCS: post-combustion
62 capture, pre-combustion capture and oxy-fuel process [8].

63 1.3 Different technical options in the context of PCC

64 PCC technologies are at various stages of development. Abanades *et al.* [9]
65 reviewed the technological readiness level for different CO₂ capture technologies in
66 the context of PCC including chemical looping, calcium looping, PCC using solvent,
67 PCC using adsorbent and PCC using membrane. The most matured process is PCC
68 using solvent [9,10].

69 However, the PCC using solvent process has several drawbacks including: (1) low
70 CO₂ loading capacity; (2) high equipment corrosion rate; (3) amine degradation by
71 SO₂, NO₂, and O₂ in the flue gases which induces a high solvent makeup rate; (4)
72 high energy consumption during solvent regeneration; (5) large equipment size [11-
73 14] (in whole PCC process, absorber and stripper account for 55% and 17% of the
74 total capital cost respectively [15]). This process is introduced in more detail in
75 Section 1.4. and Table 1 gives its performance indicators and status for chemical
76 absorption compared to adsorption and membrane technologies.

77

78 Table 1 Status of post-combustion CO₂ capture development [15-23]

	Absorbent	Adsorbent	Membrane
Commercial usage in chemical process industries	High	Moderate	Low/Niche
Operational confidence	High	High, but complex	Low to moderate
Primary source of energy penalty	Solvent Regeneration (thermal)	Solid sorbent Regeneration (thermal/vacuum)	Compression on feed and/or vacuum on permeate
Regeneration energy (MJ/kg-CO ₂)	2.2 – 6	0.5 – 3.12	0.5 – 6
Efficiency penalty (%)	8.2 – 14	5.4 – 9.0	6.4 – 8.5
Development trends	New solvent, thermal integration	New sorbent, process configuration	New membrane, process configuration

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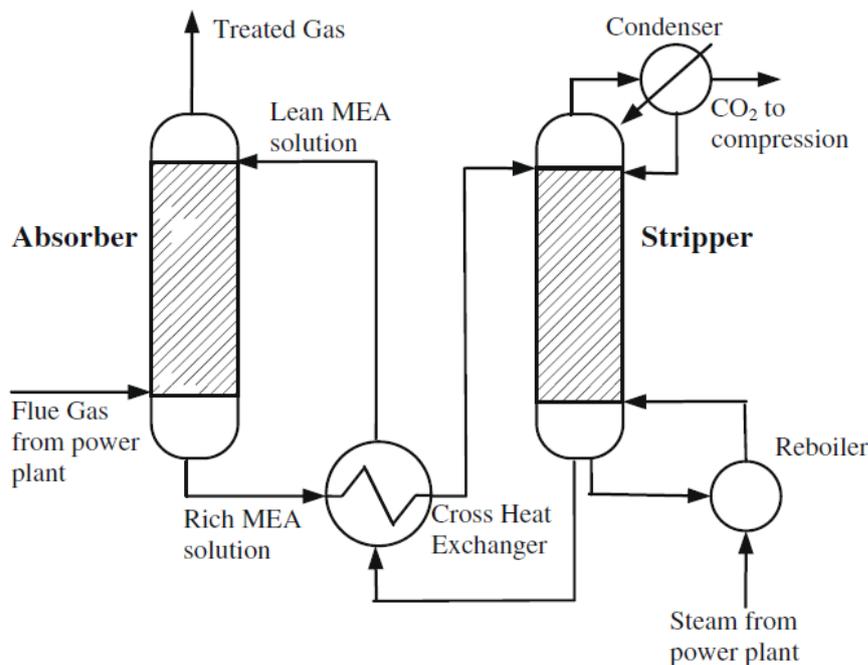
80 Chemically modified adsorbents have proved to be applicable for PCC process
81 because of large CO₂ adsorption capacity, high adsorption and desorption rates,
82 high tolerance to moisture, and high selectivity towards CO₂ over other gases [24]. In
83 terms of regeneration energy, Zhao *et al.* [25] reported that solid sorbent does not
84 have any obvious advantage over the matured MEA process in terms of energy
85 consumption in the first design (i.e. two reactor used, one for adsorption and the
86 other for regeneration). But the novel (second) design (i.e. using three reactors, one
87 for adsorption, one for regeneration and another one for formation of
88 K₂CO₃·1.5H₂O), the regeneration energy can be reduced by utilizing the waste heat
89 from the process, however this design is difficult to control because the reactors
90 operate at different pressures. A lot of researchers have focused on new adsorbent
91 development, process optimization, and equipment innovation [26-50].

92 Abanades *et al.* [9] shows that membrane process for PCC is at almost the same
93 level of technological readiness as adsorption. Therefore, more study in this area is
94 needed in order to get detailed technical performance at large scale condition. Many
95 researchers have developed new membranes that offer better performance in term
96 of selectivity. The recent key projects developing membranes and modules for CO₂
97 capture include Membrane Technology and Research (MTR) in the USA,
98 NanoGLOWA in Germany, the carbon capture project (CCP) and the CO₂CRC in
99 Australia [9]. Membranes process for PCC is beneficial because of relatively small
100 footprint, no phase change, simple mechanical system, steady-state operating
101 conditions (usually), easy scale-up and flexibility [51-53]. The major challenge for
102 membranes comes from the potential fouling of the membrane surfaces from
103 particulate matter, uncertainty about the performance and cost of large-scale efficient
104 vacuum pumps and compressors required for PCC, and the ability to integrate the
105 process into a power plant. Technological outlook of membrane system is reported in
106 Abanades *et al.* [9] and highlighted that for the technology to be competitive with
107 other PCC technology, the membrane needs to be of high CO₂ permeance (around
108 1000 gas permeance units (GPU)) to be economical.

109 The technological readiness (maturity) level of PCC based on chemical absorption
110 and the first commercial deployment of PCC using solvents plant in 2014 motivate
111 the authors to write this paper and highlight the process design option that will
112 improve the chemical absorption PCC process using the RPB technology.

113 1.4 Current status of PCC using solvent and its commercial deployment

114



115

116 Figure 1 Simplified process flow diagram of chemical absorption process for post
117 combustion carbon capture [54]

118 Figure 1 shows a simplified PCC process. Flue gas from CO₂ sources such as
119 power plant is contacted counter-currently with lean solvent solution in the absorber.
120 Solvent chemically absorbs CO₂ in the flue gas. This leaves a treated gas stream of
121 lower CO₂ content. The solvent solution (now rich solvent) is regenerated in the
122 stripper. CO₂ from the top of the stripper is compressed and transported while the
123 lean (regenerated) solvent solution is returned to the absorber column passing
124 through a cross exchanger to recover heat with rich solvent from the absorber.

125 Studies on PCC with chemical absorption process were reported mainly for fossil
126 fuel-fired power plants. Dugas [55] carried out pilot plant experimental studies of
127 PCC in the context of fossil fuel-fired power plants. Mangalapally *et al.* [56-59]
128 reported pilot plant studies of PCC for gas fired power plant. Lawal *et al.* [11,60-62],
129 Biliyok *et al.* [63], Kvamsdal *et al.* [54,64-66], MacDowell and Shah [67-69],
130 MacDowell *et al.* [70], Lucquiaud *et al.* [71,72], Errey *et al.* [73], Agbonghae *et al.*
131 [14,74] carried out steady state and dynamic modelling of CO₂ absorption for PCC
132 using solvents for fossil-fuel fired power plants. Asendrych *et al.* [75], Sebastia-Saez
133 *et al.* [76], Raynal *et al.* [77], Raynal and Royon-Lebeaud [78] studied PCC for fossil
134 fuel fired power plant using CFD.

135 There is also good progress in commercial deployment of PCC using solvent
136 technology. SaskPower's Boundary Dam Integrated Carbon Capture and Storage
137 (ICCS) Demonstration Project (that comes online on 2nd October, 2014) captures
138 over one million metric tons of CO₂ per year, reflecting a 90% CO₂ capture rate for
139 the 139 MWe coal-fired unit. This is the first commercial CCS plant in the world[79],

140 The demonstration plant of Southern Company's 25 MWe Plant Barry CCS project in
141 Alabama, USA using Mitsubishi Heavy Industries (MHI) technology has been
142 operational since Jun. 2011 and it reached full-scale capture of 500 tonnes a day in
143 September 2012 [80].

144 Petra Nova/NRG 240 MWe W.A. Parish project using the MHI technology is the
145 largest commercial PCC using solvent project in the world. It is located in southwest
146 of Houston, Texas, USA. It is installed on an existing coal-fired power plant and is
147 expected to be operational in 2016 [81]. The plant is expected to capture 1.6 million
148 tons of CO₂ annually for use in enhanced production at mature oil fields in the Gulf
149 Coast region [81]

150 However, there are two main barriers for the PCC using solvent process to be
151 commercially deployed: (a) huge capital cost; (b) high thermal efficiency penalty due
152 to solvent regeneration. Therefore it is necessary to improve the technologies that
153 can reduce various costs in PCC.

154 *1.5 Motivation for using PI in PCC with solvents process*

155 It was reported that a 500 MWe supercritical coal fired power plant operating at 46%
156 efficiency (LHV basis) releases over 8,000 tonnes of CO₂ per day [82]. PCC using
157 solvents based on the conventional technology (i.e. using packed columns) requires
158 very large packed columns. Dynamic modelling and simulation study of a 500MWe
159 sub-critical coal-fired power plant by Lawal *et al.* [11] showed that two absorbers of
160 17m in packing height and 9m in diameter will be needed to separate CO₂ from the
161 flue gas. These huge packed columns translate into high capital costs. A significant
162 amount of steam from power plants has to be used for solvent regeneration. This
163 translates into high thermal efficiency penalty. It is reported that 3.2 to 4.5 MJ energy
164 is required to capture per kg of CO₂ using MEA solvent [11,54,68,83,84]. Technical
165 approaches such as heat integration, inter-cooling among others can reduce the
166 operating cost slightly. However, they limit the plant flexibility and will make operation
167 and control more difficult [54]. On the other hand, PI has potentials of significant
168 capital cost reduction [85-89], and also to improve process dynamics.

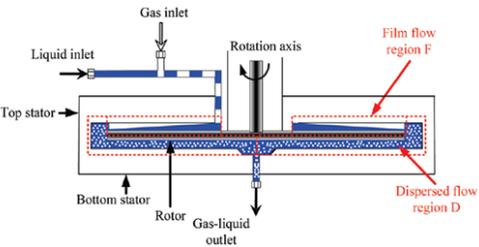
169 *1.6 Introduction to PI and Evaluation of different PI technologies for PCC*

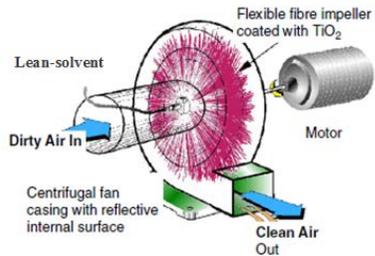
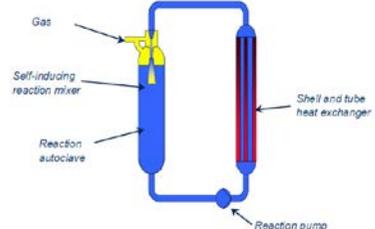
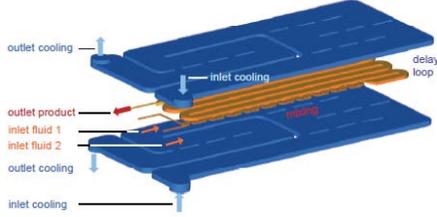
170 According to Reay [86], process intensification (PI) can be defined as: “Any
171 engineering development that leads to a substantially smaller, cleaner, safer and
172 more energy efficient technology.” There are general approaches to PI with the aim
173 to improve process performance [86]: (a) Reducing equipment size using an
174 intensified field (e.g. centrifugal, electrical, microwave); (b) Simplifying processes by

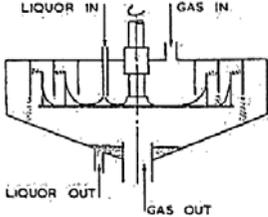
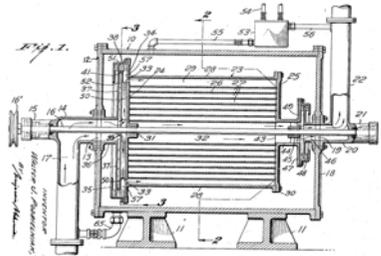
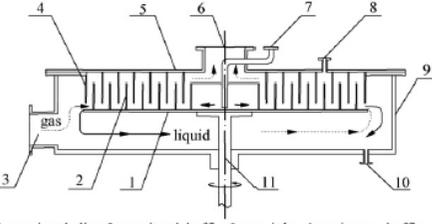
175 integrating multiple process tasks in a single item of equipment; (c) Reducing
176 equipment size by reducing its scale of structure.

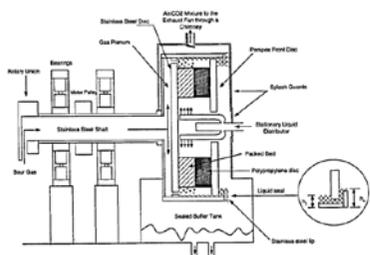
177 PI technologies differ in functions and areas of application. Some will be very good at
178 intensifying mass transfer, whilst others are good at intensifying heat transfer. Some
179 typical PI studies are presented in Table 2 to evaluate the most preferred option for
180 CO₂ capture application.

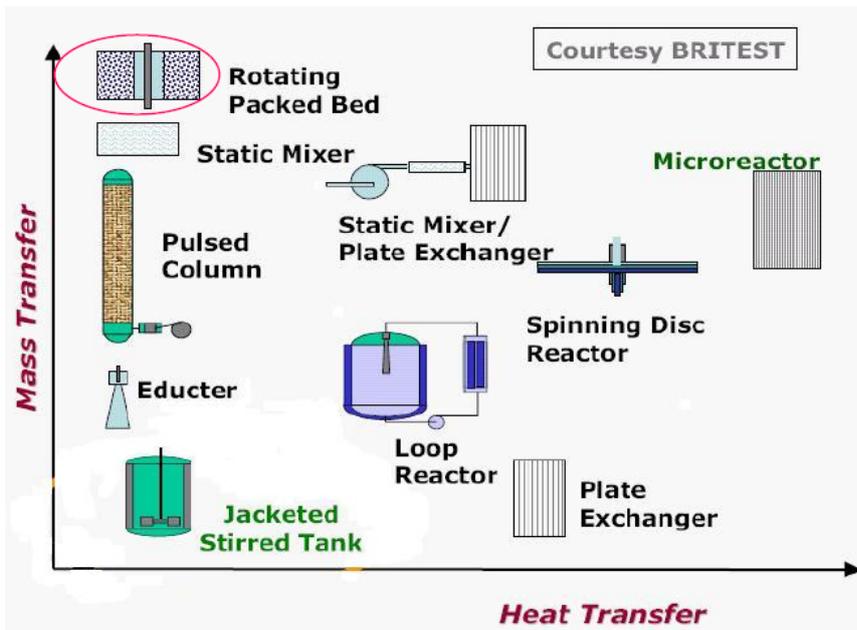
181 Table 2 Summary of Different PI Technologies

PI Equipment	Description	Mechanism for Intensification	Area of application at present	Suitability to CO ₂ capture	Limitation
Static mixer	<p>A static mixer or motionless mixer is a device inserted into a housing or pipeline with the objective of manipulating fluid streams. Different designs are available, typically consisting of plates or baffles positioned in precise angles in order to direct flow, increase turbulence and achieve mixing and reactions [90].</p> 	<p>Static mixer functions to divide, recombine, accelerate / decelerate, spread, swirl or form layers of fluid streams as they pass through the mixer. mixture components are brought into intimate contact thereby enhancing reaction processes [90]</p>	<p>Mostly this equipment is used for liquid system. e.g. Waste water treatment process (Formose treatment) [91]</p>	<p>Because of its high mass transfer capability, this can be used for CO₂ capture by combining the flue gas stream and the solvent stream. At the exit, a flash drum can be used to separate the treated gas and the rich solvent.</p>	<p>The challenge would be the high volume of flue gas to be treated (For example, 500 MWe coal-fired power plant releases 8,000 tonnes/day of pure CO₂).</p>
Spinning disc	<p>Spinning disc reactor consists of a cylindrical housing with a rotating disc in the middle between the top and bottom plates of the cylinder, the rotating disc is connected to a motor [92-94]</p> 	<p>Gas and liquid are fed together through the inlet in the centre of the top stator, close to the rotating axis. A liquid film is present on top of the rotor, and a gas-liquid bubbly dispersion is located between the rotor and the bottom stator. The combined film flow on the rotor and the dispersed flow in the remainder part of rotor-stator reactor lead to higher gas-liquid mass transfer rate [92].</p>	<p>It is used for gas-liquid and liquid-solid mass transfer process such as desorption of oxygen from oxygen saturated water, polymerization, crystallization [92-97].</p>	<p>It has the potential for CO₂ absorption, either as an absorber because of its high mass transfer capability or as a stripper because of its high heat transfer ability. It can also be used as a reboiler.</p>	<p>It may result in additional unit such as flash drum to separate the rich solvent from treated gas stream since the flow is co-current.</p>

<p>Mop fan</p>	<p>Mop fan is a device that uses flexible fibre needle impeller instead of blade impeller within a centrifugal fan casing. The flexible fibre needle impeller is connected to a motor [98]</p> 	<p>Dirty air enters the mop through its centre contacting the solvent which is sprayed on the fibre needle impeller. The rotating mop leads to increase in interfacial area for capture.</p>	<p>Mop fan is used for removal of air-borne particulates [99]</p>	<p>The system uses a flexible fibre needle impeller (mop fan) device to increase the heat and mass transfer in the absorber for CO₂ capture [98-100].</p>	<p>Huge volume of flue gas to be treated from power plants means there needs to be many mop fans for CO₂ capture. It may also result in additional unit such as flash drum to separate the rich solvent from treated gas stream since the flow might be co-current.</p>
<p>Loop reactor</p>	<p>Advanced BUSS Loop[®] reactor is made up of a reaction vessel, a circulation pump, a heat exchanger with a high performance gas/liquid ejector to achieve high mass transfer rates [101].</p> 	<p>A gas-liquid ejector consists of four main sections. Optional swirl device, nozzle that provides a high velocity jet of fluid to create suction of the gas in the gas suction chamber and entrain gas into the ejector. Liquid jet attaches itself to the mixing tube wall resulting in a rapid dissipation of kinetic energy. This creates an intensive mixing where the high turbulence produces a fine dispersion of bubbles [101].</p>	<p>Loop reactor is used for hydrogenation, phosgenation, alkylation, amination, carbonylation, oxidation and other gas-liquid reactions [101,102].</p>	<p>Because of high mass transfer and heat transfer it may be applicable to CO₂ capture [101-103].</p>	<p>Design modification will be required for it to be use for CO₂ capture also huge volume of flue gas that will be treated is another challenge</p>
<p>Microreactor</p>	<p>Schematic representation of a 2nd generation microreactor based on a plate design for performing mixing, gaining volume (e.g. to increase residence time) and integrating heat exchange [104].</p> 	<p>There are many designs for Microreactor, but the plate design offers an advantage of good mixing, longer residence time and better temperature control using cooling or heating plate [104]. Two fluid streams flow co-currently into the reactor where there are static mixers inside the reactor to enhance mixing before the final product comes out.</p>	<p>The technology is mostly applied to fine chemical and pharmaceutical industries for production specialized drugs and hazardous chemicals [104-106].</p>	<p>Because of high mass transfer and heat transfer, and its ability to operate at controlled temperature, it has good potential for CO₂ capture [105,106].</p>	<p>The expected challenge of using this technology for carbon capture is the huge volume of flue gas to be treated.</p>

<p>Chamber's centrifugal absorber</p>	<p>This centrifugal absorber was introduced by Chambers and Wall in 1954. The lower plate is rotating while the upper plate is static. Mass transfer occurs in the intermesh of concentric rings as the lean solvent contact the flue gas. No packing was utilized [107].</p> 	<p>The rotation of the lower plate makes the liquid coming into the chamber to splash as it is thrown out while the flue gas contact the liquid counter-currently leading to mass transfer in the concentric regions of the absorber [107].</p>	<p>It is used for CO₂ capture as reported by Chambers and Wall [107].</p>	<p>With more modifications in design (to incorporate packing in its bed so as to increase surface area of contact) and construction material (e.g. corrosion resistance materials), it can be used for CO₂ capture.</p>	<p>Maximum CO₂ recovery was reported to be 85%, this is below the standard for CO₂ capture above 90%.</p>
<p>Podbielniak's deodorizer</p>	<p>The contactor is basically a cylindrical rotor mounted on a shaft with contacting material [108].</p> 	<p>Gas and liquid meet counter-currently with the help of contacting material as the contactor rotates [108].</p>	<p>Used for stripping out odour and flavour substances from triglyceride oil using steam [108].</p>	<p>Has the potential for CO₂ capture especially as stripper.</p>	<p>Design modification will be needed when using it for CO₂ capture.</p>
<p>Rotating Zigzag bed</p>	<p>The RZB is characterized by a rotor coaxially combining a rotating disc with a stationary disc [109-111].</p>  <p>1-rotational disc 2-rotational baffle 3-gas inlet 4-stationary baffle 5-stationary disc 6-gas outlet 7-liquid inlet 8-Intermediate feed 9-rotor casing 10-liquid outlet 11-rotating shaft</p>	<p>In the interior of rotor, the gas flows along a zigzag path and the liquid experiences repeated dispersion and agglomeration. There are two types of gas-liquid contact. The first step is cross-current contact of two phases when the liquid is thrown by the rotational baffles. The second step is counter-current contact of two phases when the liquid falls down along the stationary baffles [109,110].</p>	<p>RZB can function without liquid distributors, eliminate one dynamic-seal, and easily accommodate and accomplish intermediate feeds in continuous distillation processes [109-111].</p>	<p>It has the potential for CO₂ absorption because of its high mass transfer capability [109-111].</p>	<p>No known limitation to CO₂ capture.</p>
<p>Rotating Packed Bed</p>	<p>The HiGee machine was constructed using a doughnut-shaped rotor, which is mounted on</p>	<p>This technology takes advantage of centrifugal fields as stimulants</p>	<p>Many studies such as Jassim <i>et al.</i>,</p>	<p>RPB has the potential for CO₂</p>	<p>No known limitation to CO₂ capture</p>

	<p>a shaft, and filled with high specific area packing [85]</p> 	<p>for process intensification. Increasing the centrifugal acceleration improves the slip velocity, which in turn improves the flooding characteristics and interfacial shear stress, and consequently boosts the mass transfer coefficient [85].</p>	<p>[85], Joel <i>et al.</i>, [87,89], Cheng and Tan [112], Cheng <i>et al.</i>, [113], Cheng and Tan [114] showed its application to CO₂ capture was successful.</p>	<p>capture [85,87-89,112,114-118].</p>	
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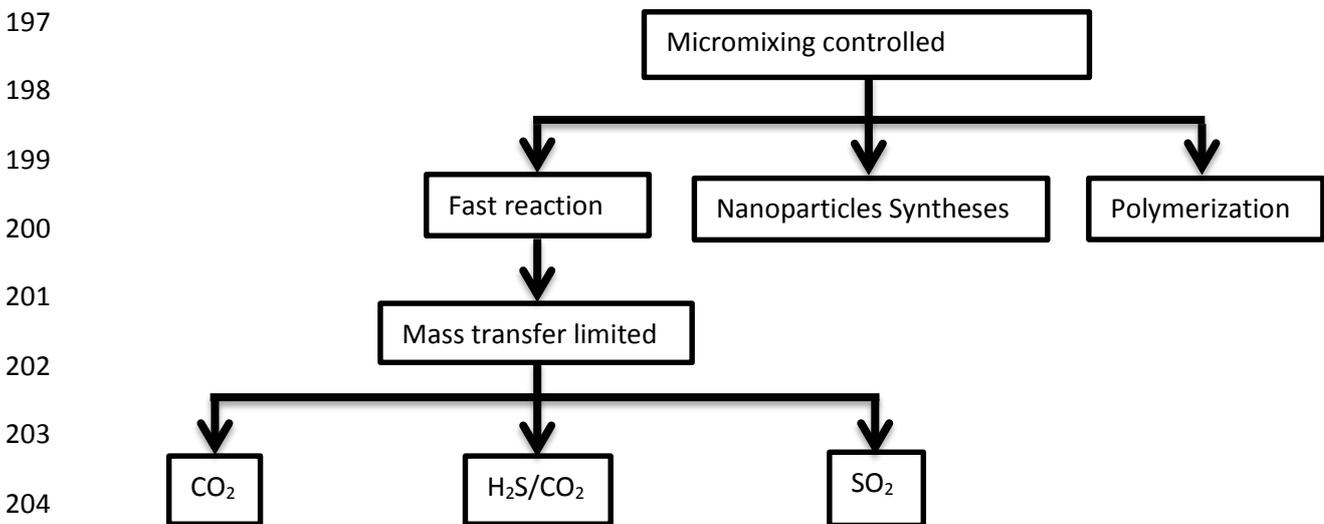


183

184 Figure 2 Mass transfer capacity in various devices [80]

185 Out of all the PI technologies studied, RPB proves to be the most suitable for
 186 intensified PCC process because of its high mass transfer performance. Figure 2
 187 shows the study done by BRITEST which indicates that RPB technology gives the
 188 best mass transfer capability compared to all other mass transfer technologies [119].
 189 Ramshaw and Mallinson [120] reported enhancement in mass transfer when using
 190 RPB. Zhang *et al.* [121] also reported that there is at least one order magnitude
 191 improvement in liquid phase mass transfer when compared to conventional packed
 192 bed.

193 Figure 3 summarised why PI technology is necessary in capture of carbon dioxide,
 194 first because reaction between CO₂ and its absorbent is a fast reaction which means
 195 it is a micromixing controlled process. Secondly it falls in the category of processes
 196 that are mass transfer limited [85,119].



205 Figure 3 Summary of why PI for PCC [80]

206 1.7 *Aim and Novelty of the Paper*

207 PCC using packed column (i.e. conventional technology) in the context of power
208 generation may result in increase of the electricity cost by more than 50%. This has
209 led to the need to search for alternative technologies since heat integration, inter-
210 cooling among others limit the operational flexibility and make control of the
211 technology more difficult [54,63,64,66]. PI technology offers the benefit of
212 significantly reducing the size of columns without compromising its production
213 capacity [120].

214 This paper aims to evaluate current status of intensified PCC (based on chemical
215 absorption) process regarding experimental rigs, experimental studies, intensified
216 heat exchanger, solvent selection, modelling and simulation and to identify the
217 knowledge gap that exists in using PI for intensified PCC process with solvents. It
218 was found that all of the experimental rigs available in the world operate as
219 standalone intensified absorber or standalone intensified stripper. Therefore no study
220 on the integrated intensified PCC process was presented in any open literature. New
221 process flow diagram (PFD) of the integrated intensified PCC process is proposed in
222 Section 7.2 of this paper. Preliminary technical and economic analysis for intensified
223 PCC process compared with conventional PCC process is presented in Section 7.7.
224 Other areas that needs research efforts on use of PI for PCC process are study of
225 Marangoni effect, systematic experimental studies, selection of suitable solvents,
226 dynamic modelling and simulation, CFD study for scale-up, optimization, techno-
227 economic analysis and life cycle analysis (LCA).

228 **2. RPB Absorber: Current Status of Experimental Rigs and Experimental**
229 **studies**

230 2.1 *RPB Absorber Experimental Rigs*

231 Since Mallinson and Ramshaw [120] introduced RPB in late 1970s, interest has
232 been continuously increasing in the use of RPB as an absorber for PCC.

233 2.1.1. *Newcastle University in the UK*

234 The group at the School of Chemical Engineering and Advanced Materials,
235 Newcastle University carried out studies on CO₂ absorption and desorption in
236 aqueous monoethanolamine (MEA) solutions in RPB. CO₂ was captured at two
237 solvents flow rates of 0.66 and 0.35 kg/s having different MEA concentrations of 30
238 wt%, 55 wt%, 75 wt% and 100 wt%. The flue gas is at a flow rate of 2.86 kmol/hr
239 [85]. The RPB rig has internal diameter of 0.156 m, outer diameter 0.398 m and axial
240 height of 0.025 m. Presently the RPB rig have been changed as reported by Lee *et*
241 *al.* [122], the new RPB rig has outer diameter of 1 m, internal diameter of 0.19 m and
242 axial depth of 0.05 m. Newcastle team is also looking at using different packing types
243 (Expamet and Knitmesh) in the RPB bed. Expamet in the inner section of the RPB

244 packing while Knitmesh at the outer section of the packing to reduce or avoid the
245 problem known as the end effect.

246 2.1.2. *BUCT in China*

247 Counter-current flow RPB was reported by Yi *et al.* [123] using Benfield solution
248 (amine-promoted hot potassium carbonate solution). The experimental rig has the
249 following specifications: inner and outer radius of the packing 0.040 m and 0.100 m
250 respectively, and axial depth of 0.031 m. Zhang *et al.* [121] reported RPB absorber
251 using ionic liquid with the following rig specifications: inner diameter of 0.020 m,
252 outer diameter 0.060 m and axial packing depth of 0.020m. Luo *et al.* [124] proposed
253 correlation for gas-Liquid effective interfacial area in a RPB using NaOH solvent to
254 absorbed CO₂. Luo *et al.* [124,125] reported the following rig specifications: inner
255 and outer radii of the rotor were 0.078 m and 0.153 m respectively, and the axial
256 height of the rotor was 0.050 m. The static casing had an inner radius of 0.248 m
257 and an axial height of 0.098 m. Zhang *et al.* [121] found that liquid side volumetric
258 mass transfer coefficient for RPB has been improved to around $3.9 \times 10^{-2} \text{ s}^{-1}$
259 compared with $1.9 \times 10^{-3} \text{ s}^{-1}$ for the conventional packed column under the same
260 operating conditions.

261 2.1.3. *Taiwan (National Tsing Hua University, Chang Gung University and Chung 262 Yuan University)*

263 The group in National Tsing Hua University in Taiwan studied CO₂ capture using
264 counter-current flow arrangement. Yu *et al.* [126] studied CO₂ capture by
265 alkanolamine solutions containing diethylenetriamine (DETA) and piperazine (PZ) in
266 an RPB. Cheng and Tan [114] studied removal of CO₂ from indoor air by
267 alkanolamine in RPB, the experimental rig used is the same as for Tan and Chen
268 [127] and Yu *et al.* [126]. The diameter of the static casing of the RPB was 0.228 m,
269 the inner and outer diameters of the packing in the RPB were 0.076 and 0.16 m
270 respectively, and the height was 0.02 m. The total volume of the packing in the RPB
271 was of 0.0003114 m³. Stainless wire mesh with a specific surface area of 803 m²/m³
272 and a void fraction of 0.96 was packed in the bed acting as packing. In all these
273 studies, there is significant improvement in overall mass transfer and height of
274 transfer unit (HTU) demonstrating the performance superiority of an RPB compared
275 to conventional packed bed. Overall mass transfer coefficient (K_{Ga}) and HTU
276 corresponding to the most appropriate operating conditions in RPB were found to be
277 higher than 5.8 s⁻¹ and lower than 1.0 cm [114]. However HTU is around 40 cm for
278 conventional PCC process.

279 The group in Chang Gung University in Taiwan studied RPB absorber using cross-
280 flow RPB for CO₂ capture which is believed to have relatively small gas flow
281 resistance and unaffected by centrifugal force. Another benefit of cross-flow is higher
282 gas velocity due to no critical gas velocity limitation. Lin *et al.* [128] and Lin and Chen
283 [129] found that cross-flow RPB absorber is effective for CO₂ absorption process. In
284 their study, the following rig specifications were used: inner radius of 0.024 m, an

285 outer radius of 0.044 m, and an axial length of 0.12 m, specific surface area of
286 $855\text{m}^2/\text{m}^3$ and a voidage of 0.95.

287 2.1.4. India

288 India Institute of Technology (IIT) Kanpur reported the use of RPB as absorber in
289 CO_2 absorption process using split packing. The two packing discs can rotate co-
290 currently or counter-currently. Their work shows that counter-current rotation of the
291 packing disc gives better performance. The technology improves slip velocity to as
292 high as 30 m/s [130]. It was also reported in their study that this type of RPB shows
293 good performance in gas phase control processes by enhancing volumetric mass
294 transfer coefficient on the gas side to about 35 – 280 times compared to those of
295 packed columns, the liquid side volumetric mass transfer coefficient enhances in the
296 range of 25 – 250 times compared to the packed column [130]. Agarwal *et al.* [131]
297 stated that continuous single block packing causes a significant reduction only in the
298 liquid side mass-transfer resistance with little or no reduction in the gas side
299 resistance. Rajan *et al.* [130] studied RPB absorber in NaOH solution and found that
300 the split packing gives the highest volumetric liquid side mass transfer coefficient as
301 compared to conventional packed column.

302 2.2 Experimental Studies on Intensified Absorber

303 Significant progress has been witnessed in the area of intensified absorber, from the
304 early work published by Chamber's Wall in early 1950's to what we have at present.
305 Newcastle research team in the UK as reported by Jassim *et al.* [85] was able to
306 establish that RPB has the potential to dramatically reduce the size and cost of
307 carbon capture units for power plants. Lee *et al.* [122] reported that there are some
308 uncertainties which need to be tackled such as (a) Power consumption; (b) Pressure
309 drop; (c) Viscous liquid distribution in a RPB, this leads to the modification of the
310 experimental rig used by Jassim *et al.* [85] with the aim of addressing these
311 challenges.

312 Research Groups in China and Taiwan also made significant progress towards the
313 commercialization of this technology. The early report of the technology based on
314 counter-current flow has now turned to look at the other flow geometries such as co-
315 current and cross-flow arrangement to take the advantage of lower gas phase
316 pressure drop and eliminating the need for air blower.

317 The research Group In India studies split packing RPB which has been suggested to
318 increase the gas-phase mass transfer coefficient as against continuous packing RPB
319 which is said to have the same gas phase mass transfer coefficient as conventional
320 packed column [132].

321 **3 RPB stripper: Current status of experimental rigs and experimental**
322 **studies**

323 *3.1 RPB stripper experimental rigs*

324 Study on intensified stripper experimental rig for intensified PCC process was
325 published by just two research groups: Newcastle University in the UK and National
326 Tsing Hua University in Taiwan. With both groups, reboiler is the same as that of
327 conventional packed column (i.e. big in size).

328 *3.1.1. Newcastle University, UK*

329 Jassim *et al.* [85] reported RPB stripper for desorption runs for 30 wt%, 54 wt% and
330 60 wt% MEA solution having flow rate range from 0.2 kg/s to 0.6 kg/s. The rich MEA
331 solution is preheated to the temperature range between 57 °C and 70 °C and at
332 atmospheric pressure before being sent into the intensified stripper. The RPB
333 stripper has internal diameter of 0.156 m, outer diameter 0.398 m and axial height of
334 0.025 m. The packed bed material was stainless steel expamet with very high
335 specific surface area (2132 m²/m³) and a moderate voidage (0.76). Comparison
336 between RPB stripper and conventional stripper operating at the same performance
337 was done by Jassim *et al.* [85] which shows that the intensified stripper height is
338 reduced by a factor of 8.4 and its diameter is reduced by a factor of 11.3.

339 *3.1.2. Taiwan*

340 With conventional packed columns, energy consumption in capturing CO₂ from a
341 conventional coal-fired power plant ranges from 3.24 to 4.2 GJ/tonne CO₂ [20].
342 Kothandaraman *et al.* [133] noted that the majority (approximately 62%) of the
343 energy consumed during the CO₂ capture process was required for the regeneration
344 of solvent. Moreover, the consumption of steam is the most important component of
345 the operating costs of alkanolamine absorption as reported in Chapel and Mariz
346 [134]; Tobiesen and Svendsen [135].

347 In order to regenerate CO₂-rich solvent solution at a temperature higher than 100 °C
348 and a pressure higher than atmospheric pressure, Cheng *et al.* [88] introduced a
349 back pressure regulator, this is the major difference between Jassim *et al.* [85] setup
350 and Cheng *et al.* [88] setup. Because of the back pressure regulator, the
351 regeneration could be operated at various temperatures over a fixed pressure, and
352 vice versa. Cheng *et al.* [88] rig specifications are as following: the diameter of the
353 static casing of the RPB was 0.228 m, the inner and outer diameters of the packing
354 in the RPB were 0.076 and 0.16 m respectively, and the packing height in the RPB
355 was 0.02 m. The total volume of the packing in the RPB was 0.000311 m³. For the
356 RPB packing, a stainless wire mesh with a specific surface area of 803 m²/m³ and a
357 void fraction of 0.96 was packed into the bed.

358 The use of an RPB apparatus instead of a packed bed not only dramatically reduces
359 the required stripper volume but also consumes less regeneration energy [88].

360

361 3.2 *Experimental studies on Intensified Regenerator*

362 Intensified regenerator for CO₂ capture was reported by Jassim *et al.* [85] and
363 Cheng *et al.* [88]. In both studies, they found that RPB regenerator can significantly
364 reduce the size of column as compared to conventional stripper, but the reboiler size
365 is still as big as before since it has not been intensified. Cheng *et al.* [88] improved
366 from Jassim *et al.* [85] by introducing a back pressure controller, this will help in
367 operating the regenerator at higher pressure and temperature.

368 **4 Intensified Heat Exchanger**

369 In conventional PCC process, there is a cross heat exchanger. This is huge in
370 volume. In addition to this, the piping surrounding the cross heat exchanger has
371 high footprint. Therefore, the cross heat exchanger has to be intensified.

372 *4.1 Technologies available to choose*

373 *4.1.1 Printed Circuit Heat Exchanger (PCHE)*

374 The PCHE was invented in 1980 in Australia and applied to refrigerators in 1985 by
375 Heatric (UK) [136]. The PCHE is a high-integrity plate type compact heat exchanger
376 in which fluid flow channels are produced by chemical etching on flat metal plates.
377 Etched plates are stacked to produce single block by diffusion bonding [136-139].

378 Because of the compactness provided by PCHE design, the volume of PCHEs are
379 typically 4–6 times smaller and lighter than conventional shell-and-tube heat
380 exchangers designed for the same thermal duty and pressure drop as shown in
381 Figure 4 [140,141]. Low pressure drop in PCHE can be found based on design. Kim
382 *et al.* [138] compared air-foil fin PCHE and zigzag channel PCHE which have the
383 same heat transfer performance but the pressure drop of airfoil fin PCHE is one-
384 twentieth of zigzag PCHE. PCHE effectiveness was reported to be more than 98%
385 and can operate at maximum allowable pressure of 600 bar and more than 800 °C
386 maximum operating temperature (limited by material of construction). PCHE has
387 multi-fluid capability (intensification achieved by reducing the number of exchanger
388 units) [141].



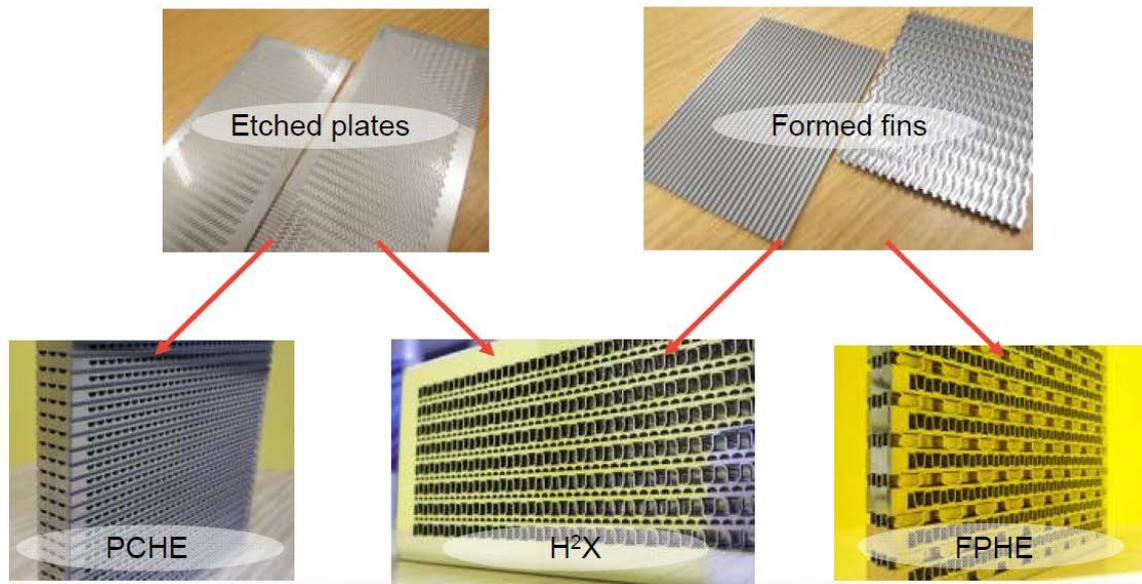
389
390 Figure 4 Printed Circuit Heat Exchanger (Courtesy of Heatric Ltd): the big one at the
391 back is shell-and-tube heat exchanger; while the small one in front is PCHE.

392 *4.1.2 Formed Plate Heat Exchanger (FPHE)*

393 The FPHE uses the same concept as various fin plate heat exchangers, but adds the
394 advantage of replacing the brazing process with the diffusion-bonding process. This
395 is shown in Figure 5. Heatric Ltd reported that FPHE has bigger channels size (about
396 3mm x 3mm) than the PCHE, this leads to lower pressure drop than PCHE [141].
397 FPHE has effectiveness of more than 98%. Maximum allowable pressure for FPHE
398 is 200 bar and can operate at higher temperature of up to 800 °C, it has multi-fluid
399 capability (Intensification by reducing number exchanger units)[141].

400 *4.1.3 Hybrid Heat Exchanger (H²X)*

401 H²X technology developed by Heatric Ltd combines the etched plates and the
402 formed fins to form what is known as Hybrid Heat Exchanger [141]. The heat
403 exchanger takes some of the advantages offered by PCHE and FPHE. H²X has
404 bigger hydraulic diameter than PCHE because of the presence of FPHE and can
405 also withstand higher pressure than FPHE because of the advantage offered by the
406 presence of PCHE. Typical hydraulic diameter of PCHE is in the range of 0.1 mm to
407 3 mm while that of FPHE is in the range of 1.2 mm to 3.3 mm [141].



408

409 Figure 5 Summary of Technology of PCHE, H2X, and FPHE (Courtesy of Heatric)

410 *4.1.4 The Marbond heat exchanger*

411 The manufacturing procedures of Marbond heat exchanger are similar to those of
 412 the PCHE. It is formed of slotted flat plates which have been chemically etched
 413 through. The plate pack is then diffusion-bonded together [142,143]. In contrast
 414 with the PCHE, several thinner, slotted plates are typically stacked to form a single
 415 sub-stream, thus giving the potential for very low hydraulic diameters [142]. In some
 416 applications, it has a substantially higher area density than the PCHE. For example,
 417 a doubling of porosity, other factors being equal, results in a halving of the volume
 418 for a given surface area [144].

419 *4.1.5 Spiral heat exchanger (SHE)*

420 SHE refers to a helical tube configuration. The term refers to a circular heat
 421 exchanger with two long metal strips of plate rolled together to form a pair of
 422 concentric spiral channels of rectangular cross-section, one for each fluid. The
 423 passages can be either smooth or corrugated, in some cases studs are welded onto
 424 one side of each strip to fix the spacing between the plates, to provide mechanical
 425 strength and to induce turbulence that increases heat transfer [145].

426 The internal void volume is lower (less than 60%) than in a shell-and-tube heat
 427 exchanger [145], and this yields a compact and space-saving construction that can
 428 be readily integrated in any plant and reduces installation costs. The heat transfer
 429 surface ranges from 0.05 m² for refrigeration applications up to about 500 m² with a
 430 maximum shell diameter of 1.8 m and the sheet metal thickness range is 1.8–4 mm
 431 for industrial processes [146]. The surface area requirement is about 20% lower than
 432 that for a shell-and-tube unit for the same heat duty [145]. SHEs are often used in
 433 the heating of high viscosity and dirty fluids. It exhibits lower tendency to fouling

434 [147]. When a SHE requires cleaning, all heat transfer surfaces are readily
435 accessible by simply removing the heads.

436 4.2 *Recommendation for intensified Heat Exchanger for PCC application*

437 To make a decision on best intensified heat exchanger to be used for PCC
438 application, many factors need to be considered. Some of them are listed as follows
439 [140]: (a) Operating Pressure limits; (b) Thermal performance (also known as the
440 effectiveness of the heat exchanger); (c) Expected working temperature range; (d)
441 Product mix to be used in the exchanger (liquid-to-liquid or gas-to-gas); (e) Pressure
442 drop desired across the expected heat exchanger; (f) The expected fluid flow
443 capacities over both sides of the heat exchanger; (g) Method of cleaning employed,
444 maintenance and repair issues associated with heat exchanger; (h) Materials
445 required for construction; (i) Ease of expansion of exchanger when it becomes
446 necessary; (j) The cost of the heat exchanger .

447 Compromise would therefore have to be made in most cases when selecting a heat
448 exchanger. For instance, cost of the exchanger is a paramount factor, but it should
449 not be the determining factor. If just for a cheaper heat exchanger, certain desired
450 performance demands of the heat exchanger would have to be forfeited.

451 The authors believe that the PCHE and the Marbond heat exchanger look promising
452 for use in intensified PCC process because of its many benefits such as high
453 efficiency (>98%), Compactness to improve safety and economics, weight saving,
454 low pressure drop, high temperature and retrofit options [141-143]. PCHE has been
455 reported to have additional advantage of being multi-fluid, meaning it can be used for
456 preheating of rich-MEA stream and also as a condenser for CO₂ – stream.

457 5. **Solvents used for intensified PCC process**

458 There are extensive studies in solvent selection from both academia and industry
459 trying to identify alternative solvents for conventional PCC process. There are very
460 few studies on PI using different solvents [85,87,89,121,123-126,128,129,148-150].

461 5.1 *Factors to consider*

462 Factors to consider when conducting solvent screening for conventional PCC
463 process and intensified PCC process is similar to some extent but the major
464 difference comes from residence time of solvent in different technologies. In
465 intensified PCC process, the residence time is relatively short (less than 10% of the
466 conventional PCC process). Therefore the factors to consider are: (1) CO₂
467 absorption reaction kinetics, (2) CO₂ absorption capacity, (3) heat of absorption, (4)
468 solvent toxicity, (5) solvent volatility, (6) solvent corrosivity, (7) solvent degradation,
469 (8) solvent foaming, (9) solvent viscosity, (10) solvent surface tension and (11) cost.

470 (1) CO₂ reaction kinetics: This determines the rate at which CO₂ will be captured.
471 Fast reaction kinetics is essential for intensified PCC process since the residence
472 time is very short.

473 (2) CO₂ absorption capacity: This is related to the solvent flow rate required and
 474 the sensible heat requirement. Higher CO₂ absorption capacity would require lower
 475 solvent flow rate and subsequent less regeneration energy demand.

476 (3) Heat of absorption: This would be an important factor affecting reboiler heat
 477 duty. Lower heat of absorption will require less regeneration energy input to reverse
 478 the chemical reaction and release absorbed CO₂.

479 (4) Solvent stability, operational issues and environmental impact: These are the
 480 other factors to be evaluated when selecting solvents. Solvent degradation (which
 481 may be controlled by having high stability against oxygen and thermal stress) and
 482 corrosion will cause an increase in operation and maintenance (O&M) costs by
 483 making up solvent and reducing the lifetime of the equipment. Higher solvent
 484 viscosity would increase the pump work in circulating the solvent between the
 485 absorber and regenerator. Cost and availability of potential solvents in commercial
 486 scale could contribute to limitations of the process feasibility. Environmental impacts
 487 such as solvent toxicity and volatility deserve serious attention when judging the
 488 potential of a solvent since causing secondary pollution while capturing CO₂ is not a
 489 scenario the public would be willing to take.

490 Other solvent characteristics such as surface tension and foaming tendency are also
 491 important factors to consider when judging a solvent's potential. Table 3 gives
 492 evaluation of solvent properties based on relative importance on a scale of 0 to 10,
 493 with 10 being the most important property and 0 the least important property [151].
 494 These may provide insights for solvent selection in intensified PCC process.

495 Table 3 Evaluation of Solvent Properties [151]

Property	Importance	Potential Show-stopper	Evaluation Methods
Reaction Kinetics	10	Yes	Literature, wetted-wall column(WWC)
Absorption Capacity	10	Maybe	Calculation from VLE
Heat of absorption	10	Yes	Literature, Calorimetry Measurement, Calculation from VLE
Toxicity	6	Yes	Literature/material safety data sheet(MSDS)
Volatility	4	Maybe	Literature
Corrosivity	6	Maybe	Literature, Laboratory Test
Degradation	6	Yes	Literature, Laboratory Test
Foaming	4	Maybe	Literature
Viscosity	4	Yes	Literature, Pilot Test
Surface tension	4	Maybe	Literature
Cost	2	Maybe	Vendor

496 5.2 Solvents Used

497 Different solvents were used for intensified PCC process by different research
498 groups. Some researchers use one solvent while others mix solvents so as to benefit
499 from properties each solvent offers.

500 5.2.1. Alkanolamine

501 The use of MEA for CO₂ capture in RPB was reported in Jassim *et al.* [85]. MEA has
502 high reactivity but is rapidly replaced by more efficient solvents because of its
503 corrosive nature, toxicity and high heat of reaction with CO₂. Diethanolamine (DEA)
504 is much slower to react with CO₂. It is not good for intensified PCC process itself.
505 Methyldiethanolamine (MDEA) has become an important alkanolamine because of
506 its low energy requirement, high capacity and high stability but has the disadvantage
507 of low rate of reaction with CO₂. Lin *et al.*, [128] presents study on the evaluation of
508 various alkanolamine solutions for CO₂ removal in cross-flow RPB. The reaction rate
509 of these solvents with CO₂ followed the order of Piperazine (PZ) > MEA > 2-amino-
510 2-methyl-1-propanol (AMP).

511 Yu *et al.* [126] reported study on CO₂ capture by alkanolamine solutions containing
512 diethylenetriamine (DETA) and PZ in RPB. They found that the CO₂ capture
513 efficiency of DETA in terms of overall mass transfer coefficient $K_G a$ and HTU was
514 superior to that of MEA in RPB. This is because DETA possesses higher CO₂
515 absorption capacity and reaction rate with CO₂ than MEA. Higher boiling point and
516 lower vapour pressure of DETA will lead to lower energy requirement and less loss
517 of solvent in stripper compared with MEA, suggesting DETA as a promising solvent
518 to substitute MEA for CO₂ capture. The mixed solution DETA + PZ exhibited higher
519 CO₂ capture efficiency than DETA indicating PZ was a great promoter for capturing
520 CO₂. This was because the promoter PZ possesses higher reaction rate with CO₂
521 than DETA [126].

522 5.2.2 NaOH

523 Munjal *et al.* [152] reported the use of NaOH for absorption of CO₂. Their study
524 shows that the gas-liquid mass transfer could be improved. Lin *et al.* [153] compared
525 the overall volumetric mass-transfer coefficient ($K_G a$) of RPB for different solvents
526 (i.e. NaOH, MEA and AMP) and found that $K_G a$ values for the CO₂-MEA system
527 were approximately 2-5 times higher than those for the CO₂-AMP system also $K_G a$
528 values for MEA were at least 20% higher than those for NaOH at the same operating
529 conditions. Therefore rate of reaction for CO₂ capture in RPB follows the order MEA
530 > NaOH > AMP [154]. But AMP has higher absorption capacity than MEA. Lin and
531 Chen [81], Luo *et al.* [125] studied chemisorption's of CO₂ using NaOH in RPB. They
532 found that NaOH has the potential for use as solvent in RPB, but one of the major
533 challenges is the formation of stable salt which make solvent regeneration difficult.

534 5.2.3 *Ionic liquid (1-n-butyl-3-methylimidazolium hexafluorophosphate)*

535 The use of ionic liquids for CO₂ capture is gaining interest due to their unique
536 characteristics (i.e., wide liquid ranges, thermal stabilities, negligible vapour
537 pressures up to their thermal decomposition points, tunable physicochemical
538 characters, and high CO₂ solubility) [121,155]. However, ionic liquids are commonly
539 high or superhigh viscosity liquids with poor fluidities. A significant limitation for large-
540 scale application of a continuous CO₂ capture process for conventional packed
541 columns by ionic liquid is the great resistance of mass transfer and low gas – liquid
542 mass transfer rate due to the high viscosity. As reported in Chen *et al.* [118], the
543 dependence of $k_L a$ on liquid viscosity in RPB is less than that of packed column.

544 5.2.4 *Potassium Carbonate (K₂CO₃)*

545 The use of K₂CO₃ is receiving great attention because of its high CO₂ absorption
546 capacity. Firstly, K₂CO₃ is a more efficient solvent for CO₂ than either MEA or DEA
547 [156]. This means that for a given amount of solvent, K₂CO₃ can absorb more than
548 the other two solvents. In addition, the cost of this solvent is lower because less is
549 needed and K₂CO₃ is cheaper than other traditional solvents [156]. Secondly, this
550 eliminates the need for cross heat exchanger because the stripper runs at lower
551 temperature than the absorber [156]. Thirdly, K₂CO₃ increases the safety in CO₂
552 removal by not only absorbing CO₂ but also small amounts of hydrogen. Hydrogen
553 possesses a safety threat. Flashing off hydrogen can cause fires or explosions if
554 proper precautions are not taken. Lastly K₂CO₃ is not volatile, which means minimal
555 losses of the solvent with the exit gas occur. Since K₂CO₃ is not prone to the
556 degradation reactions associated with MEA, there is no loss of solvent associated
557 with degradation [133]. However one of the major drawbacks of using K₂CO₃ in RPB
558 is its low rate of reaction. This necessitated the need for promoter so as to increase
559 its rate of reaction. Kothandaraman *et al.* [133] reported regeneration energy (without
560 energy recuperation) of 3.2 MJ/kg for K₂CO₃ when treating flue gas (12 vol% CO₂).

561 5.2.5 *Benfield solution (Amine-promoted hot K₂CO₃ solution)*

562 Amine-promoted hot K₂CO₃ solution, which is called Benfield solution, is used in
563 Benfield process [157]. The amine promoter could significantly enhance the reaction
564 rate while the carbonate–bicarbonate buffer offers advantages of large capacity for
565 CO₂ capture and ease of regeneration [123,157]. Pilot plant studies by Field *et al.*
566 [158] shows that hot-carbonate system is particularly effective for removing CO₂,
567 especially when present at high partial pressure. Steam consumption is one-third to
568 one-half that of ethanolamine. Therefore, Benfield Process is known as an economic
569 and efficient way of removing large quantities of CO₂ from flue gases and can be
570 effectively used in RPB [123].

571 5.2.6 *Piperazine (PZ)*

572 PZ is a diamine solvent whereby one amine group is involved in a fast reaction with
573 CO₂ to form the carbamate while the other amine absorbs the released proton [159].

574 PZ reacts rapidly with CO₂ and thus has attracted interest for usage in CO₂ capture,
 575 particularly as a reaction rate promoter for CO₂ absorption in carbonate and tertiary
 576 amine solutions [148,159-162]. Chemical reactions which describe the absorption of
 577 CO₂ in PZ solutions are more complex than MEA [159,160].

578 Reaction rate between CO₂ and aqueous PZ solution is high [148,160-162].
 579 However, the absorption has to take place at high temperatures because its solubility
 580 in water is limited [161]. Freeman *et al.* [160] suggested the use of concentrated PZ
 581 solution for CO₂ capture because of its effective resistant to oxygen degradation and
 582 thermal degradation. Despite its high reactivity with CO₂, it has some challenges
 583 such as limited solubility in water and more volatile than MEA.

584 5.3 Proprietary Commercial Solvents

585 To avoid high thermal efficiency penalty due to high regeneration energy, new
 586 solvents were developed and commercialised. Econamine FG+ is MEA-based
 587 solvent with proprietary inhibitors [163,164]. Sander and Mariz [163] reported
 588 resultant solution circulation factor (m³ solvent per m³ Econamine FG+ solvent) for
 589 Econamine FG+ solvent as 1 while 18wt% MEA solvent has 1.7. The Kansai
 590 Electric power Co. and Mitsubishi Heavy Industries, Ltd. have developed new
 591 aqueous solutions of sterically-hindered amines designated as KS-1, KS-2 and KS-3
 592 [165-167]. The world's largest commercial PCC plant (Petra Nova/NRG 240 MWe
 593 W.A. Parish project) planned to use KS-1 solvent when commissioning in 2016 [81].
 594 The first commercial CCS plant (SaskPower's Boundary Dam 139 MWe project)
 595 uses Consolv solvent [79], the solvent is based on tertiary amines, and probably
 596 includes a promoter to yield sufficient absorption rates for low pressure flue gas
 597 streams. H3 solvent is Hitachi's proprietary solvent formulation which has much
 598 lower regeneration energy compared with MEA [168]. Regeneration energy and
 599 thermal efficiency penalty for different proprietary commercial solvents were
 600 compared in Table 4.

601 Table 4 Performance of proprietary commercial solvents [159]

Solvent	Regeneration Energy (GJ/t-CO ₂)	Efficiency penalty (%)	References
Econmaine FG+	3.12	9.2	IEAGHG [169]
KS-1	3.08	8.4	IEAGHG [169]
KS-2	3.0	9.3	Gibbins and Crane [170]
CANSOLV	2.33	8.2	Just [171], Shaw [172]
H3	2.8	7.8	Wu <i>et al.</i> [168], Stover <i>et al.</i> [173]

602 5.4 Recommendations on solvent selection for PCC process

603 Selection of solvents for CO₂ capture process is a very important design decision for
 604 both conventional and intensified PCC processes. Firstly, the residence time in the
 605 intensified PCC process is less than 10% of the conventional PCC process.

606 Therefore solvent for intensified PCC process should have fast kinetics to capture
607 CO₂. That is why most studies on RPB absorber uses primary or secondary
608 alkanolamines due to their fast kinetics. Concentration of the solvent in RPB is
609 usually high in order to have high reaction rate. High concentration solvent generally
610 has high viscosity, which prevents its use in conventional PCC process. However,
611 this is not a problem in RPB case. Secondly to achieve high CO₂ absorption
612 capacity and reaction kinetics fast, mixing solvents such as amine-promoted K₂CO₃
613 will play a significant role. Thirdly the regeneration energy of solvents should be low
614 in addition to fast kinetics and high absorption capacity. Oexmann *et al.* [174]
615 reported mixing MDEA and PZ gives lower regeneration energy of 2.52 GJ/t-CO₂.
616 Lastly when an intensified PCC plant is to be built in non-compliant area, volatility of
617 the solvent will have a big impact on whether or not the project will be permitted.

618 **6. Modelling/Simulation for CO₂ Capture Process using RPB**

619 Modelling and simulation can be used to circumvent the difficulties with the
620 experimental approach, and complement the experimental studies.

621 *6.1 Correlations for Mass/Heat Transfer*

622 Significant progress has been noticed in the development of correlations for
623 mass/heat transfer in RPB. Tung and Mah [132] developed correlations for liquid
624 phase mass transfer coefficient based on the penetration theory. In developing Tung
625 and Mah [132] correlation, the effect coriolis acceleration and packing material
626 geometry were neglected. Chen *et al.* [175] developed correlation that takes into
627 consideration of those terms that were neglected by Tung and Mah [132]. Chen [176]
628 reported mass transfer correlation for gas-phase mass transfer coefficients which
629 has predicted experimental data reasonably well. The challenge of predicting
630 accurately the interfacial area in RPB is another issue of concern. Onda *et al.* [177]
631 correlation for gas-liquid interfacial area for conventional packed column was
632 modified by replacing the gravity term with centrifugal gravity term, but this does not
633 take into account different types of packing factors such as wire diameter and the
634 wire mesh opening. Luo *et al.* [124] developed gas-liquid interfacial area correlation
635 for wire mesh packing which takes into account the effect of wire diameter and wire
636 mesh opening. Burns *et al.* [178] correlation predicts the liquid hold-up for a high
637 voidage structured packing in an RPB.

638 *6.2 Modelling/Simulation of Intensified Absorber*

639 Few open literature discussed modelling and simulation of intensified absorber. The
640 group in Taiwan modelled the RPB as a series of continues stirred tank with
641 contactors. Cheng and Tan [114] reported that five CSTRs with a contactor can
642 achieve the set target for a given case through simulation study. The research group
643 at University of Hull, UK reported modelling and simulation of RPB absorber using
644 Aspen Plus® and visual FORTRAN [87,89]. Their key findings include: (a) the

645 packing volume can be reduced 52 times and the absorber size can be reduced 12
646 times; (b) there is no temperature bulge observed so far inside the packing [87,89].

647 6.3 *Modelling/Simulation of Intensified Stripper*

648 Experimental studies of intensified regenerator were only reported in Jassim *et al.*
649 [85] and Cheng *et al.* [88]. No modelling and simulation of intensified stripper was
650 reported in open literature.

651 6.4 *Modelling and Simulation of the whole plant*

652 Open literature on modelling and simulation of whole intensified PCC process was
653 not available as at the time of this review.

654 7. **Prospective of applying PI Technology into PCC using solvents**

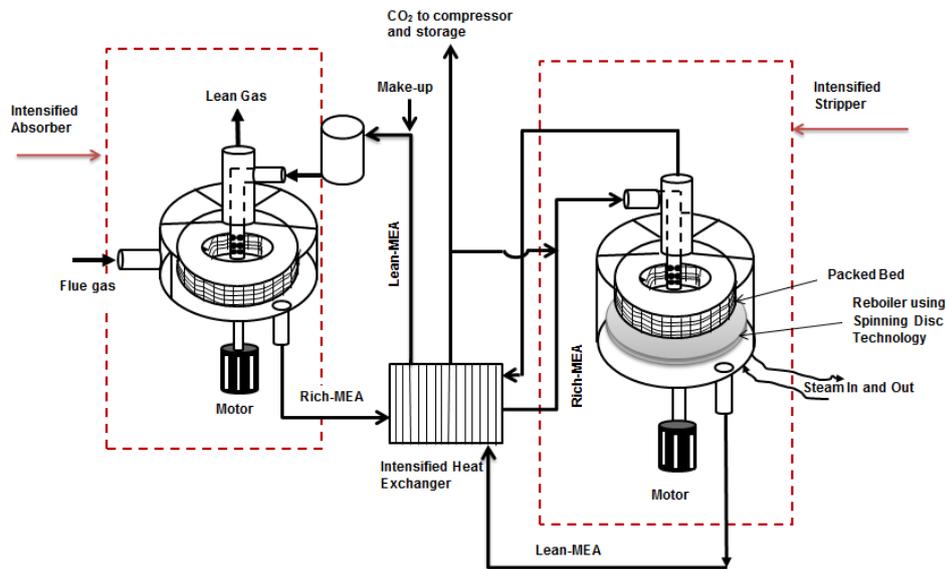
655 With the high potential to reduce capital and operating costs for carbon capture, the
656 UK Engineering and Physical Sciences Research Council (EPSRC) recently
657 awarded a consortium project worth £1.27 million to the universities of Hull,
658 Newcastle, Sheffield and Imperial College London to apply PI technology into PCC
659 with solvents process.

660 7.1 *Fundamental study regarding Marangoni effect and enhanced mass transfer*

661 Interfacial turbulence (i.e. Marangoni effect) which is caused by surface tension
662 gradient have been analysed by Semkov and Kolev [179], Kolev and Semkov [180],
663 Sternling and Scriven [181] and Buzek *et al.* [182]. The main reason for the
664 phenomenon of instability can be the local disturbances of temperature and /or
665 concentration near the interface [182]. Marangoni effect can significantly enhance
666 mass transfer rate by a factor of two or more, but it can easily be damped by a
667 surface active agent. Therefore, amine solutions used for absorption should not be
668 contaminated by even traces of surfactants [182]. A future task is to use simple
669 experiments to observe whether interfacial turbulence exists in RPB absorber.

670 7.2 Proposed schematic PFD for whole intensified PCC process

671 Since there is no pilot plant of whole intensified PCC process in the world, we
672 propose a simplified whole intensified PCC process. Figure 6 describes potential
673 technologies (RPB, Spinning disc and PCHE) proposed for the intensified PCC.



674

675 Figure 6 Simplified PFD of intensified chemical absorption process for PCC

676 Comparing Figure 6 (intensified PCC using solvent) with Figure 1 (conventional PCC
677 using solvents), there are three main differences: (1) Rich solvent is regenerated in
678 the stripper which use spinning disc reboiler incorporated outside RPB packing in
679 order to use the same motor for rotation; (2) the cross heat exchanger in the
680 conventional PCC technology is replaced with the intensified heat exchanger based
681 on PCHE. The intensified heat exchanger is multi-fluid which can be used as
682 condenser for the CO₂ stream and also pre-heating rich solvent. Therefore this
683 integration can reduces or eliminate condenser cooling cost; (3) Intensified absorber
684 and stripper are driven by motors.

685 Checked against the definition of PI in Section 1.6, the proposed PFD in Figure 6
686 achieves PI in the following ways: (a) the size of the intensified absorber and stripper
687 is reduced significantly due to the centrifugal field; (b) the reboiler and the stripper
688 are merged together into a single item of equipment. The condenser is also merged
689 into the multi-fluid intensified heat exchanger.

690 7.3 Experimental rigs and Experimental studies

691 For intensified absorber or stripper, conscious judgement on the best flow
692 arrangement (i.e. counter-current, co-current or cross-flow) is necessary in RPB
693 design of experimental rig so as to decide which flow geometry will have better RPB
694 performance and lean at minimum energy penalty (due to vapour phase pressure drop).

695 Currently, there are only standalone intensified absorber or stripper worldwide. The
696 experimental rig for the whole intensified PCC process is necessary to build so as to

697 understand the dynamic behaviour of the whole capture process and to provide
698 foundation for studies in optimal design, operation and control.

699 Mass transfer performance of the intensified absorber has been fully determined by
700 experimental studies and modelling & simulation. More studies are needed to
701 understand the actual surface area in the RPB which over the years not been fully
702 determined. Dynamic flow behaviour in RPB is another aspect that needs to be
703 studied.

704 7.4 Selection of Solvents

705 Studies in conventional packed column showed that solvent with high viscosity
706 affects its performance. Zhang *et al.* [121] reported that the impact of high viscosity
707 on CO₂ capture process is not so severe for intensified CO₂ capture process.
708 Therefore study on ionic liquid solvent which has high absorption capacity and
709 selectivity can be a good driver in improving the performance of intensified PCC
710 process. Again studies on mixed solvents in intensified PCC process should be
711 emphasized in order to bring in balance during absorption and desorption so as to
712 have high capture performance in the absorber and less amount of regeneration
713 energy to be consumed in the stripper.

714 7.5 Modelling and simulation of intensified CO₂ capture process

715 Thermodynamic and transport property data is central to any modelling and
716 simulation of intensified CO₂ capture process and these properties were readily
717 available for up to 30 wt% aqueous MEA solution. However higher MEA
718 concentration is required for RPB absorber or stripper due to low residence time.
719 There is very limited data for CO₂ with high concentration MEA in literature.

720 Solubility (i.e. VLE) data for MEA concentration up to 60 wt% and for temperature
721 range of 40 to 120 °C were reported in Aronu *et al.* [183]. Mason and Dodge [184]
722 reported solubility for MEA concentration up to 75 wt%. More solubility data at
723 80wt% or even 100 wt% MEA concentrations is needed for intensified PCC process.
724 Therefore there is a need for more solubility studies for higher solvent
725 concentrations. This can be done by experiment or using molecular simulation
726 software to interpolate or extrapolate.

727 Other areas that require experimental or molecular simulation study are reaction
728 equilibrium constants and kinetic parameters determination for higher solvent
729 concentration with CO₂.

730 Most studies through modelling and simulation focused on intensified absorber with
731 few researches on intensified stripper for PCC. Studies on steady state simulation of
732 intensified absorber were reported in Joel *et al.* [87,89] with main aim on process
733 analysis for design and operation. However, these studies could not check the
734 dynamic behaviour of the absorber or the whole intensified PCC process. Therefore
735 future study on dynamic modelling is necessary in order to meet such challenges. It

736 is believed that if a dynamic model is developed and validated, sensitivity analysis
737 will be done for optimal design, operation and control.

738 7.6 Scale-up

739 Commercial-scale capture on operating power plants has yet to be undertaken,
740 leading to uncertainty regarding scale-up and integration of existing technologies
741 [185]. Harzog [186] reported that the challenge for CCS commercial deployment is
742 to integrate and scale up these components (absorber, heat exchanger and the
743 regenerator). Shi *et al.* [187] and Yang *et al.* [188] use computational fluid dynamic
744 (CFD) to study fluid flow in RPB. But more studies are required for scale-up of RPB
745 columns. To be able to carry out the scale-up study of an intensified PCC process, it
746 is recommended to couple process modelling software with CFD software so as to
747 accurately predict the hydraulic behaviour and the mass transfer behaviour of the
748 RPB.

749 7.7 Evaluation of Technical, Economical and Environmental Performance

750 So far, no detailed and systematic studies were reported on the technical,
751 economical and environmental impact on the use of intensified PCC process
752 compared with conventional PCC process.

753 There are some operational benefits when using RPB in intensified PCC process.
754 The first is its ability to be operated at higher gas and/or liquid flow rates owing to the
755 low tendency of flooding compared to the conventional packed bed [128]. The
756 second benefit of using RPB is its better self-cleaning, avoidance of plugging in the
757 system, and being unaffected by a moderate disturbance in its orientation [189].

758 Joel *et al.* [87] carried out comparative study between conventional absorber and
759 intensified absorber, and found the size reduction factor of about 12 times. Jassim *et al.*
760 [85] reported stripper height reduction factor of 8.4 and stripper diameter
761 reduction factor of 11.3 as compared to conventional packed column. Cheng *et al.*
762 [88] reported a reduction factor for RPB stripper of at least 10 times as compared to
763 conventional stripper. Li *et al.* [190] reported that for PCHE performing the same duty
764 as shell-and-Tube heat exchanger has size reduction factor of 4-6 times. From the
765 analysis above, the size of main components in intensified PCC process can be
766 significantly reduced.

767 Because of much less gas–liquid contact time that occurs in RPB absorber than in a
768 conventional packed bed columns, the selection of solvent with a fast reaction rate
769 with CO₂ is crucial [85,87,89,112,113,120,127,191]. This necessitates the use of
770 higher concentration solvent (such as 55 wt% or 75 wt% MEA reported in
771 [85,87,89]), but this comes with another challenge of corrosion as reported by
772 Barham, *et al.* [192]. This corrosion problem can be managed by the use of (a) more
773 expensive construction material such as stainless steel rather than the commonly
774 used carbon steel; (b) coating with high performance polymer on the surface of
775 stainless steel.

776 In addition to steam consumption for solvent regeneration, electricity will be
777 consumed to drive the intensified absorber and stripper in the intensified PCC
778 process. This means parasitic energy consumption added to carbon capture in
779 intensified PCC process. Cheng *et al.* [88] reported that regeneration energy of RPB
780 stripper is smaller than that of conventional packed column (excluding the energy for
781 rotating the RPB stripper). This is caused by decrease in the amount of vapour lean
782 MEA required from reboiler to RPB due to improved heat transfer zone inside RPB
783 thereby decreasing its reboiler duty. Only Agarwal *et al.* [131] studied electricity
784 consumption to drive the motors in RPB absorber using DEA solvent for carbon
785 capture. Generally the higher the rotating speed, the higher the electricity
786 consumption by the motor. The study in Agarwal *et al.* [131] indicates that electricity
787 consumption is quite low at 900 rpm while the number increased significantly at 1500
788 rpm. More experimental studies are required to quantify the contribution of electricity
789 used by motors to overall energy consumption in intensified PCC process.

790 The authors have performed preliminary technical and economic analysis for
791 intensified PCC process compared with conventional PCC process. The initial
792 prediction is that the capital cost of the whole intensified PCC process can reduce to
793 1/6 (i.e. 16.7%) compared with the same capacity conventional PCC process. The
794 reasons behind this prediction are: (a) the average size reduction is around 12 times;
795 (b) Due to corrosion, stainless steel has to be used instead of carbon steel. Unit
796 material price will roughly double. The initial prediction on energy consumption in
797 capturing unit mass of CO₂ will be similar between the two processes. The reasons
798 behind this prediction are: (a) the steam consumption (or the regeneration energy) in
799 intensified PCC process will be lower due to higher concentration of solvent used; (b)
800 electricity consumed to drive the intensified absorber and stripper in the intensified
801 PCC process is highly related to rotating speed. The second item can only be
802 determined by specific design conditions.

803 In summary, it is necessary to quantify overall costs (capital & operating costs) used
804 for capturing unit mass of CO₂ in intensified and conventional PCC processes based
805 on detailed and accurate process models. Detailed life cycle analysis (LCA) for
806 intensified PCC process should be performed in order to compare with conventional
807 PCC process.

808 **8. Conclusions**

809 The paper presents a critical evaluation of current research status in intensified PCC
810 regarding experimental rigs (including intensified absorber and stripper, intensified
811 heat exchanger), experimental studies worldwide, solvent selection, modelling and
812 simulation. It was found: (a) there is no experimental rig for whole intensified PCC
813 process apart from standalone intensified absorber or stripper. There have been no
814 efforts to intensify the reboiler and the cross heat exchanger so far. (b) There is no
815 systematic study on solvent selection for intensified PCC process. (c) There are

816 some papers on steady state modelling and simulation of intensified absorber. No
817 modelling and simulation of intensified stripper was reported in open literature.
818 Future research efforts and potential breakthrough on different aspects of intensified
819 PCC process have been discussed. These include: (a) A schematic PFD for
820 intensified PCC process has been proposed. (b) It is important to use simple
821 experiments to observe whether interfacial turbulence (i.e. Marangoni effect) exists
822 in RPB absorber. (c) It is vital to develop dynamic models for the whole intensified
823 PCC process for future work in process control. (d) It is necessary to combine CFD
824 study and process modelling for scale-up study. (e) A preliminary technical and
825 economic analysis for the intensified PCC process has been carried out in
826 comparison with conventional PCC process. More detailed and systematic technical,
827 economic and environmental performance analysis should be performed.

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