

1 **Physical solvents and techno-economic analysis for pre-combustion CO₂ capture: A review**

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10
11 **Abstract**

12 This manuscript reviews the use, development, and economic performance of physical solvents
13 for pre-combustion CO₂ capture from high pressure H₂ rich syngas streams. Commercially
14 available physical solvents are presented, followed by an assessment of the ideal properties that
15 are important for development of novel solvents for CO₂ capture from high-pressure syngas
16 streams. To compare the technical and economic performance of traditional and novel physical
17 solvents, a review of the methods, assumptions and models used in techno-economic analysis
18 (TEA) studies was conducted. It was found that, although some novel solvents show promising
19 technical performance in the laboratory (e.g., high CO₂ absorption capacity and low vapor
20 pressure), other issues (e.g., solvent viscosity and cost) may limit their industrial applications.
21 Process simulations were useful tools for modeling the technical performance of processes using
22 traditional and novel solvents. However, model predictions are most reliable when the methods
23 and correlations used to develop the process simulation are validated with representative
24 experimental data, in particular highly accurate baseline models are required for fair comparison
25 among physical solvents. The key inputs and assumptions in pre-combustion CO₂ capture TEAs
26 have also been summarized. Some studies showed that the promising technical performance of
27 novel physical solvents can be offset by the high and often unknown costs of these solvents. Future
28 development of novel physical solvents for pre-combustion CO₂ capture will benefit from more
29 studies that conduct in-depth techno-economic analysis, specifically with validated process
30 simulations and transparent economic models.

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32 **Key words:** solvent absorption, pre-combustion, blue hydrogen, carbon dioxide, physical solvent,
33 techno-economic analysis, review

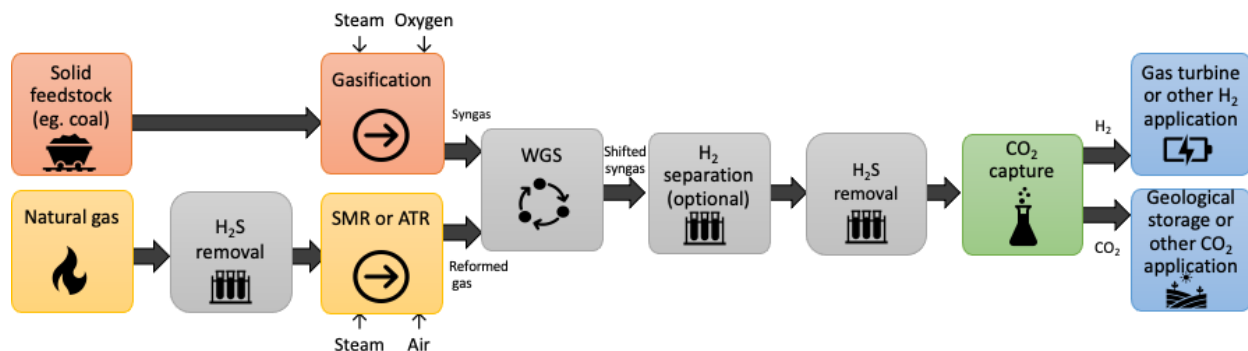
34 **Highlights:**

- 35 • A review of both commercial and novel physical solvents for pre-combustion CO₂ capture
- 36 • Review of techno-economic analysis (TEA) studies using physical solvents
- 37 • Key assumptions of the TEA studies have been summarized
- 38 • Lists areas of improvement for future TEAs for physical solvents

39 **1. Introduction**

40 As reported in the latest IPCC assessment report, in order to limit global warming, “strong, rapid,
41 and sustained reductions in carbon dioxide, methane and other greenhouse gases are necessary”
42 (IPCC, 2021). The primary sources of greenhouse gas (GHG) emissions in the United States in
43 2019 were transportation (29%), electricity production from fossil fuels (25%) and industries that
44 burn fossil fuels for energy or produce GHG emissions from chemical processes (23%) (EPA,
45 2021). Carbon capture and storage (CCS) technologies are considered critical for reducing fossil
46 fuel sourced carbon dioxide (CO₂) emissions in order to meet global climate targets (Global CCS
47 Institute, 2020). In the first step of CCS, CO₂ emissions are captured using a gas separation process,
48 such as solvent absorption, membrane separation, adsorption or cryogenic separation, and the
49 resulting CO₂ rich gas stream is then compressed, transported, and stored geologically in depleted
50 oil and gas fields or deep saline formations.

51 Many forms of CO₂ capture will be required to achieve global climate goals, including but not
52 limited to: (a) direct air capture, (b) post-combustion capture from large power plants and similar
53 point sources, and (c) pre-combustion capture from mid-stream facilities, gasification plants and
54 chemical refineries. The demand for clean or near-zero emissions hydrogen produced from fossil
55 fuels with CCS, also known as clean hydrogen or ‘blue hydrogen’, is increasing due to its potential
56 to make a significant contribution to emissions reduction in the power generation, transportation,
57 and industrial sectors (Global CCS Institute, 2021). Blue hydrogen is produced from fossil fuels
58 (e.g. steam methane reforming (SMR) from natural gas or gasification from a solid fuel like coal)
59 with carbon capture and storage (Lau et al., 2021; NETL, 2021). As such, this review will be
60 important for future implementation of blue hydrogen technologies. Figure 1 provides a high-level
61 overview of the main processes involved in the production of blue hydrogen.



62
 63 **Figure 1: Block flow diagram of clean or blue hydrogen production processes with carbon**
 64 **capture and storage (CCS) including (1) coal gasification and (2) natural gas reforming**
 65 **(Steam Methane Reforming, SMR or Autothermal Reforming, ATR) with water gas shift**
 66 **(WGS), optional H₂ membrane separation and acid gas removal (AGR) or CO₂ capture**

67 In this literature review, we focus only on physical solvents for pre-combustion CO₂ capture.
 68 Though, the physical solvents analyzed here are relevant to CO₂ capture from a wide range of high
 69 pressure syngas streams containing H₂ and CO₂. Furthermore, we focus mainly on literature in this
 70 field which attempts to estimate the total cost of pre-combustion CO₂ capture using physical
 71 solvents. Additionally, here we only focus on the capture of CO₂ at the site making the shifted
 72 syngas, though it's important to note that achieving net-zero emissions from blue hydrogen
 73 production will require upgrades to associated upstream/midstream pipelines, mining and
 74 processing centers in order to prevent the emission of CO₂ and other GHGs, such as methane. The
 75 GHG emission association with pipelines, mining and processing centers, as well as the needed
 76 technologies to limit upstream/midstream GHGs is outside the scope of this literature review.

77 In this literature review, we focus on pre-combustion CO₂ capture from shifted syngas streams,
 78 which typically implies capture of CO₂ (and/or other acid gases, such as H₂S and COS) from fuel
 79 gas streams containing high concentrations of CO₂ and/or at high pressure (i.e. high CO₂ partial
 80 pressure). Some examples of industrial processes producing gas streams with high CO₂ partial
 81 pressure include: petrochemical processes, synthetic liquid fuels, ammonia, steam methane
 82 reforming for hydrogen production, and Integrated Gasification Combined Cycle with CO₂ capture
 83 and storage (IGCC-CCS) power or chemical plants. A full review of precombustion capture
 84 technologies, such as solvents, sorbents, and membranes, can be found in Jansen *et al.* (2015).

85 In IGCC power generation, a solid feedstock (coal, biomass, and/or solid waste) is partially
 86 oxidized in a gasifier using steam and oxygen/air to produce a synthesis gas (syngas). This syngas,
 87 which is at high temperature and high pressure, is a mixture of fuels including hydrogen (H₂),

88 carbon monoxide (CO) and methane (CH₄) as well as other gaseous species including CO₂. To
89 apply CO₂ capture, a water-gas shift reactor (WGSR) is used to convert CO and water (H₂O) in
90 the syngas into additional H₂ and CO₂, resulting in a high pressure shifted syngas mixture that can
91 contain CO₂ at concentrations up to 45% (McJannett, 2012). The shifted syngas is then cleaned
92 (particulates, tars, and H₂S are typically removed) before being sent to a gas turbine to generate
93 power. In an IGCC power plant with CCS, a CO₂ removal process would be added after the H₂S
94 acid gas removal (AGR) process (or possibly combined with the H₂S AGR process), to produce a
95 high CO₂ concentration, high-pressure gas stream. This CO₂ rich gas stream can then be sent to a
96 pipeline for geological storage or for utilization in another process, such as enhanced oil and gas
97 recovery, provided that produced oil and gas does not contribute to further GHG emissions.

98 **2. Physical solvent absorption processes for CO₂ capture**

99 One of the most frequently reported pre-combustion CO₂ capture technology is physical solvent
100 absorption. When using physical solvents for CO₂ capture, there is no chemical reaction so the
101 driving force for CO₂ absorption is the solubility of CO₂ in the solvent which generally increases
102 with pressure and decreases with temperature in accordance with Henry's law. Physical solvents
103 are most commonly regenerated via some combination of: (a) pressure reduction, (b) temperature
104 increase, and/or (c) sweep gas. The captured CO₂ is typically released to the atmosphere unless
105 there is nearby CO₂ compression and storage equipment, such as the Great Plains Synfuels Plant
106 in North Dakota which captures H₂S and CO₂ using Rectisol[®] solvent and then compresses and
107 transports the H₂S and CO₂ to sour oil fields in Saskatchewan, Canada (NETL, 2020b). In the
108 future, the practice of venting CO₂ (and CH₄) from natural gas processing centers (which in 2019
109 accounted for approximately 0.3% of total US emissions of CO₂ equivalent emissions (EPA,
110 2021)) will need to be adjusted in order to capture the released GHGs. The techno-economic
111 analyses discussed in this report will be relevant to existing natural gas processing centers, in
112 addition to future steam methane reforming and gasification facilities that also generate blue
113 hydrogen. The techno-economic analysis summarized here covers both baseline cases as well as
114 advanced processes to lower the cost of pre-combustion capture using physical solvents that (a)
115 modify the regeneration conditions to recover the CO₂ at a higher pressure, (b) improve selectivity
116 to reduce H₂ losses, and (c) develop a solvent that has a high CO₂ loading at a higher temperature
117 in order to integrate waste heat into the process.

118 **2.1 Current commercial physical solvent processes**

119 There are potentially many physical solvents for AGR processes, but the most commonly used and
120 proven commercial physical solvent processes are Selexol™ (Honeywell UOP, Des Plaines, IL,
121 USA) and Rectisol® (Air Liquide, Houston, TX, USA). Both processes provide good CO₂
122 absorption capacity and selectivity but are relatively energy intensive due to their operation below
123 room temperature.

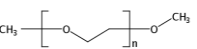
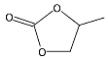

124 The Selexol process, which uses a solvent containing a mixture of dimethyl ethers of polyethylene
125 glycol (DEPG or PEGDME), has several benefits including high solvent stability and low vapor
126 pressure (compared with some solvents, though high vapor pressure compared with ionic liquids)
127 (Porter et al., 2017). DEPG can be used for selective H₂S removal as well as bulk CO₂ removal in
128 a two-stage process with two absorption columns and multiple regeneration flash tanks. DEPG
129 will dehydrate the gas, which is ideal for natural gas processing facilities, but likely not ideal for
130 SMR and IGCC applications. At low temperatures DEPG has a relatively high viscosity which
131 reduces mass transfer efficiency, (Burr and Lyddon, 2008), which must then be balanced with the
132 increased CO₂ capacity at low temperatures (typical operating temperature is around 10 °C for
133 Selexol). The Selexol solvent process has been used to remove CO₂ from natural gas streams at a
134 range of processing facilities including the Century Plant (Texas, USA), Terrell Natural Gas plant
135 (Texas, USA), Lost Cabin Gas plant (Wyoming, USA) and Shute Creek Gas processing facility
136 (Mumford et al., 2015). Selexol is also used in the Coffeyville Gasification plant (Kansas, USA)
137 to remove CO₂ during fertilizer production (Folger, 2013). All of these Selexol processes utilize
138 the CO₂ for downstream enhanced oil recovery (EOR) applications. In addition, Selexol was the
139 solvent process chosen for the Kemper County IGCC-CCS power plant (Mississippi, USA.)

140 The Rectisol process, which uses an inexpensive and readily available organic solvent (methanol),
141 is one of the oldest and well-known processes for achieving very high purity syngas streams and
142 CO₂ product streams. For example, it has a very high selectivity for H₂S over CO₂ and can also
143 remove COS. Methanol requires operation at very low temperatures (typically -15 to -60°C)
144 (Jansen et al., 2015) to achieve increased capacity for H₂S and CO₂ at an acceptable viscosity.
145 Methanol also has a high vapor pressure resulting in the need for costly solvent recovery processes
146 to prevent high solvent loss (Burr and Lyddon, 2008) and hence, the operation at cryogenic
147 temperatures. The Rectisol process has been used commercially for a range of processes including
148 CO₂ removal at the Great Plains Synfuels Plant (Saskatchewan, Canada) and at the Alberta Carbon
149 Trunk Line (Alberta, Canada) with the CO₂ being used for EOR in both locations (Folger, 2013;

150 Heal and Kemp, 2013; Loria and Bright, 2021). The Rectisol process has also been used
 151 extensively in Sasol’s synfuel plants in South Africa (Gatti et al., 2014).

152 Two other relatively common commercial physical solvents are: (a) propylene carbonate or PC
 153 (Fluor process), which provides similar CO₂ capacity but at low temperatures, so it is necessary to
 154 cool the syngas before CO₂ capture; and (b) N-Methyl-2-Pyrrolidone (NMP), which is a physical
 155 solvent used in the commercial process known as Purisol and is known for its high selectivity for
 156 H₂S over CO₂ regardless of the operating temperature (typically 15 to 40 °C). However, NMP also
 157 has a relatively high vapor pressure leading to the requirement of water washes of the gas streams
 158 to enable recovery of the solvent (Burr and Lyddon, 2008). A summary of the physical properties
 159 for these commercially available CO₂ capture physical solvents can be found in Table 1. For
 160 properties such as molecular weight, boiling point temperature, and viscosity, PC and NMP have
 161 values between DEPG and methanol. However, as will be shown in Table 2, most of the not-yet
 162 commercial, hydrophobic solvents being considered have values of these properties even higher
 163 than DEPG.

164 **Table 1. Summary of physical properties for commercially available hydrophilic physical**
 165 **solvents for CO₂ capture (Bhattacharyya et al., 2017; Burr and Lyddon, 2008; Kohl and**
 166 **Nielsen, 1997b; Mumford et al., 2015; Theo et al., 2016)**

Solvent	Dimethyl ethers of polyethylene glycol (DEPG)	Methanol	Propylene carbonate (PC)	N-Methyl-2-pyrrolidone (NMP)
Process name	Selexol	Rectisol	Fluor	Purisol
Molecular formula	CH ₃ O(C ₂ H ₄ O) _n CH ₃ where n has a specific distribution	CH ₄ O	C ₄ H ₆ O ₃	C ₅ H ₉ NO
Chemical structure		CH ₃ —OH		
Molecular Weight	280	32	102	99
Viscosity @ 25 °C (cP) (dry)	5.8	0.6	3	1.7
Density @ 25 °C (kg/m ³)	1030	785	1195	1027
Vapor pressure @ 25 °C (Pa)	0.097	13,000	11	53
Melting point (°C)	- 28	- 92	- 48	- 24
Boiling point at 1 atm (°C)	275	65	240	202
Thermal conductivity @ 25°C (W/m.K)	0.19	0.21	0.21	0.16

Specific heat @ 25°C (J/g.K)	2.05	0.327	1.42	1.67
Operating temperature (°C)	- 7 to 40	- 70 to 0	- 18 to 65	- 15 to ambient
CO ₂ Henry's Law Constant (dry) @ 25°C (mol/(MPa.L))	1.6	1.4 (3.6 @ - 10 °C. 6.7 @ - 30 °C)	1.5	1.6
H ₂ S solubility/CO ₂ solubility @ 25°C	8.9	7.1 @ - 25 °C	3.3	10.2
CH ₄ solubility/CO ₂ solubility @ 25°C	0.067	0.051 @ - 25 °C	0.038	0.072
H ₂ solubility/CO ₂ solubility @ 25°C	0.013	0.0054 @ - 25 °C	0.0078	0.0064

167

168 These commercial absorption processes all use hydrophilic solvents that are well suited to treating
169 raw natural gas streams at ambient temperatures where both acid gases (e.g. CO₂ and H₂S) and
170 water vapor need to be removed prior to injection into a pipeline with specification on (a) energy
171 density, (b) corrosion, and (c) water dew point. However, what makes these hydrophilic solvents
172 ideal for natural-gas-sweetening applications can cause difficulties for the IGCC-CCS application.
173 For example, in pre-combustion CO₂ capture, the shifted syngas enters the AGR process at a much
174 higher temperature, which means significant gas cooling is required before using a CO₂ absorption
175 process with the commercially available hydrophilic solvents operating at below room
176 temperature. Development of novel physical solvents that can selectively absorb CO₂ at higher
177 temperatures than the current commercial solvents will result in energy and cost advantages. In
178 addition, for natural-gas-sweetening applications, the commercially available physical solvents are
179 typically regenerated by pressure reduction and the CO₂ is just vented, though there are some cases
180 where the CO₂ is collected, compressed and sold as a pressurized product to industry. If the solvent
181 regeneration process can also occur at higher pressures using waste or low-grade heat, further
182 energy and cost advantages could result for pre-combustion carbon capture, (Dave et al., 2020)
183 which will be crucial for reducing the CO₂ emissions from mid-stream AGR NG processing
184 facilities.

185 **2.2 Novel physical solvents for CO₂ capture**

186 For high pressure IGCC-CCS applications, there are processing and economic benefits to operating
187 physical solvent absorption and regeneration processes at higher temperatures and pressures. As
188 such, a number of researchers have been developing novel physical solvents that can absorb CO₂
189 at higher temperatures from high pressure shifted syngas streams containing CO₂, H₂O and H₂

190 (Heintz et al., 2008; Miller et al., 2010; Shi et al., 2016; Shi et al., 2015; Siefert et al., 2016a; Tang
191 et al., 2016). An ideal physical solvent for IGCC-CCS would have the following properties:

- 192 • High CO₂ and H₂S capacity (reduces solvent flow rate and equipment sizes)
- 193 • Favorable selectivity for CO₂ over other fuel gas components, including CH₄, H₂, N₂, and
194 CO
- 195 • Low vapor pressure (reduces solvent losses and allows operation at higher temperature)
- 196 • Non-foaming
- 197 • Low water uptake
- 198 • High CO₂ diffusivity
- 199 • High thermal stability (enables operation above ambient temperature)
- 200 • Low cost
- 201 • Low toxicity
- 202 • Simple solvent synthesis
- 203 • Environmentally benign

204 Numerous physical solvents, with many of these properties, have been proposed in the literature
205 for removing CO₂ from high pressure gas streams. For example, the Estasolvan process, which
206 utilizes tri-n-butyl phosphate (TBP) as the solvent, has shown potential as a physical solvent under
207 pilot plant conditions, but has not been implemented commercially (Carranza-Abaid et al., 2021;
208 Franckwiak and Nitschke, 1970). Another recent study has compared the CO₂ absorption
209 performance of TBP and three analogous organophosphate solvents in which the length of the
210 PEG-substitution was varied (Thompson et al., 2019a). Although acceptable densities and
211 viscosities were obtained, CO₂ absorption performance was reduced, and increased water
212 absorption was observed.

213 Several small volatile compounds such as methanol and acetone (Miller et al., 2011) and
214 hydrophobic polymers such as polydimethyl siloxane (PDMS) (Shi et al., 2015), poly(propylene
215 glycol) dimethylether (PPGDME) (Enick et al., 2013) and propylene carbonate mixtures with
216 poly(propylene glycol) monobutylether (PPGME340) (Li et al., 2020) have been evaluated as
217 physical solvents for CO₂ absorption. PDMS has the advantage of not absorbing any water,
218 however its relatively low selectivity for CO₂ over H₂ can be problematic (Enick et al., 2013; Shi
219 et al., 2016). Some solvents, like PPGDME, were found to gel in the presence of water, which will

220 likely limit industrial application (Enick et al., 2013). Water is also known to compete with CO₂
221 for interaction with these solvents leading to lower CO₂ loading and reduced CO₂/H₂ selectivity
222 (Koronaos et al., 2016). Generally, hydrophobic solvents do not absorb water, which improves
223 CO₂ absorption performance and prevents energy intensive gas cooling requirements.

224 The promising features of these hydrophobic solvents has led to the search for other hydrophobic
225 solvents with increased CO₂ solubility as well as improved CO₂/H₂ selectivity. One study
226 investigated polyethylene glycol siloxane solvent (referred to as PEG-Siloxane-1 or PEG-PDMS-
227 1), which is an amphiphilic solvent with polar ethylene glycol segments and a non-polar siloxane,
228 which provides favorable hydrophobicity that is characteristic of PDMS combined with improved
229 CO₂ solubility and CO₂/H₂ selectivity that is typical for polyethylene glycol (PEG) (Shi et al.,
230 2016; Siefert et al., 2016a). However PEG-PDMS-1 was reported to have foaming issues which
231 led to the development of PEG-PDMS-3 (or disub-4PEG), which avoided foaming issues while
232 also providing high CO₂/H₂ selectivity at above room temperature conditions (CO₂/H₂ selectivity
233 of ~70 at 25 °C and ~60 at 40 °C). Operation at these higher temperatures will also allow for waste
234 heat utilization for solvent regeneration (Siefert and Hopkinson, 2018). In a related study, a solvent
235 named CASSH-1 showed promising potential as a hydrophobic physical solvent with absorption
236 capacity and selectivity predicted to be comparable to Selexol and PEG-PDMS-3 but with no
237 foaming issues and very low vapor pressure (Shi et al., 2018). CASSH-1's favorable absorption
238 and physical properties were also highlighted in a related computational screening study by Shi et
239 al. (2021) where physical solvents for pre-combustion CO₂ capture were screened via a model
240 developed by integrating the commercial NIST database, an in-house NETL computational
241 database, chem-informatics, and molecular modeling. In another development, nine new
242 hydrophobic PEG-substituted solvents were found to be good candidates for pre-combustion CO₂
243 capture applications because of their “hydrophobicity, high CO₂ solubility, low evaporation rate,
244 and lack of foaming” (Thompson et al., 2019b).

245 Many other studies have proposed the use of ionic liquids (ILs) as novel physical solvents for CO₂
246 capture (Basha et al., 2014; Basha et al., 2013; García-Gutiérrez et al., 2016; Heintz et al., 2009;
247 Jamali et al., 2017; Liu et al., 2021; Taheri et al., 2018; Wang et al., 2021; Zhai and Rubin, 2018;
248 Zhao et al., 2020; Zhao et al., 2018). ILs allow a range of cation/anion combinations, which can
249 enable the solvent to be customized to different applications. ILs have very low to negligible vapor
250 pressure (which prevents solvent loss to the atmosphere), low corrosivity (which allows cheaper

251 construction with carbon steel), low water miscibility and high thermal stability. ILs typically
252 operate at higher temperatures, which reduces the gas cooling requirements and may allow the
253 solvent to be regenerated with waste heat at higher pressure. A large number of imidazole and
254 imidazolium-based ionic liquid species have been reported as physical solvents for CO₂
255 absorption, including but not limited to, 1-butyl-3-methylimidazolium hexafluorophosphate
256 ([bmim][PF₆]), 1-butyl-3-methylimidazolium tetrafluoroborate ([bmim][BF₄]), 1-hexyl-3-
257 methylimidazolium bis(trifluoro-methylsulfonyl)amide ([hmim][Tf₂N]), 1-butyl-3-
258 methylimidazolium bis(trifluoromethylsulfonyl)imide ([bmim][Tf₂N]) and 1-butyl-3-
259 methylimidazolium bromide ([bmim][Br]) (Basha et al., 2014; Basha et al., 2013; Jalili et al., 2010;
260 Jamali et al., 2017; Safavi et al., 2013; Taheri et al., 2018; Zhai and Rubin, 2018). A hydrophobic
261 ionic liquid called allyl-pyridinium bis(trifluoromethylsulfonyl)imide or [aPy][Tf₂N] was
262 synthesized and found to have similar CO₂ solubility to Selexol but significantly lower vapor
263 pressure and relatively low viscosity (~28 cP at 25 °C) for an ionic liquid (Siefert et al., 2016a).
264 Another study investigated the use of a PEG-ionic liquid solvent blend to intensify the CO₂
265 absorption capacity while reducing the viscosity (Li et al., 2016). In a study by Song et al. (2020),
266 a dataset containing 10,116 CO₂ solubilities in ionic liquids was collected and used to develop two
267 machine learning models for predicting CO₂ solubility. The relationship between CO₂ solubility
268 and IL structure, temperature and pressure were correlated using group contribution methods.
269 Additional studies by these authors found computer-aided molecular design (CAMD) could be
270 used to find optimal ILs by using solubility and viscosity as design objectives (Zhang et al., 2021;
271 Zhou et al., 2021). Finally, a new generation of solvents named deep eutectic solvents (DESs) have
272 shown potential for use in carbon capture applications. DESs share some of the promising features
273 of ILs including low melting point and very low volatility, but they are also non-flammable, non-
274 toxic and easily manufactured (Liu et al., 2021; Sarmad et al., 2017; Wibowo et al., 2021; Xin et
275 al., 2020).

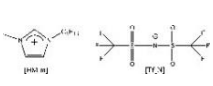
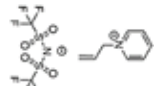
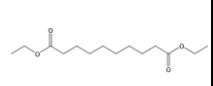

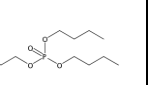
276 Despite the promising technical performance shown by many of these studies, the high cost of
277 synthesizing the new IL, which is often unknown at commercial scale, is likely to limit further
278 application. Further analysis on the cost and scale-up of IL processes is required before application
279 in industrial CO₂ capture processes. A review of studies covering economic analysis of novel
280 physical solvents including ILs can be found in Section 3. Moreover, although ILs can provide
281 enhanced CO₂ solubility, the transport properties (i.e. viscosity) and thermochemical properties

282 (i.e. heat capacity) may preclude their use at process scale due to increased operating costs as a
283 result of these important properties (Mota-Martinez et al., 2018). Physical absorption of CO₂ by
284 ILs in commercial packed columns was also reported to be strongly kinetically controlled (i.e.
285 mass transfer is dependent on solvent properties including viscosity, surface tension and CO₂
286 diffusivity) and concluded that viscosity was the key parameter when selecting a physical solvent
287 (de Riva et al., 2017; Palomar et al., 2019).

288 A summary of the physical properties for some selected novel physical solvents for CO₂ capture
289 can be found in Table 2.

290

291 **Table 2. Summary of physical properties for some selected novel hydrophobic physical**
 292 **solvents proposed for CO₂ capture (Basha et al., 2013; Enick et al., 2013; Shi et al., 2016;**
 293 **Shi et al., 2015; Shi et al., 2010; Shi et al., 2019; Siefert and Hopkinson, 2018; Siefert et al.,**
 294 **2016a)**

Solvent	[hmim][Tf ₂ N]	[aPy][Tf ₂ N]	Diethyl sebacate (CASSH-1)	PEG-PDMS-3 (or disub-4PEG)	Tributyl phosphate (TBP)
Solvent type	Ionic liquid	Ionic liquid	Hydrophobic solvent	Hydrophobic solvent	Hydrophobic solvent
Molecular formula	C ₁₂ H ₁₉ N ₃ F ₆ O ₄ S ₂	C ₁₀ H ₁₀ N ₂ S ₂ O ₄ F ₆	C ₁₄ H ₂₆ O ₄	C ₂₆ H ₆₀ O ₁₀ Si ₃	C ₁₂ H ₂₇ O ₄ P
Chemical structure					
Molecular Weight kg/kmol	447	399	258	620	266
Viscosity @ 25°C (cP)	71	28	5.1	12.2	2.9
Density @ 25 °C (kg/m ³)	1371	1507	960	987	979
Vapor pressure @ 25 °C (Pa)	small	<0.09	0.0012	small	0.15
Melting point (°C)	- 3 ± - 2	-	5	-	- 80
Boiling point (°C)	635	-	312	high	289
Thermal conductivity @ 25°C (W/m·K)	0.12	0.23	0.15	0.17	-
Specific heat @ 25°C (J/g·K)	1.41	1.11	-	-	1.426
CO ₂ Henry's Law constant	~0.8	~1.1	~1.3	~1.2	1.1 *

@ 25°C (mol/(MPa.L))					
CO ₂ /H ₂ selectivity @ 25 °C	~30	~100	66	58	61 *

295 *prediction from simulation Shi et al. (2018), - data not found

296
 297 Other physical solvents that have been proposed for pre-combustion CO₂ capture include
 298 carbonates (dimethyl carbonate, diethyl carbonate and diethyl succinate) (Gui et al., 2010; Zhao et
 299 al., 2017), hexamethyl phosphorotriamide (Blanc et al., 1979), perfluorinated compounds (Heintz
 300 et al., 2008), N-formyl morpholine (Jou et al., 1989), polypropyleneglycol dimethylether and
 301 polydimethyl siloxane (Miller et al., 2010). Also, solvents with ether and ester groups (methyl
 302 heptanoate, ethyl hexanoate, methyl benzoate, ethylene glycol methyl ether acetate, propylene
 303 glycol methyl ether acetate, 3-methoxy butyl acetate, diethylene glycol diethyl ether, ethylene
 304 glycol butyl ether acetate and carbitol acetate) (Gui et al., 2014; Li et al., 2018a; Li et al., 2018b)
 305 and 1,2,3-Trimethoxypropane (Flowers et al., 2017). Common solvents including ethanol, decane
 306 and acetone have also been reported as physical solvents for CO₂ capture (Chang et al., 1997; Kohl
 307 and Nielsen, 1997a; Miller et al., 2011; Shi et al., 2019). A number of reviews have been published
 308 on solvent selection for CO₂ capture, including some discussion on the research and development
 309 of physical solvents (Ban et al., 2014; N.Borhani and Wang, 2019; Shi et al., 2019; Theo et al.,
 310 2016; Vega et al., 2018).

311 **2.3 Outlook for development of new physical solvents for CO₂ capture**

312 Significant research effort is currently underway to develop novel higher performing physical
 313 solvents for pre-combustion CO₂ capture. However, commercially proven hydrophilic physical
 314 solvents, like the Selexol process, would continue to be the industry standard for IGCC-CCS until
 315 the higher performing and potentially less costly solvent processes are demonstrated at scale. Most
 316 of the recent studies published on both commercial and novel physical solvents for pre-combustion
 317 CO₂ capture have been limited to bench scale experimental data (e.g. measurement of physical
 318 properties, CO₂ solubility and CO₂ selectivity) (Gui et al., 2010; Gui et al., 2014; Jalili et al., 2015;
 319 Jamali et al., 2017; Jou et al., 1989; Miller et al., 2011; Miller et al., 2010; Rayer et al., 2012a, b;
 320 Zhao et al., 2017; Zhao et al., 2018) or process simulations (Ashkanani et al., 2020, 2021; Basha
 321 et al., 2013; Dave et al., 2016; Field and Brasington, 2011; Gatti et al., 2014; Kapetaki et al., 2015;

322 Shi et al., 2015; Sun and Smith, 2013; Taheri et al., 2018; Zhao et al., 2018). Fewer studies
323 (Ashkanani et al., 2020, 2021; Field and Brasington, 2011; García-Gutiérrez et al., 2016; Siefert
324 et al., 2016a; Zhai and Rubin, 2018) have assessed both the technical and economic performance
325 of physical solvent absorption processes. As reported by Mota-Martinez et al. (2018), it is
326 important to conduct a systemic assessment of both the non-monetized properties as well as
327 monetized properties (e.g. CAPEX, OPEX) when considering a new solvent process for CO₂
328 capture. In the literature, most of the research effort, particularly with ILs, is focused on improving
329 CO₂ solubility or solvent capacity, however, transport properties (like viscosity) and
330 thermochemical properties (like heat capacity) have been shown to have a significant impact on
331 the economic performance of a CO₂ capture process, which could prevent practical implementation
332 of these solvents (Mota-Martinez et al., 2018). Other solvent properties like solvent foaming,
333 which generally cannot be predicted using process simulation tools, can also challenge industrial
334 implementation of novel solvents. A hypothetical modeling study by Xin et al. (2020) highlighted
335 the importance of using a solvent with high CO₂ solubility and also found solvent viscosity is
336 important in some mass transfer correlations used in rate-based models. Therefore, although
337 research into novel high performing solvents is important and warranted, this research must be
338 coupled with a robust assessment of the whole process (such as a techno-economic analysis) in
339 order to understand the overall cost potential of CO₂ capture with these novel solvents.

340 **3. Techno-economic analysis (TEA) of physical solvents for CO₂ capture**

341 Reducing the cost of capture is one of the biggest challenges for deployment of CCS technology.
342 Process innovations using better solvents are expected to reduce the capital and energy costs
343 associated with CO₂ capture. For CO₂ capture with physical solvents, the economic cost is known
344 to depend on a range of solvent properties (Siefert et al., 2016a) and therefore completion of TEA
345 will encourage commercial implementation of novel processes. A TEA will initially assess the
346 technical performance of the process, to obtain equipment sizes and energy demands based on
347 specified design targets (e.g. CO₂ capture rate, product pressure, etc.) using process simulation
348 software like Aspen Plus. These technical parameters are then used in an economic analysis to
349 determine the capital and operating costs of the process.

350 NETL has developed a comprehensive set of publicly available “Baseline Studies for Fossil
351 Energy Plants” to provide “estimates for the cost and performance of combustion- and gasification-
352 based power plants, as well as options for co-generating synthetic natural gas and fuels - all with

353 and without carbon dioxide capture and storage - for several ranks of coal” (James et al., 2019;
354 NETL, 2020a). These studies provide an important resource for comparing high-level relative
355 costs of energy generation due to their evaluation of different cases using a common methodology
356 and consistent set of technical and economic assumptions.

357 The TEA studies discussed in this review focus on the CO₂ capture process using physical solvents
358 and highlight the importance of assessing the technical performance of each process alongside
359 economic performance in order to understand what parameters will minimize the cost of CO₂
360 capture. While completing this review, it was found that, when comparing TEA results from
361 different research groups, it is important to consider what methods, assumptions, and models that
362 have been used in their TEA since these can have a significant impact on the overall outcomes.
363 For this reason, the main goal of this literature review is to analyze and compare the methods and
364 **input** assumptions that have gone into previous TEAs for physical solvents rather than to directly
365 compare the **output** results, such as levelized cost of cost (LCOC: \$/ton CO₂) or levelized cost of
366 hydrogen (LCOH: \$/kg H₂). The following sections aim to provide an overview of some of the
367 process and economic models, along with their inputs and assumptions, which were used to assess
368 the technical and economic performance of physical solvents for CO₂ capture.

369 **3.1 Process simulations used in TEAs to assess the technical performance of physical solvents** 370 **for CO₂ capture**

371 When developing a process simulation for a physical solvent absorption process, it is important to
372 consider the models chosen for predicting the VLE (vapor liquid equilibrium) and the mass transfer
373 behavior. In addition, models for predicting properties such as density, viscosity, surface tension,
374 vapor pressure, heat capacity, thermal conductivity are also important as they can directly or
375 indirectly impact the VLE and mass transfer models (Bhattacharyya et al., 2017). Several equation
376 of state (EOS) models, including SRK-EOS, PC-SAFT-EOS and PR-EOS, have been reported to
377 be suitable for predicting the VLE behavior in physical solvent absorption processes
378 (Bhattacharyya et al., 2017). Recently, a cheminformatics-based machine learning model has been
379 developed to predict CO₂ solubility in physical solvents using “organic structural and bond
380 information, thermodynamic properties, and experimental conditions” as input variables (Li et al.,
381 2019). The mass transfer performance of a physical solvent is also important to understand,
382 particularly when the viscosity of the solvent is relatively high, such as with Selexol and some of
383 the novel IL solvents discussed above. Overall, limited studies are available in the literature for

384 the mass transfer performance of physical solvents (Heintz, 2012). Generally, the gas phase
385 resistance is considered to be negligible if the partial pressure of the physical solvent in the gas
386 phase is low. Therefore, the mass transfer resistance can be assumed to be only from the liquid-
387 phase, as represented by the volumetric liquid phase mass transfer coefficient, $k_L a$, which has been
388 reported to vary with pressure and temperature. Xin et al. (2020) developed a rate-based model for
389 the Selexol process using different mass transfer correlations, including Bravo et al. (1985), Billet
390 and Schultes (1993), and Hanley and Chen (2012), and compared the results to the equilibrium
391 modeling approach. Solvent viscosity was found to strongly affect the mass transfer rate using the
392 Hanley and Chen (2012) correlation, but solvent viscosity only had a minor impact on the mass
393 transfer rate using the other two correlations. This study highlights that although there are
394 numerous correlations for predicting important properties like mass transfer coefficient (k_L) and
395 interfacial area (a) in the literature and built into process simulations, they should be selected with
396 caution.

397 A number of studies have reported simulations that propose process innovations for improving the
398 efficiency and cost of CO₂ capture using physical solvents within the IGCC process. However,
399 comparing their technical performance can be challenging due to the range of modeling
400 assumptions and lack of transparency in the models used to predict process performance. With this
401 in mind, Field and Brasington (2011) presented a baseline model for an IGCC power plant with
402 carbon capture using Selexol based physical solvent absorption “to enable researchers to have a
403 consistent and transparent framework for analyzing IGCC flowsheets and potential innovations”.
404 The Aspen Plus model, which is freely available online, includes all major pieces of equipment at
405 an IGCC-CCS power plant. For the AGR, a single DEPG component was chosen to represent the
406 DEPG mixture (Selexol) and PC-SAFT parameters were used to predict equilibrium. However,
407 over the course of completing this literature review, we compared the publicly available IGCC-
408 CCS model developed by Field & Brasington and found important differences between model
409 predictions and experimental laboratory-scale studies that also used DEPG or Selexol solvent
410 substitutes (Chen et al., 2020; Gainar and Anitescu, 1995; Li et al., 2007). While this baseline
411 model provides a useful framework for developing simulations in AspenPlus, we want to highlight
412 some considerations including (1) the CO₂ uptake at a given temperature and partial pressure of
413 CO₂ may be lower than experimental values (Chen et al., 2020); (2) the predicted dependence of
414 H₂ uptake with temperature is opposite to that obtained with experimental studies (Chen et al.,

415 2020; Gainar and Anitescu, 1995) and (3) the effect of adding water to DEPG on the viscosity is
416 the opposite compared with experimental studies (Li et al., 2007). While this freely available
417 simulation is a useful first step and provides a lot of detail for modeling the overall IGCC-CCS
418 power plant, a new IGCC-CCS baseline model is needed which more accurately mimics the
419 thermodynamics of physical solvents including the water/DEPG mixtures in contact with the
420 syngas species, such as the experimentally-measured increase in density and viscosity when water
421 is added (as shown in Figure 1 and 2 of Li et al. (2007)).

422 While only focusing on the CO₂ capture and compression portions of an IGCC-CCS power plant,
423 Siefert et al. (2016a) modified the previously mentioned Aspen Plus simulation and regressed in
424 experimental gas uptake values for the AGR process presented by Field and Brasington in order
425 to compare the performance of the novel hydrophobic solvent PEG-Siloxane-1 and the ionic liquid
426 [aPy][Tf₂N]. The absorber was modeled as an adiabatic Rate Based Rad-Frac block in Aspen Plus
427 with physical properties predicted using the PC-SAFT equation of state (EOS) for Selexol and
428 PEG-Siloxane-1 while the ENRTL-RK method was used for the ionic liquid. The resulting process
429 flowsheets from these simulations were then used to complete a techno-economic analysis for each
430 physical solvent.

431 Dave et al. (2016) assessed the performance of DMEPEG (Selexol) solvent using a rate-based
432 mass transfer model with ProTreat[®] simulation software. A non-ionic liquid package was used to
433 predict the thermodynamic properties which included an activity coefficient model for the liquid
434 phase and the Peng-Robinson EOS for the vapor-phase. Detailed process design was presented
435 along with a sensitivity study for recovering co-absorbed H₂ from the solvent.

436 Basha et al. completed two studies using Aspen Plus software to develop a process simulation that
437 captures CO₂ from an IGCC process using three different ionic liquids (TEGO IL K5, TEGO IL
438 P51P and [hmim][Tf₂N]) (Basha et al., 2014; Basha et al., 2013). The conceptual process has four
439 adiabatic absorbers in parallel modeled with a rate-based model with the Peng-Robinson EOS and
440 Boston-Mathias α function and standard mixing rules. The physical properties of the ionic liquids
441 were measured or obtained from literature and modeled as a function temperature. Their results
442 showed that the three ionic liquids were able to capture between 90-95 % CO₂.

443 Park et al. (2015) modeled three physical solvent absorption processes (Selexol, Rectisol and
444 Purisol) for the two stage (H₂S and CO₂) pre-combustion capture process for a 100 MW class
445 IGCC system. The Rad-Frac model in Aspen Plus was used to model the absorber with the PC-

446 SAFT property method used to predict the physical properties. The Selexol process was found to
447 be the most efficient when considering electric and thermal energy consumption.

448 Zhai and Rubin (2018) have used an “integrated technical and economic modeling framework” for
449 assessing the potential to use ionic liquids for pre-combustion CO₂ capture. The study used a
450 computer modeling program, developed by Carnegie Mellon University with support from
451 DOE/NETL, called the Integrated Environmental Control Model (IECM) (IECM, 2020). IECM
452 performs “systematic cost and performance analyses of emission control equipment at coal-fired
453 power plants” and can be used as a baseline case for the Selexol solvent absorption process. Zhai
454 and Rubin’s study investigated the separation of CO₂ and H₂ using the IL [hmim][Tf₂N] and
455 compared its performance to the Selexol process. The phase equilibrium model, Redlich-Kwong
456 Equation of State (RK-EOS), was used to predict the phase behavior of the gases in the solvent. A
457 multistage equilibrium model was used to model the absorption process. The study found that the
458 energy penalty when using ILs comes mainly from the process and product stream compression
459 and solvent pumping, while the absorbers and compressors represent the major capital costs.

460 Zhao et al. (2018) investigated the physical solvents DMC, [Omim][BF₄] and their mixtures for
461 CO₂ capture from high pressure sources. The COSMOtherm software package was used with the
462 COSMO-RS model to predict the Henry’s law constant and vapor pressure. Taheri et al. (2018)
463 also developed a process model for CO₂ absorption in an ionic liquid as well as methanol and their
464 mixtures. In this study a new thermodynamic method was used, called the UNIFAC-Lei model, to
465 predict the solubility of CO₂ in MeOH/[AMIM][Tf₂N]. The absorption process was then modeled
466 using the Rad-Frac equilibrium model in Aspen Plus to assess energy requirements, CO₂ capture
467 potential and solvent losses.

468 Roussanaly et al. (2020) compared the techno-economic performance of three technologies for
469 CO₂ removal from a lignite fired IGCC power plant located in the Czech Republic. The Rectisol
470 physical solvent absorption process was included in the analysis. Aspen Plus was used to model
471 the Rectisol process using an equilibrium column and the PR-EOS for estimating properties. In
472 addition to the carbon capture plant, the overall simulation was modified to include water-gas-shift
473 reactors and nitrogen addition from the air separation unit to the H₂-rich fuel to act as a diluent and
474 reduce the temperature of the gas turbine. H₂S and water removal from the syngas was assumed to
475 occur before the CO₂ absorption column.

476 Padurean et al. (2012) completed a techno-economic analysis for an IGCC power plant using
477 Aspen Plus to develop the process simulation for pre-combustion carbon capture using Selexol,
478 Rectisol, Purisol and the chemical solvent MDEA. The study found that Selexol was the most
479 energy efficient option due to its low energy (heat) consumption for solvent regeneration as well
480 as a relatively simple process configuration.

481 Urech et al. (2014) used Aspen Plus to assess different solvent-based capture technologies for a
482 black coal IGCC power plant with CCS. Selexol physical solvent absorption was used for CO₂
483 capture in addition to the chemical solvents MDEA and hot potassium carbonate. The study aimed
484 to assess the energy penalty of pre-combustion CO₂ capture by comparing the energy efficiency
485 of the power plant with and without CO₂ capture. All these capture processes reduced the overall
486 plant efficiency by between 7.6 and 8.6%. The highest efficiency case of 39.3% was found with
487 the hot potassium carbonate solvent, however, it was noted that this “does not necessarily give the
488 most economic cost of electricity or cost of CO₂ capture” and that a “full economic evaluation and
489 thermo-economic optimization” is required (Urech et al., 2014).

490 Heintz et al. (2008) developed a preliminary conceptual process design using the perfluorinated
491 compound PP25 for CO₂ capture from a shifted fuel gas mixture generated with an E-Gas gasifier.
492 The solubility of CO₂, N₂ and H₂ in PP25 were measured while the other components were
493 predicted using the PR-EOS in Aspen Plus. The process simulation was used to assess both
494 pressure swing and temperature-pressure swing regeneration options. Sun and Smith (2013)
495 developed an Aspen Plus simulation for a single stage Rectisol wash and a two-stage configuration
496 using the PC-SAFT model with revised binary interaction parameters (Sun and Smith, 2013).

497 Ashkanani et al. (2020) performed techno-economic analysis for CO₂ capture from an average fuel
498 gas stream in a pre-combustion power plant (543 MW) using the following five physical solvents:
499 Selexol, PEGPDMS-1, NMP, [aPy][Tf₂N] and [hmim][Tf₂N]). The absorption process operated
500 over a range of temperatures. Aspen Plus was used to develop the process simulation with the PC-
501 SAFT equation of state used to predict physical properties and gas solubility. The absorption
502 column was found to be most efficient with structured packing, as opposed to random packing,
503 due to improved mass transfer performance resulting in lower solvent flow rates and smaller flash
504 equipment. Additionally, lower temperatures resulted in better CO₂ absorption, due to increased
505 solubility of CO₂ in the solvent at lower temperatures.

506 Xin et al. (2020) used a rate-based modelling approach to compare the benchmark Selexol solvent
507 absorption process to novel deep eutectic solvent (DES) absorption for pre-combustion capture.
508 The liquid phase was modelled as plug flow while the gas phase was considered to be fully mixed.
509 Three mass transfer coefficient correlations were adopted in the rate-based process simulation
510 using Aspen Plus. For Selexol two of these correlations (Bravo et al. (1985) and Billet and Schultes
511 (1993)) were similar to the equilibrium model while the correlation by (Hanley and Chen, 2012)
512 deviated from equilibrium particularly at high L/G ratios and for high viscosity solvents.
513 Table 3 provides a summary of selected process simulations for CO₂ capture using physical
514 solvents including main model/simulation assumptions, syngas properties, CO₂ gas specifications
515 and scale of CO₂ capture. In addition, Table 4 provides a summary of available electricity or energy
516 consumption detail from the processes listed in Table 3. As discussed in this review, these results
517 should be compared with caution due to different assumptions between process designs, different
518 system boundaries, different scales and different operating parameters. Some authors model both
519 the full IGCC process and then the full IGCC-CCS process which allows estimation of differences
520 in the power output from the combined cycle due to operation with a H₂ rich rather than CO₂ rich
521 syngas. This lower power output from the combined cycle can be comparable or even larger than
522 the energy requirements for the whole CO₂ capture and compression processes. Although a plant-
523 wide approach is important for assessing energy requirements when adding CCS to an IGCC plant,
524 the focus of this review, including Table 3 and Table 4, is on physical solvent absorption processes
525 for CO₂ capture. But readers should be aware that the CO₂ capture and compression sub-systems
526 are not the only causes of lower power output between an IGCC-CCS and IGCC with equal feed
527 flowrates. For example, in the NETL baseline study (James et al., 2019) case B5B (IGCC with
528 Selexol two stage process), the net power is 556 MW whereas case B5A (IGCC with no CO₂
529 capture with equal feed flow rate) has a net power of 656 MW. However, as shown in Table 4, the
530 CO₂ capture and compression for case B5B comprises only 43 MW, which represents less than
531 half of the overall power loss when including CCS to the IGCC plant. Other important factors to
532 consider when comparing overall energy consumption between different physical solvent studies
533 are H₂S removal, CO₂ compression conditions, use of hydraulic turbine and net changes to
534 combined cycle power output. A sensitivity analysis completed by Zhai and Rubin (2018) also
535 showed the capture process power use can depend on multiple parameters including operating
536 temperature, absorption pressure, regeneration pressure and CO₂ removal efficiency.

Table 3: Summary of input conditions modeled in simulation studies for CO₂ capture using physical solvents

Reference	Solvents	Gasification type and/or syngas composition details	Gas flow rate, kg/s	Gas flux, kg/s/m ²	Gas Pressure, bar	Gas Temp. °C	Solvent Flow rate, kg/s	Liquid Flux, kg/s/m ²	L/G	Solvent Temp. °C	Absorber Diameter, m	Absorber height, m	Physical property model	Process simulator	CO ₂ capture rate, %	Flash pressure HP/MP/LP, bar	CO ₂ compression specifications
Dave et al. (2016)	Selexol	307 MWe Syngas: 42.3% CO ₂ , 46.6% H ₂	163	5.1	37	26	1,604	50	10	20	6.4	16	PR-EOS	ProTreat v5.2 2013	90	35.2	n/a
Basha et al. (2013)	[hmim][Tf ₂ N]	400 MWe E-Gas with Pitt. Coal #8, 23.9% CO ₂ , 37.5% H ₂	25.6*	5.7	30	227	1,038	229	40	24.9	2.4*	30*	PR-EOS & Span-Wager EOS	Aspen Plus	97.3	20/10/1	153 bar 260 K
Basha et al. (2014)	TEGO IL K5 TEGO IL P51P	400 MWe E-Gas with Pitt. Coal #8, 23.9% CO ₂ , 37.5% H ₂	25.6*	1.6	30	227	900 250	57 16	10- 35	24.9	4.5*	27*	PR-EOS & Span-Wager EOS	Aspen Plus v7.2	92	20/10/1	153 bar 260 K
Siefert et al. (2016a)	Selexol PEG-Siloxane1 IL – [aPy][Tf ₂ N]	464 MW GEE gasifier, 40.3% CO ₂ , 56.3% H ₂	147	4.4	52	40	1,514- 3,154	46-74	10- 17	10-40	6.5	36	PC-SAFT	Aspen Plus	90	20.7/11/1.5	15 MPa 323 K
Field and Brasington (2011)	Selexol (DEPG)	464 MW GEE gasifier, 40% CO ₂ , 56% H ₂	227	n/a	52	17	1,939	n/a	8.5	10	n/a	n/a	PC-SAFT	Aspen Plus v7.1	90	20.7/11/1.5	153 bar 50 °C
Park et al. (2015)	Selexol (DEPG) Rectisol (MeOH) Purisol (NMP)	Udhe entrained flow coal gasifier	n/a	n/a	37	0	95 7.3 145	35 1 24	n/a	-10 -20 to - 40	1.87 3 2.8	25 28 28	PC-SAFT	Aspen Plus v7.3	90	3 stage LP/MP/HP	n/a
Zhai and Rubin (2018)	[hmim][Tf ₂ N]	Shell gasifier, 41% CO ₂ , 59% H ₂	~78 ₊	n/a	29.6	29	2,439	n/a	~30	29.4	n/a	n/a	modified RK EOS	IECM v9.5	90	3 stages 10 to 0.8 bar	153 bar
Taheri et al. (2018)	Methanol 50:50 MeOH/IL [AMIM][Tf ₂ N] [AMIM][Tf ₂ N] [AMIM][Tf ₂ N]	Conceptual gas stream: 47% H ₂ , 34% CO ₂ , 9% CO	105	n/a	60	25	395 395 395 790	n/a	4-8	-30 -30 -30 -7.5 15	n/a	10 stages 10 stages 20 stages 22 stages 24 stages	UNIFAC-Lei	Aspen Plus	varies	0.1/0.8	98.8-100 % CO ₂
Roussanaly et al. (2020)	Rectisol	278 MW Shell gasifier (lignite) 38.6% CO ₂ , 53.4% H ₂	68	n/a	28.4	30	n/a	n/a	n/a	n/a	n/a	n/a	PR-EOS	Aspen Plus	89	3 stages	110 bar, 313 K, 95% purity
Urech et al. (2014)	Selexol	Shell gasifier 1200 MWth	n/a	n/a	30	40	2,554	52	n/a	40	7.9	16	n/a	Aspen Plus	90	18/2/0.3	100 bar
Ashkanani et al. (2020)	Selexol PEGPDMS-1 NMP [aPy][Tf ₂ N] [hmim][Tf ₂ N]	NETL case B5B, GEE IGCC – 543 MW, 40.8% CO ₂ , 56.8% H ₂	155	4.7-5.5 3.8-5.5 4.7-5.5 3.5-5.5 2.6-3.9	30	35	1300-2900 900-2400 1100-2500 2000-5000 2300-5200	39-87 22-58 33-75 45-113 38-86	5-35	0-50	6-6.5 6-7.25 6-6.5 6-7.5 7.1-8.75	38.6-42.9 39.4-47.1 39.4-43.5 39.4-50.6 45.9-55.8	PC-SAFT	Aspen Plus v8.8	90	15-22 /10.5/1	152.7 bar <600 ppm H ₂ O
Padurean et al. (2012)	Selexol Purisol Rectisol	Coal/biomass mixture 425-450 MWe, 39.9% CO ₂ , 54.7% H ₂	125	248	32	40 -15 to 20 -30 to-40	n/a	n/a	n/a	n/a	0.8	20	n/a	Aspen Plus v7.0	91.4 91.2 90.6	13	122 bar <200 ppmv H ₂ O
Xin et al. (2020)	Selexol Hypothetical DES	550 MW medium sulfur coal GE quench, 40% CO ₂ , 56% H ₂	~160	n/a	29.58	29.44	~3,500 - 6,500	n/a	20- 40	25-30	72% flooding criteria	10-20 m / 50 stages	PC-SAFT	Aspen Plus v11.0	70	17.3/6.7/2.7/1.05	130 bar 25 °C

Notes: n/a = not available in literature; *4 absorbers in parallel +2 trains for capture process

539 **Table 4: Summary of electricity consumption reported for physical solvent processes listed in Table 3**

Reference	Solvent	Electricity consumption details from manuscript	CO ₂ capture flow rate* (in kg/s)	Corresponding physical solvent process electricity (or equivalent) consumption (in kJ/mol CO ₂ captured)	Notes
Dave et al. (2016)	Selexol	Solvent pump power consumption: 7 MW	120 kg/s	CO ₂ capture solvent pump only: 2.6 kJ/mol CO ₂	Solvent pump only; H ₂ S not included in analysis; CO ₂ compression energy detail not included in manuscript.
Basha et al. (2013)	[hmim][Tf ₂ N]	AGR power requirement: 22.1 MW	53.7 kg/s	CO ₂ capture + compression: 18 kJ/mol CO ₂	H ₂ S removal excluded; CO ₂ compression included.
Basha et al. (2014)	TEGO IL K5 TEGO IL P51P	Power requirement of conceptual AGR process: TEGO IL P51P: 31.54 MW; TEGO IL K5: 39.18 MW	52.1 kg/s	CO ₂ capture + compression: TEGO IL P51: 27 kJ/mol CO ₂ TEGO IL K5: 33 kJ/mol CO ₂	H ₂ S removal excluded; CO ₂ compression included.
Siefert et al. (2016a)	Selexol PEG-Siloxane1	Overall difference in electricity generation between IGCC and IGCC-CCS: 31.7-32.6 kJ/mol CO ₂ (PEG-Siloxane1); 32.2 kJ/mol CO ₂ (Selexol) Estimated lowered power from combined cycle: 15.4 kJ/mol CO ₂ CO ₂ Compression: 10 kJ/mol CO ₂	124 kg/s	CO ₂ capture + compression: PEG-Siloxane1: 16.3-17.2 kJ/mol CO ₂ Selexol: 16.8 kJ/mol CO ₂	Overall values include direct capture energy & indirect energy costs, such as lowered power from gas turbine/steam turbine combined cycle. H ₂ S removal excluded; CO ₂ compression included.
Field and Brasington (2011)	Selexol (DEPG)	Major power consumption for 2-stage Selexol plant and CO ₂ compression: Acid gas removal subsystem: 16.7 MW _e + 7.1 MW _{e,eq} = 23.8 MW _{e,eq} CO ₂ compressor subsystem: 28.3 MW _e	129 kg/s	AGR + CO ₂ compression: 17.8 kJ/mol CO ₂	2 stage H ₂ S & CO ₂ AGR plus CO ₂ compression. Thermal energy for H ₂ S regen = 31.6 MW _{th} ≈ 7.1 MW _e eq. (See SI in Siefert et al. (2016b)).
Park et al. (2015)	Selexol Rectisol Purisol	Total electric power consumption: 3.214 MW (Selexol); 3.819 MW (Rectisol); 3.635 MW (Purisol) Total thermal power consumption: 0.858 MW (Selexol), 1.256 MW (Rectisol), 2.099 MW (Purisol).	Selexol: 17.3 kg/s Rectisol: 18.8 kg/s Purisol: 18.8 kg/s	Not calculated here due to inclusion of thermal energy.	2 stage H ₂ S & CO ₂ AGR; CO ₂ compression included.
Zhai and Rubin (2018)	[hmim][Tf ₂ N]	Solvent pumping: 18.1 MW; Process compression: 14.0 MW; CO ₂ product compression: 30.4 MW; Hydraulic turbine recovery power: -9.4 MW Total net system power use (IL-based CO ₂ capture): 53.1 MW	113.2 kg/s	CO ₂ capture + compression: 21 kJ/mol CO ₂	Net power includes power generated from hydraulic turbine. H ₂ S removal excluded; CO ₂ compression included.
Taheri et al. (2018)	Methanol 50:50 MeOH/IL [AMIM][Tf ₂ N]	Process specific energy penalty (combination of electrical & thermal energy): Pure methanol: 0.353 GJ/kmol captured CO ₂ Methanol + IL: 0.197 GJ/kmol captured CO ₂ Pure IL: 0.052, 0.142, 0.241 GJ/kmol captured CO ₂ (@ 243K, 266K, 288K)	Pure MeOH: 72.0 kg/s MeOH + IL: 72.0 kg/s Pure IL: 73.9, 65.9, 73.7 kg/s @243K, 266K, 288K	Not calculated here due to inclusion of thermal energy	No H ₂ S present in syngas. No detail provided for CO ₂ compression.
Roussanaly et al. (2020)	Rectisol	CO ₂ capture unit: 1.2 MWe CO ₂ conditioning unit: 13.9 MWe	50.2 kg/s	CO ₂ capture + conditioning: 13.2 kJ/mol CO ₂	H ₂ S removal excluded; CO ₂ conditioning included.
Urech et al. (2014)	Selexol	CO ₂ capture power consumption (Selexol plant): 39.41 MW	94.5 kg/s	Selexol only: 18.3 kJ/mol CO ₂	2 stage H ₂ S & CO ₂ AGR plus CO ₂ compression included.
Ashkanani et al. (2020)	Selexol PEGPDMS-1 NMP [aPy][Tf ₂ N] [hmim][Tf ₂ N]	Selexol: 47.1 – 52.4 MW PEGPDMS-1: 44.5 – 76.0 MW NMP: 56.4 – 73.5 MW [aPy][Tf ₂ N]: 46.7 – 70.1 MW [hmim][Tf ₂ N]: 53.4 – 73.2 MW	126.5 kg/s	Selexol: 16.4 – 18.2 kJ/mol CO ₂ PEGPDMS-1: 15.5 – 26.4 kJ/mol CO ₂ NMP: 19.6 – 25.6 kJ/mol CO ₂ [aPy][Tf ₂ N]: 16.2 – 24.4 kJ/mol CO ₂ [hmim][Tf ₂ N]: 18.6 – 25.5 kJ/mol CO ₂	Power consumption used in OPEX calculations for the paper. H ₂ S assumed to be removed from syngas. CO ₂ compression included.
Padurean et al. (2012)	Selexol Rectisol Purisol	Power duty (MW _{el}): 14.117 (Selexol); 14.121 (Rectisol); 22.845 (Purisol) Cooling duty (MW _{th}): 6.809 (Selexol); 8.244 (Rectisol); 94.434 (Purisol) Heat duty (MW _{th}): 34.08 (Selexol); 35.488 (Rectisol); 103.205 (Purisol)	~ 99 kg/s	Not calculated here due to inclusion of thermal energy	Includes both H ₂ S and CO ₂ removal but excludes compression and drying. (Add 25-35 MW for compression & drying).
NETL Report (James et al., 2019)	Selexol	Case B5B Acid gas removal (H ₂ S & CO ₂): 11.55 MWe; Case B5A (no CO ₂ capture) AGR (H ₂ S): 2.95 MWe; Case B5B CO ₂ compression: 31.67 MWe	126 kg/s (Case B5B)	AGR (Selexol CO ₂ & H ₂ S) + CO ₂ compression: 15.1 kJ/mol CO ₂	2 stage Selexol process includes CO ₂ & H ₂ S plus CO ₂ compression included.

540 *If captured CO₂ flow rate was not reported in manuscript, CO₂ capture flow rate was calculated from syngas flow rate, syngas composition and capture efficiency values presented in Table 3

541 From the summary above and Table 3, it can be seen that many of these process simulations have
542 some features in common, including the use of Aspen Plus to develop the simulation, requirement
543 of 90% CO₂ capture, and CO₂ product gas at around 153 bar. However, some parameters vary
544 significantly between each study, including operating pressure of the absorber (or syngas feed
545 pressure) which varies from 30 to 60 bar, varying gas and liquid flow rates and subsequently
546 varying absorption column dimensions. Most systems have a 3-stage flash process for solvent
547 regeneration, but other studies regenerate the solvent in a single stage or don't state the conditions
548 for which the solvent is regenerated or do not include compression of CO₂ gas stream.
549 Additionally, some simulations have validated physical properties and solubility predictions with
550 experimental data whereas others rely on built in correlations and thermodynamic models which
551 may not accurately represent the solvent system being assessed, particularly for newer solvents
552 (e.g., IL and DES (Xin et al., 2020)). It is also important to consider the absence or presence of
553 other syngas processes such water-gas-shift or the removal of other gas components like H₂S
554 before the CO₂ capture plant or simultaneously with the CO₂ capture process. These factors will
555 also influence the overall energy requirements as reported in Table 4 and therefore should be
556 compared with caution. Although process simulations can be a useful tool for predicting and
557 comparing the performance of different physical solvents for pre-combustion CO₂ capture, the
558 assumptions and models incorporated into the simulation must be transparent in order to conduct
559 a fair comparison between different studies, particularly when these results are then used as inputs
560 to a subsequent economic analysis.

561 **3.2 Economic models used in TEAs to assess the economic performance of physical solvents** 562 **for CO₂ capture**

563 To compare the techno-economic analysis results of CO₂ capture with different physical solvents,
564 the assumptions made in the economic models need to be consistent and transparent. When
565 estimating capital and operating costs, it is important to consider parameters, such as what other
566 gas components (e.g. sulfur) have already been removed from the syngas, electricity cost, capacity
567 factors, plant life and construction time, operating and maintenance costs, CO₂ capture rate,
568 inflation/discount rates, solvent costs, currency conversion, location factors, etc. Despite recent
569 advancements in the transparency and depth of CCS costing studies (Guandalini et al., 2019;
570 Rubin, 2019), van der Spek et al. (2019) have highlighted the significant need for improvement in
571 applying suitable cost engineering practices, including recent methodical advancements and

572 adaptations to the latest energy policies (van der Spek et al., 2019). This review also highlighted the
573 importance of including all assumptions and outputs transparently, including realistic “equipment
574 design and sizing; cost indices and location factors; process and project contingency costs; CO₂
575 transport and storage costs; and uncertainty analysis and validation”. A limited number of studies
576 have assessed the economic performance of pre-combustion CO₂ capture using physical solvents,
577 and the detail and depth of analysis in each study varies, leading to a range of outcomes, as
578 described below.

579 Siefert et al. (2016a) reported on the economic results for CO₂ capture from an IGCC process using
580 three physical solvents including Selexol, PEG-Siloxane-1 and the ionic liquid [aPy][Tf₂N].
581 Detailed information is provided for the equations used to determine the levelized cost of capture
582 (LCOC) as well as the associated assumptions used in the economic analysis. It was found that the
583 majority of the LCOC was independent of solvent used and as such there were no significant
584 differences in the economic performance of the chosen solvents. However, it was reported that
585 more significant reductions in cost could be achieved when using a hydrophobic solvent, such as
586 an IL, as it could be operated at higher temperature resulting in reduced operating costs and due to
587 its higher mass density, which reduces the size of absorption and flash equipment. The study also
588 highlighted the impact of assumptions made for unknown parameters, such as solvent cost, in the
589 economic analysis. A sensitivity analysis of IL solvent cost showed notable changes in the LCOC.
590 At an IL synthesis cost of \$40/L, the IL lowered the LCOC by 7% when compared to Selexol.
591 However, if the cost of the IL increased to \$400/L, then the cost of CO₂ capture using the IL could
592 be 16% larger than Selexol. In most studies, the cost of synthesizing novel solvents at commercial
593 scale is unknown, but this study shows solvent cost has an important impact on the overall
594 economics of carbon capture processes.

595 Padurean et al. (2012) conducted a techno-economic analysis on pre-combustion CO₂ capture from
596 an IGCC power plant using a range of solvents, including an alkanolamine (chemical solvent
597 MDEA), dimethyl ethers of polyethylene glycol (Selexol), chilled methanol (Rectisol) and N-
598 Methyl-2-pyrrolidone (Purisol). The total investment capital costs were assessed for the different
599 solvents at CO₂ capture rates ranging from 70 to 90%. Selexol was found to be the most energy
600 efficient and as such an economic analysis was then completed for this solvent. This study
601 highlighted the impact of operational parameters and assumptions on economic outcomes as the
602 capital costs increased from 19.55% for 70% CO₂ capture up to 22.5% for 90% CO₂ capture.

603 Zhai and Rubin (2018) developed an integrated technical and economic framework for assessing
604 the feasibility of pre-combustion CO₂ capture using the ionic liquid [hmim][Tf₂N] and compared
605 results to the Selexol process. The cost method applied in this study used models that predict
606 capital cost, annual operating costs and maintenance costs and finally the total annual levelized
607 cost of electricity (\$/MWh). The minimum cost of CO₂ separation was found to occur at 90% CO₂
608 removal efficiency. Cost comparisons for the entire IGCC-CCS process between the IL (\$63 per
609 ton of CO₂ avoided) and Selexol (\$65 per ton of CO₂ avoided) suggested that there was no strong
610 data for ILs to replace the established Selexol process. To understand what solvent properties are
611 required to improve the economic viability of a high capital cost solvent like ILs, a hypothetical
612 study was conducted with an IL that had a 10 % increase in CO₂ solubility. Results of the
613 simulations showed that the separation cost could be reduced by 29% if a solvent with 10% higher
614 CO₂ solubility could be developed. The plant-level evaluation (IGCC with CCS) completed in this
615 study highlighted the importance of new compression technology to reduce costs as well as
616 system-wide IGCC cost reductions (including process intensification), as the cost of CO₂ capture
617 accounts for less than 10% of an IGCC plant's total capital requirement.

618 Mota-Martinez et al. (2018) evaluated the economic performance of ILs for pre-combustion CO₂
619 capture. Their study proposed a methodology that used both monetized and non-monetized process
620 performance indices. The economic performance was based on the total annualized cost (TAC)
621 which comprised the sum of yearly capital expenditures (CAPEX) including a capital recovery
622 factor and yearly operating expenditures (OPEX). The CAPEX for each unit was estimated with
623 cost correlations that were dependent on key properties of the unit, such as heat transfer area for a
624 heat exchanger. As opposed to previous IL studies that focused on improving CO₂ solubility in the
625 solvent, this study found that solvent transport properties like viscosity and thermochemical
626 properties like heat capacity also had a strong impact on the total annualized cost. For example,
627 when comparing the capital costs for CO₂ capture with ILs, the costs of heaters and heat
628 exchangers were two orders of magnitude higher than the cost of the absorber (due to increased
629 viscosity) highlighting the fact that solvent transport properties can be just as important as solvent
630 capacity.

631 Cormos and Cormos (2019) assessed the techno-economic performance of an IGCC power plant
632 with CCS to evaluate the impact of a number of plant wide process configurations including the
633 choice of chemical (MDEA) or physical (Selexol) solvent when adding CCS. The economics of

634 each process were assessed by calculating the capital cost of equipment (using cost correlations),
635 the operating and maintenance costs, levelized cost of electricity and hydrogen, CO₂ capture costs
636 and cash flow analysis. The capital cost and the interest rate were found to have the greatest impact
637 on the electricity cost and the CO₂ avoided cost. For pre-combustion CO₂ capture, physical solvent
638 absorption was found to have slightly better economic performance when compared to the
639 chemical absorption process.

640 Porter et al. (2017) completed a techno-economic assessment of CO₂ capture for a range of power
641 plants, including pre-combustion CO₂ capture, with a focus on processes that deliver different
642 levels of CO₂ product gas purity. For pre-combustion, three different options for capturing sulfur
643 gas species and CO₂ were considered: (i) Selexol for co-capture of CO₂ and sulfur species and (ii)
644 Selexol for separate capture of CO₂ and sulfur species and (iii) Rectisol for separate capture of
645 CO₂ and sulfur species. The Integrated Environmental Control Model (IECM) (IECM, 2020) was
646 used to perform the techno-economic calculations as it allows for the numerous factors that impact
647 the power plant output, cost of electricity, emissions and cost of CCS at combustion based power
648 plants including IGCC. For 95% CO₂ removal, the Selexol co-capture plant was found to have the
649 lowest LCOE, mainly due to the lower capital cost and higher efficiency. The cost of CO₂ avoided
650 was highest for the Rectisol solvent process.

651 Chen and Rubin (2009) assessed the overall plant performance and cost for an IGCC power plant
652 with CCS using the Selexol process for CO₂ capture. This study investigated parameters such as
653 CO₂ removal efficiency and quality/cost of coal on the overall IGCC plant performance and cost.
654 The IECM framework (IECM, 2020) was used to simulate the performance and cost of the
655 different systems. The CO₂ avoidance cost was lowest for a CO₂ removal efficiency of
656 approximately 90%.

657 Cormos (2012) investigated the techno-economic indicators for IGCC with CCS using a Selexol
658 process for 90% CO₂ removal. The capital costs for each section of the plant were determined from
659 published cost data correlations and detailed information was provided for the operating and
660 maintenance costs. Adding CCS with 90% CO₂ removal increased the LCOE by 30-35% compared
661 to the plant without CCS. Roussanaly et al. (2020) conducted a techno-economic analysis of three
662 technologies, including Rectisol physical solvent absorption, for CO₂ capture from a lignite based
663 IGCC power plant located in the Czech Republic. In this study, the LCOE increased by 40% when
664 Rectisol solvent absorption was added compared to the plant without CO₂ capture. Additionally,

665 this study showed that lower CO₂ capture ratios (CCR) generally increase the CO₂ avoidance cost.
666 A later study by Roussanaly et al. (2021) highlighted the challenges and factors that can have a
667 significant influence on CCS cost evaluations, including “cost metrics, energy supply aspects,
668 retrofitting costs, CO₂ transport and storage and maturity of the capture technology”. Using
669 specific case study examples, the quantitative impacts of cost assumptions and methods were
670 shown to be significant and as such highlights the importance of clearly reporting any assumptions
671 and methods used in CCS TEA studies.

672 Xin et al. (2020) conducted an economic analysis of pre-combustion CO₂ capture for the
673 benchmark Selexol process and compared it to a hypothetical deep eutectic solvent to determine
674 how solvent characteristics contribute to the total cost of CO₂ separation. This study highlighted
675 variation in solvent performance and therefore economics when using different mass transfer
676 coefficient correlations. The optimum capture cost of Selexol ranged from 26.66 to 30.84 \$/ton
677 CO₂ (2018) depending on what correlation was selected in the mass transfer model. The analysis
678 also found that the DES could outperform Selexol with two of three mass transfer correlations,
679 however, as the viscosity of the DES increased the economics were less favorable.

680 Ashkanani et al. performed a techno-economic analysis for pre-combustion CO₂ capture using five
681 physical solvents from a fuel gas stream produced at a 543 MW power plant (Ashkanani et al.,
682 2019; Ashkanani et al., 2020). The levelized cost of CO₂ capture (LCOC) was calculated and
683 results showed that solvents operating at low temperatures provided the lowest LCOC values
684 compared to those operating at higher temperatures. The reduced LCOC was due to the increased
685 CO₂ solubility in the selected physical solvents at lower temperatures, which resulted in smaller
686 absorption columns and lower solvent circulation rates, which offset the higher cooling
687 requirements. The hydrophobic PEGPDMS-1 solvent had the lowest LCOC and was considered
688 the most promising due to its low capital and operating costs in addition to its lower corrosivity,
689 which enabled the use of less expensive materials of construction for the processing equipment.
690 Due to the recent interest in smaller or modular scale gasification technology, Ashkanani et al.
691 (2021) conducted another study to assess the impact of plant scale on the cost of pre-combustion
692 CO₂ capture using physical solvents. Solvent absorption processes with physical solvents
693 (PEGPDMS-1, PEGPDMS-3, [bmim][Tf₂N], and [emim][Tf₂N]) were modeled using the PC-
694 SAFT EOS in Aspen Plus v8.8 for power plant capacities ranging from 54 to 543 MW. The
695 simulations indicated that increasing the power plant capacity from 54 to 543 MW increased the

696 operating expenditure (OPEX) of the solvent absorption plant from 2.6 to 30 MM\$/year and the
697 capital expenditure (CAPEX) of the solvent absorption plant from 10 to 58 MM\$. However,
698 increasing power plant capacity decreased the OPEX and CAPEX when expressed in \$/ton CO₂
699 captured, reducing the levelized cost of CO₂ capture (LCOC) from 12.50 to 7.58 \$/ton CO₂
700 captured, due to the increased amount of the CO₂ removed at larger scale.

701 Jansen et al. (2015) have summarized the economic evaluation results for integrating CO₂ pre-
702 combustion capture in IGCC plants with wide variability in the cost of CO₂ avoided (30-86 \$/ton
703 CO₂) and the cost of electricity, COE (65-150 \$/MWh) reported. The main reason proposed for
704 the variability in these costs is the different assumptions and methodologies used to complete the
705 economic analysis, for example different contingencies, inclusion/exclusion of inflation rate, etc.
706 van der Spek et al. (2019) have also highlighted the importance of conducting “more rigorous
707 application of good cost engineering practices; the inclusion (and progression) of recent
708 methodological advances; and adaptation to changing policy focus” when assessing CCS
709 technology.

710 Zhang et al. (2020) conducted a comparative economic analysis on physical, chemical, and hybrid
711 physical-chemical absorption processes for a range of industrial scenarios. Comparative results
712 showed that for CO₂ removal from high pressure feed gases, a hybrid absorption process that
713 combines chemical and physical solvents was preferred from an economic perspective. The
714 economic analysis conducted in this study focused only on total annualized costs for physical
715 absorption, chemical absorption and a hybrid absorption process.

716 **3.3 Outlook for techno-economic analysis of physical solvents for CO₂ capture**

717 Significant research effort is underway to develop novel physical solvents for efficient CO₂ capture
718 from high pressure gas streams. However, as highlighted in this review, it is the cost of the overall
719 CO₂ capture process that will ultimately determine if a solvent is to be successfully implemented
720 in industrial applications. Fewer studies report on the economic performance of novel solvents
721 compared to the number of studies reporting on promising technical performance such as CO₂
722 solubility. Therefore, it is important to highlight the importance of conducting comprehensive
723 economic analyses for solvents with promising technical performance. A lack of transparency,
724 consistency and detail were generally found when reviewing the literature for specific parameters
725 and associated assumptions used in the economic models of TEAs for physical solvent CO₂
726 capture. For example, some studies specify an inflation rate (even if it is 0%) while no detail can

727 be found in other studies. Additionally, some studies report a real discount rate while others use a
728 nominal rate or do not state what is used. A summary of some of the economic model parameters
729 and assumptions have been provided in Table 5. As shown by the sensitivity analysis conducted
730 by García-Gutiérrez et al. (2016), economic parameters such as capital investment, plant operating
731 hours, interest rates, plant lifespan, electricity cost and solvent costs can have an impact on the
732 overall economic outcomes. In this sensitivity study, production costs were found to be most
733 sensitive to plant operating hours. Additionally, capital expenditure, which is often estimated with
734 errors of $\pm 30\%$, was found to result in big variations in production costs. The values assumed for
735 these parameters not only impact on the economic outcomes of the specific case being studied, but
736 also make it challenging when comparing performance and economic results from different
737 studies. These conclusions were also supported by a broader study on CCS techno-economic
738 analysis by van der Spek et al. (2020) which discusses the importance of uncertainty analysis to
739 provide more robust and meaningful CCS performance and cost conclusions.

740 **Table 5: Summary of input parameters used in selected economic models for CO₂ capture using physical solvents**

Reference	Physical solvent/s	CO ₂ capture rate, %	WGS costs included?	Base year	Discount rate, per year	Electricity price, \$/MWh	Plant life, yr	Construction time, yr	Capital charge factor, per yr	Capacity factor, fraction of max. design	Operating & maintenance costs, per year	Solvent cost, \$/L	CO ₂ transport & storage cost, \$/ton CO ₂
Chen and Rubin (2009)	Selexol	90	Yes	2008	n/a	102.8	30	4	14.8 %	75 %	5 % plant fixed costs	5 \$/kg (~5 \$/L)	9.5 \$/ton
Cormos (2012)	Selexol	90	Yes	n/a	8 %	n/a	25	3	n/a	n/a	3.5 %	6500 Euro/ton (~ 7 \$/L)	n/a
Siefert et al. (2016a)	Selexol PEG-PDMS-1 [aPy][Tf ₂ N]	90	Yes	2012	7 %	50	30	1 capture plant only	10 %	80 %	4 % total upfront capital/yr	4 \$/L Selexol 4 \$/L PEGPDMS1 40-400 \$/L IL	excluded
Zhai and Rubin (2018)	[hmim][Tf ₂ N]	90	Yes	2000	7.09 %	50	n/a	3	11.3 %	75 %	5 % total plant cost	10,000 \$/ton (~ 10 \$/L)	10 \$/ton
Mota-Martinez et al. (2018)	ILs	90	No	2017	10 %	50 \$/ton of coal	25	n/a	n/a	92 %	n/a	10 \$/kg (~ 14 \$/L)	20-25 \$/ton
Roussanaly et al. (2020)	Rectisol	85	Yes	2015	10 %	Internal utility of plant	25	3	n/a	85 %	4.5 % of EPC (Engineering, procurement, construction costs)	n/a	excluded
Ashkanani et al. (2020)	NMP, Selexol, PEGPDMS-1, [aPy][Tf ₂ N], [hmim][Tf ₂ N], PEGPDMS-3, [bmim][Tf ₂ N], [emim][Tf ₂ N]	90	No	2019	10 %	50	30	n/a	10.6%	80 %	4 % / year	2 \$/L NMP 4 \$/L Selexol 4 \$/L PEGPDMS-1 400 \$/L [aPy][Tf ₂ N] 415 \$/L [hmim][Tf ₂ N] \$4/L PEGPDMS-3 \$14.37/L [bmim][Tf ₂ N] \$30.38/L [emim][Tf ₂ N]	excluded
Xin et al. (2020)	Selexol DES	90	Yes	2018	12 %	120	25	3	Total overnight cost details provided	75 %	10% EPC	7,200 \$/ton Selexol 7,200 \$/ton DES	excluded

741 n/a = detail/parameter not available in manuscript.

742 4. Conclusions and future directions

743 CO₂ capture from high-pressure gas streams using physical solvents can be considered a mature
744 technology option as shown by the currently available commercial processes that have been in
745 operation for many years, including the Selexol and Rectisol processes. However, as recent studies
746 have shown, there are opportunities to improve the technical and economic performance of these
747 physical solvent processes, particularly when considering the solvents specifically for pre-
748 combustion CO₂ capture applications from IGCC processes rather than removal of acid gas from
749 synthetic or natural gas streams. In addition, physical solvents will be needed at AGR processing
750 facilities that can selectively capture and regenerate CO₂ at high pressure. Physical solvents that
751 can absorb more CO₂ at higher temperatures with high CO₂/H₂ selectivity and then undergo
752 regeneration at higher pressures than the current commercial solvents have the potential to reduce
753 the energy requirements and costs for all pre-combustion CO₂ capture applications. A number of
754 hydrophobic physical solvents and tunable ionic liquids have shown promising CO₂ absorption
755 performance at bench scale and in subsequent techno-economic analysis studies. However, as also
756 highlighted in this review, it is important to ensure that the process simulations and economic
757 analyses used to come to these conclusions are validated with reliable experimental data and also
758 ensure that the cost models utilize appropriate economic parameters with transparent assumptions.
759 In some cases, the improved absorption performance was shown to be offset by the higher cost of
760 the solvent or as a result of the need for more expensive ancillary equipment (such as heat
761 exchangers/pumps) due to unfavorable physical properties of the novel solvent.

762 As shown in this review, it is clear that novel physical solvents have potential to improve the
763 efficiency and economics of pre-combustion carbon capture and will be needed for meet future
764 net-zero CO₂ eq. emission goals. However, in order to overcome the challenges of (a) comparing
765 the costs of novel physical solvents to baseline solvents and (b) understanding the uncertainties
766 around these new processes, more in-depth TEAs are required that use (1) validated process
767 models, (2) clear economic assumptions, and (3) comparison to baseline technologies. Future
768 studies should also include critical analysis of economic operating parameters, such as solvent
769 cost, to fully understand the overall impact of cost assumptions and associated uncertainties on
770 process design and economic performance. These steps will be important in advancing the
771 development of novel physical solvent processes as understanding both the technical and economic

772 performance is an essential step for future demonstration and commercialization of physical
773 solvent processes for pre-combustion carbon capture.

774 **5. Acknowledgment**

775 This technical effort was performed in support of the US Department of Energy’s ongoing research
776 in Carbon Capture. The research was executed through the NETL Research and Innovation
777 Center’s Transformational Carbon Capture field work proposal (FWP). We thank Mr. Rui Wang
778 at the University of Pittsburgh and Dr. Dushyant Shekhawat for their assistance in reviewing this
779 manuscript.

780 **6. Disclaimer**

781 This work was funded by the Department of Energy, National Energy Technology Laboratory, an
782 agency of the United States Government, through a support contract with Leidos Research Support
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