Energy Minimization of Carbon Capture and Storage by means of a Novel Process Configuration

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11 Abstract

12 Carbon capture and storage is considered a key technology for decarbonizing the heat and power 13 industries and achieving net zero emission targets. However, the significant energy requirements of the process as currently utilized hinders its widespread implementation. This work presents a novel process 14 15 configuration by which the energy expenditures of carbon capture and storage can be minimized. This 16 configuration is intended to enhance heat integration during the capture process through an innovative 17 combination of three stripper modifications, namely lean vapor compression, a rich solvent split with vapor heat recovery and reboiler condensate heat recovery using a stripper inter-heater in a single flow-sheet. For 18 19 carbon dioxide compression, a novel pressurization strategy involving carbon dioxide multi-stage 20 compressors, a heat pump system and a supercritical carbon dioxide power cycle was designed and 21 evaluated. The heat pump was used for carbon dioxide liquefaction while the supercritical carbon dioxide 22 power cycle was employed to recover the intercooling heat. Through a comprehensive parametric 23 investigation of the proposed configuration, the optimum value of the key operating parameters i.e., the 24 split fraction, flash pressure, stripper inter-heater location, stripper inter-heater solvent flowrate, carbon 25 dioxide liquefaction pressure and supercritical carbon dioxide cycle turbine pressure ratio were estimated. 26 The performance of the proposed design at the optimized condition was quantified in terms of the reboiler 27 heat duty, the carbon dioxide pressurization power and the equivalent work and compared to a baseline 28 case post-combustion carbon capture and storage process. The proposed case reduced the reboiler heat duty 29 from 3.36 GJ/TonneCO₂ to 2.65 GJ/TonneCO₂ and the electric power required for carbon dioxide 30 compression from 16,691 kW to 14,708 kW. The results demonstrate that the new design can significantly

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- 31 reduce the reboiler duty, compression power and equivalent work by 21.1%, 11.88%, and 15.8%,32 respectively.
- 33
- 34 **Keywords:** Post-combustion carbon dioxide capture, Novel capture process configuration, Hybrid carbon
- 35 dioxide pressurization system, Supercritical carbon dioxide power cycle
- 36 Nomenclature
- 37 Stoichiometric coefficient of component *i* in reaction equation. a_i 38 $a^{g,l}$ Area of the gas-liquid interface Conventional multistage compression established as the baseline case 39 BC 40 CCS Carbon capture and storage Concentration of the ith component 41 C_i Specific heat capacity at constant pressure (J/kg.K) 42 CP Temperature difference 43 dT 44 E Activation energy (Cal/mol) Electrolyte non-random two-liquid **ENRTL** 45 46 Genetic algorithm GA Enthalpy (J/kg) 47 h Henry Constant 48 Η 49 Pre-exponential factor k 50 Κ Overall mass transfer coefficient 51 m Mass flow rate (kg/s) Rate of mass transfer 52 Μ Monoethanolamine 53 MEA Exponent of Temperature, which is zero 54 n 55 Ν Number of components in the reaction 56 ORC Organic Rankine Cycle 57 Р Pressure (kPa) 58 PP Pinch point in the heat transfer process Rate of heat transfer (W) 59 Q Reaction rate. 60 r 61 R Universal Gas Constant Refrigerants' degree of superheat at the HP's compressor inlet (K) 62 SH 63 Entropy (J/kg.K) s Temperature (K) Т 64 HP Heat pump 65 66 W Power (W) 67 Х Quality 68 State 1CO₂–12CO₂ CO₂ states 69 State 1Refr-4Refr Refrigerant R290 states 70

71	Greek Symbols	
72	φ	Percentage of savings in equivalent work
73	η	Isentropic efficiency of compressors and pumps
74	γ	Activity coefficient in liquid phase
75		
76	Subscripts	
77	BC	Conventional multistage compression established as the baseline case
78	С	Cooling temperature
79	Comp	Compressors
80	Comp2	CO ₂ compressor at the end of CCS chain
81	CW	Cooling water
82	Eq	Equivalent work
83	Eva	Evaporator
84	FG	Flue gas
85	In	Incoming CO ₂
86	is	Isentropic
87	0	Ambient conditions
88	Out	Conditioned CO_2 at the outlet
89	ORC	Organic Rankine Cycle
90	PC	Proposed design
91	R	Ratio
92	Refr	Refrigerant or HP working fluid
93	SC	Subcooling
94	sCO ₂	Supercritical CO ₂
95	State 1CO ₂ -12CO ₂	CO ₂ states
96	State 1Refr-4Refr	Refrigerant R290 states
97	Turb	Turbine
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99		
100	1. Introduction	

101 Carbon capture and storage (CCS) is a key technological measure for reducing the emission of 102 greenhouse gases (GHG) from stationary emission sources such as thermal power plants and ensuring a 103 sustainable and reliable global energy supply. CCS has the potential to significantly reduce carbon dioxide 104 (CO₂) emissions and mitigate climate change [1] and is, therefore, regarded as one of the most important 105 research frontiers for the development of a sustainable future [2]. Different technologies, including oxy-106 fuel combustion, pre-combustion and post-combustion [3], as well as different materials that are capable of 107 selective sorption of CO₂ (chemical or physical) [4] are available for CO₂ sequestration.

Post-combustion CO₂ capture, making use of an aqueous amine absorption process, is an especially
 promising technology because it is inherently flexible and can be integrated into various energy systems or

retrofitted into existing facilities [5]. The carbon capture step is followed by CO_2 transportation to the storage site, as shown in Fig. 1. Both of the major steps involved in the CCS process – CO_2 capture and transportation to a storage site – are energy-intensive processes and can produce up to a 14% reduction in net plant efficiency. This significantly high energy penalty hinders the implementation of CCS into power plants [6].



Fig. 1. Carbon capture and storage process.

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116 In the amine absorption process, CO_2 is absorbed at 40-60 °C in an absorber column that makes use 117 of an aqueous amine solvent. This CO_2 -rich solvent is then thermally treated at 110-130 °C in a stripper 118 column to produce pure CO₂. The thermal treatment of the CO₂-rich solvent requires a large amount of 119 energy and accounts for approximately 70% of the total process cost [7]. Some researchers have focused 120 on integrating CCS with renewable heat sources such as solar energy in order to provide the energy required 121 for the thermal treatment of the solvent. However, this option remains economically unfeasible, as a large 122 solar field is required to meet the high energy needs of solvent thermal treatment [8]. Therefore, various 123 research activities are aimed at curtailing the energy expenditure of the CCS process by either improving 124 the solvent for the absorption process [9] or the process configuration [10]. Improvement in the process 125 configuration is easier and more effective, because it can be achieved with minimal retrofitting of any 126 existing facility.

127 Different process configurations such as absorber inter-cooling [10], stripper inter-heating [11], 128 rich solvent splitting [12], stripper overhead compression [13], lean vapor compression (LVC) [14], rich 129 vapor compression [14] and many other process modifications have been reported to reduce the energy 130 penalty of the process. Dmartiz et al. [15] compared the energy consumption of various amine absorption process configurations and concluded that improvement to the configuration has substantial potential to 131 132 improve the energetic efficiency of the process. Furthermore, the combination of different process 133 configurations has more potential to reduce the energy consumption of the process. A combination of 134 absorber inter-cooling, advanced rich solvent splitting and stripper inter-heating was simulated using ASPEN PLUSTM and resulted in an 18% reduction in reboiler duty [16]. Haider et al. [17] also examined 135 136 the combination of capture process modifications and achieved 14% reduction in the reboiler duty.

137 The complete CCS process entails transporting sequestered CO_2 from the capturing facility to the permanent storage site. Pipelines are considered a reliable and efficient mean of CO₂ transportation where 138 139 the required CO_2 pressure ranges are from 150-200 bar [18]. Therefore, in the first step of the storage 140 process, the pressure of the CO_2 is to be boosted from the captured pressure (1.2-3.5 bar) to the pipeline 141 pressure. The CO_2 pressurization can result in an energy penalty of as high as 12% of the loss of power 142 plant efficiency [19]. Therefore, to curtail this significant energy expenditure, extensive past research has 143 focused on improving the CO₂ compression process. Witkowski et al. [20] investigated a range of 144 compression chain strategies and concluded that the requisite compression power can be reduced by 145 designing a compression chain that makes use of CO_2 liquefaction and pumping. This technique involves 146 compressing the incoming CO_2 from the capture unit to some intermediated liquefaction pressure, after 147 which it is liquefied and then pumped to the target pressure. The liquefaction of CO_2 requires sub-zero 148 condensing temperatures, which makes the use of ambient sink impractical. Therefore, some research 149 activities sought to design refrigeration cycles for CO₂ liquefaction [21]. Alabdulakarem et al. [22] designed a vapor compression cycle (VCC) for CO₂ liquefaction and pumping and explored various refrigerants. 150 151 They concluded that by complementing the multistage compression with an ammonia-based vapor 152 compression cycle, the power consumption could be reduced by 5.1% compared to conventional multi-153 stage compression.

The efficiency of the CO₂ compression process can also be improved by recovering compression heat to produce useful work. Romeo et al. [23] optimized the CO₂ compression process by investigating the compression ratio and converting the compression heat of the CO₂ to low-pressure steam in the plant. Meanwhile, Kurtulus et al. [24] integrated an Organic Rankine Cycle (ORC) with CO₂ compressors and conducted a thermodynamic analysis of their system. Pei et al. [25] conducted a similar analysis and concluded that the coupling of an ORC to shockwave-based CO₂ compression is more promising than simple intercooling compression. Farajollahi et al. [26] studied the impact of integrating an ORC into a 3250 MW thermal power plant with post-combustion CO_2 capture and found out that the efficiency of the plant increased from 31.26% to 33.4% through the application of ORC.

Recently, to reduce the energy expenditure of the CCS process, the integration of fossil-fueled 163 164 plants with solar hybrid systems has also been aggressively researched. Xu et al. [27] investigated a novel, 165 direct-fired, oxy combustion supercritical CO₂ (sCO₂) power plant integrated with a solar-driven coal 166 gasification array. They found that the proposed design reduced coal consumption by 29.9% and achieved a net energy efficiency of 43.4% with near zero carbon emissions. Similarly, Ghorbani et al. [28] proposed 167 168 a tri-generation system to produce liquefied natural gas (LNG) and liquefied CO₂ using an adsorption refrigeration system, and desalinated water. Their analysis revealed that hybrid poly-generation systems 169 170 can enhance the exergetic efficiency of the system up to 88.97%. Although hybrid systems exhibit promising potential to decarbonize the industry for greenfield plants, they lack the capacity to be retrofitted 171 172 with the existing facility. Therefore, this study sets out to design an advanced CCS process configuration 173 that can be adapted to greenfield plant designs or retrofitted with existing plants to reduce the energy 174 expenditure of the CCS process.

175 Previous studies demonstrate that extensive research activities have focused on improving the 176 capture process configuration and investigating a liquefaction system design for CO₂ pressurization [29]. 177 However, the majority of these research efforts have addressed these problems separately; therefore, this 178 study aims to remedy this gap by evaluating the entire process chain of CO_2 capture and liquefaction. 179 Recently, Aliyon et al. [30] examined the compete CO_2 capture and liquefaction process; however, their 180 study employed a conventional capture process configuration and ship-based CO₂ transportation. The CO₂ 181 target pressure for their ship-based transportation was 19.7 bar and 26.5 bar, which makes the design of 182 their CO₂ pressurization system significantly differs from pipeline-based transportation.

183 The originality of this work includes incorporating the complete CCS process while proposing and 184 optimizing a new design for capture, as well as the pressurization process. For the capture process, the combination of stripper modifications has shown improvement potential. Jin et al. [31] reported a 28% 185 186 reduction in energy consumption by incorporating a combination of stripper modifications, which included 187 an air stripper, flue gas membrane pre-separation, inter-cooler and rich solvent split. However, their study 188 did not consider the auxiliary loads of the added equipment, while the installation of a membrane increases 189 the complexity of the process. In this study, three stripper modifications, namely LVC; a rich solvent split 190 with vapor heat recovery; and heat recovery from reboiler condensate using stripper inter-heater are 191 incorporated into a single flowsheet and auxiliary loads are accounted for while calculating net energy

192 consumption. For the pressurization process, Muhammad et al. [29] found that integrating liquefaction and 193 pumping into the pressurization system can save the electric power, while Kurtulus et al. [24] demonstrated that the intercooling heat of CO₂ compressors can be recovered to produce power. This study consolidated 194 195 the effects of liquefaction and pumping and the recovery of intercooling heat by devising a hybrid CO₂ 196 pressurization scheme. The proposed pressurization scheme involves multi-stage compressors, a heat pump 197 (HP) system for CO_2 liquefaction and an sCO₂ power cycle to recover the intercooling heat. Unlike previous 198 studies that investigated ORC to recover the intercooling heat, this study employs an sCO_2 cycle. The sCO_2 199 cycle offers better efficiency compared to the ORC while harnessing low- to mid-temperature heat sources 200 [32] and integrating the sCO₂ cycle with the proposed CO₂ pressurization scheme does not require any 201 additional working fluid inventory. The capture process configurations were simulated and optimized using ASPEN PLUSTM V10.1 rigorous rate-based modeling, whereas the sCO₂ cycle integrated pressurization 202 203 scheme was modelled and optimized in the MATLAB environment. The energy expenditure of the 204 proposed capture and pressurization configuration was then calculated at the optimized conditions. The 205 resulting reduction in heat duty and electric power using the proposed design is converted to equivalent work (W_{Eq}) for a fair comparison with the baseline case (BC). 206

207 2. Amine-based carbon dioxide capture process description and modelling

208 The monoethanolamine (MEA) was used as an absorbent during the CO₂ capture process. The 209 liquid phase was modelled using Electrolyte Non-Random Two Liquid (ENRTL) property package while 210 the vapor phase was modelled using the Redlich-Kwong (RK) equation of state. Absorber and stripper 211 columns were simulated using rigorous rate-based models. FLEXIPAC 250Y was used as packing material 212 in the absorber and stripper columns. Absorber and stripper specifications for the CCS process are presented 213 in Table 1. The CO₂ capture process was designed for 90% efficiency at a removal rate of 190 Tonne/hr. 214 The CO₂ capture process was designed for 300 MW coal-based power plants with a designed capacity of 215 1.5 million tonnes of CO_2 captured per annum. The flue gas composition was set in accordance with the guidelines provided by the Department of Energy (DOE) for the CO_2 capture process. The flue gas molar 216 217 composition and other specifications are provided in Table 2. These simulation bases were retained 218 throughout the analysis.

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Component		Specification		
Plan	t type	Coal-based, ~300 [MW]		
Absorber	Height	24 [m]		
	Diameter	13 [m]		
Stripper	Height	11 [m]		
Suppor	Diameter	8 [m]		
Amine	flowrate	2900 [Tonne/hr]		
CO ₂ capture	e rate (m _{CO2})	190 [Tonne/hr]		
CO ₂ capture efficiency		90%		
CO_2 proc	luct purity	99 wt.%		

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Table 2. Flue gas specifications

Component	Molar composition
CO ₂	13 [mole %]
O ₂	5 [mole %]
H ₂ O	10 [mole %]
N_2	72 [mole %]
Pressure (P _{FG})	1.5 [bar]
Temperature (T _{FG})	40 [°C]
Flowrate (m _{FG})	300 [kg/s]

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226 2.1. Process chemistry

The capture process, essentially a chemical process involving a variety of species and a series of reactions, was modelled using Aspen Plus® [33]. A water, amine and CO₂ mixture were thermodynamically modeled using the ENRTL property package in Aspen Plus® [33]. The chemical reactions taking place in the process were modelled using the power law given in Eq. (1). The reactions and their kinetic parameters; rate constant (k) and activation energy (E) used in Eq. (1), taken from [31], are given in Table 3.

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$$r = k(T)^n \exp\left[\left(\frac{-E}{R}\right)\right] \prod_{i=1}^N C_i^{a_i}$$
(1)

Reactions	Туре	Rate constant (k)	Activation energy (E) [Cal/mol]
$H_2O + MEAH^+ \leftrightarrow MEA + H_3O^+$	Equilibrium	-	-
$2H_2O \leftrightarrow H_3O^+ + OH^-$	Equilibrium	-	-
$HCO_3^- + H_2O \leftrightarrow CO_3^{2-} + H_3O^+$	Equilibrium	-	-
$CO_2 + OH^- \leftrightarrow HCO_3^-$	Kinetic	4.32e+13	13249
$HCO_3^- \leftrightarrow CO_2 + OH^-$	Kinetic	2.38e+17	29451
$MEA + CO_2 + H_2O \leftrightarrow MEACOO^- + H_3O^+$	Kinetic	9.77e+10	9855.8
$MEACOO^- + H_3O^+ \leftrightarrow MEA + CO_2 + H_2O$	Kinetic	3.23e+19	156554

235 2.2. Mass transfer in the carbon dioxide capture process

The mass transfer rate in the gas-liquid interface is expressed by Eq. (2) [34], below. The overall
mass transfer coefficient is calculated by Eq. (3) [34].

238

$$M_{i} = a^{g,l}N_{i} = a^{g,l}K_{tot,i}(P_{i}^{eq} - P_{i}) = a^{g,l}K_{tot,i}(H_{E,i}\gamma_{i}c_{i} - P_{i})$$
(2)
$$\frac{1}{K_{eq}} = \frac{RT}{R_{eq}} + \frac{H_{E,i}}{R_{eq}}$$
(3)

$$\overline{K_{tot,i}} = \overline{k_i^g} + \overline{k_i^l} \tag{3}$$

239

The individual coefficients of mass transfer for species "i" are represented by k_i^g and k_i^l in the gas and liquid phases, respectively. Henry's Constant for CO₂ solubility in water is obtained by regressing Vapor Liquid Equilibrium data [35], while that for CO₂ in MEA is obtained from Wang et al. [36]. The non-random two-liquid (NRTL) model's interaction parameters between MEA & water are obtained from Austgen et al. [37]. The NRTL interaction parameters are set to zero between CO₂ and water. The interfacial area ($a^{g,l}$) and mass transfer coefficient in the liquid (k_i^l) and gas (k_i^g) phases are derived from Bravo et al. [38].

247 2.3.1 Baseline case: The carbon dioxide capture process

The conventional CO₂ capture process is shown in Fig. 2. The 30 wt.% MEA solvent enters from
the top of the absorber column and absorbs CO₂ from the flue gas emanating from the bottom of the absorber.
After the absorption of CO₂, clean gas exits from the top of the absorber. The CO₂-rich solution from the

251 bottom of the absorber is then pumped to an economizer before regenerating the CO_2 -rich solvent in the 252 stripper column. In the stripper column, CO_2 is stripped from the CO_2 -rich solvent by heating it in a reboiler. 253 The stripped CO_2 , along with the evaporated water vapors, exit from the top of the stripper column and 254 move to the condenser, where they are cooled. The cooled 2-phase mixture is separated in a flash column 255 that sends the condensed water back to the stripper as reflux and the separated CO_2 to the compression unit 256 to increase the pressure up to 150 bar for transportation and storage. The lean solvent from the stripper 257 bottom exchanges heat in the economizer with CO₂-rich solvent and is cooled to 40 °C before feeding back 258 into the absorber.

259



Fig. 2. Baseline case CO₂ capture process flowsheet.

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261 **2.3.2.** Baseline case for carbon dioxide pressurization

The CO₂ pressurization is an essential step to prepare the captured CO₂ for transportation. Conventionally, the CO₂ is pressurized using multi-stage compression, which is taken as a BC in this study. Each stage of the multi-stage compression comprises a compressor and intercooler, as shown in Fig. 3. The number of stages required depends on the captured CO₂ pressure (P_{In}), target pressure (P_{Out}) and the stage pressure ratio (P_R). The process parameters taken for CO₂ compression are tabulated in Table 4, while the net electric power consumed in the conventional compression process (W_{Comp,BC}) is given by Eq. (4).

$$W_{Comp,BC} = \sum_{i=1}^{N} W_{Comp} \tag{4}$$

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Fig. 3. Baseline case CO₂ compression unit.

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- 272

Table 4. Performance parameters of CO₂ pressurization during multi-stage compression

Parameters	Values
Compressor and pump isentropic efficiency (η)	0.80
Captured CO ₂ pressure (P _{In})	1.9 [bar]
CO ₂ target pressure (P _{Out})	150 [bar]
Cooling temperature (T _C)	30 [°C]
Pinch point (PP)	5
Compressor pressure ratio (P _R) across each stage	3

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274 2.4.1. Proposed carbon dioxide capture process configuration

275 This proposed novel configuration aims to reduce the heat required for the regeneration of amine 276 solvent by the addition of a HP and integrating process waste heat within the system, as shown in Fig. 4. 277 The CO₂-rich solution stream splits into two sub-streams before entering the economizer. One stream 278 follows the conventional path by entering the economizer and then moving to the stripper column. The 279 other moves to the vapor heat exchanger and recovers heat from the hot CO₂/H₂O vapors mixture exiting 280 the stripper top. This modification (shown with red lines in Fig. 4) will not only reduce the stripper duty, 281 but also the condenser duty by recovering a part of the heat that was being wasted in the condenser. The 282 solvent from the vapor heat exchanger enters the stripper and the H_2O/CO_2 mixture moves to condenser. 283 The stripper column is integrated with a pump-around heater (stripper inter-heater). Some of the solvent 284 from the stripper column is withdrawn and the exchange heat in the stripper inter-heater is then pumped back to the stripper. Saturated steam at 130 °C is used as the heating utility in the stripper reboiler. The
condensate from the reboiler outlet is used as a heating utility in the stripper inter-heater. The modification
is marked with violet lines in Fig. 4.

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Fig. 4. Proposed CO₂ capture process flowsheet.

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290 A HP was installed to reduce lean loading and add energy to the system. Instead of pumping the CO_2 -lean solution from the stripper reboiler into the economizer, it was flashed in a two-phase separator, 291 292 removing more CO_2 and reducing the lean loading of the CO_2 -lean solution. The gas separated in the flash 293 column was compressed back to the stripper pressure and returned to the stripper (as marked with green 294 lines in Fig. 4). The lean solvent from the flash column was pumped to the economizer and further cooled 295 before entering the absorber column. Even though additional electric power is needed to run the added 296 compressor, the reduction in the reboiler's heat duty remains more prominent. For a reasonable comparison, 297 the heat duty and electric power are brought to a common basis in terms of equivalent work, which is 298 discussed later.

299 2.4.2. Proposed design for the carbon dioxide pressurization process

300 In the proposed pressurization schematics, the multi-stage compression is assisted by the HP 301 system, as shown in Fig. 5. Increasing the pressure of the liquid CO₂ is significantly less energy-intensive than gaseous compression. Therefore, the HP is used as a refrigeration cycle to liquefy the CO₂, which is
 subsequently pumped to the required pressure.

The incoming CO_2 from the capture unit is 1.9 bar, whereas the triple point pressure of CO_2 is 5.17 bar. Therefore, the initial two stages of compression are necessary to raise the CO_2 pressure beyond the triple point pressure. The HP system can thus be installed after either the second or third stages. This study evaluated the performance of the proposed design with HP installed after the second as well as third stage, which is discussed in Section 3.1.5.





Fig. 5. Proposed heat pump assisted CO₂ compression schematics.

The difference between the conventional and the proposed approach for CO₂ pressurization is elucidated in a P-h diagram in Fig. 6. The baseline approach is multi-stage compression while in the proposed case the last stage is replaced by CO₂ liquefaction and subsequent pumping to the target pressure. 313



Fig. 6. Thermodynamic routes for CO₂ pressurization.

314 2.4.3. Modelling framework for proposed carbon dioxide pressurization process

The refrigerant of the HP, which absorbs heat (Q_{Eva}) from the CO₂, is then compressed and adjusted 315 316 to expel heat into the environment. Propane (R290) is considered as the HP refrigerant in this study. In the evaporator, the saturation parameters of R290 are determined by the pinch point (PP) limit and CO_2 317 318 liquefaction temperature. The CO_2 liquefaction temperature is the saturation temperature, corresponding to 319 its liquefaction pressure (P_{7CO2}). State 1Refr in Fig. 5 is determined by identifying the refrigerant's 320 saturation temperature in the evaporator and assuming a degree of superheating (SH) at the HP's 321 compressor inlet. The ambient conditions, which are considered the heat sink for R290, determine the 322 saturation temperature and pressure of R290 in the condenser and, consequently, the State 3Refr. State 323 2Refr and 4Refr were determined by using a compressor model with isentropic efficiency and assuming 324 isenthalpic expansion.

The calculation procedure of the HP system in the proposed schematics is summarized in Table 5. The discretization scheme is used for the modeling of heat exchangers, details of which can be found in [39]. The net power consumed in the proposed schematics ($W_{Net,PC}$) is given by Eq. (5), where $W_{Comp,CO2}$, W_{Pump} and $W_{Comp,Refr}$ are the power consumed by the initial multi-stage CO₂ compressors, the CO₂ pump and the HP compressor, respectively.

$$W_{Net,PC} = W_{Comp,CO2} + W_{Pump} + W_{Comp,Refr}$$
⁽⁵⁾

330

339

Table 5. Calculation schematics of the proposed heat pump system

Step	State	Temperature	Pressure	Enthalpy	Comments
1	7CO ₂	$T_{7CO2} = T_C$	P _{7CO2}	f(T _{7C02} , P _{7C02})	Using the set values of P_R , T_C , and η_{Comp} ; State $7CO_2$ is solved.
2	8CO ₂	$T_{8CO2} = T_{Sat,8CO2}$ $= f(P_{8CO2})$	$P_{8CO2} = P_{7CO2}$	f(T _{8C02} , X=0)	Calculate Q _{Eva} using heat exchanger model.
3	9CO ₂	$T_{9CO2} = f(P_{9CO2}, \label{eq:general} \eta_{Pump})$	P _{9CO2}	f(T _{9CO2} , P _{9CO2})	Pump model.
4	1Refr	$T_{1Refr} = f(T_{Sat,4Refr} \ , \label{eq:transf}$ SH)	$P_{1Refr} = P_{Sat,1Refr} = f(T_{4Refr})$	$f(P_{1Refr}, T_{1Refr})$	SH is taken as 5 K.
5	2Refr	$T_{2Refr} = f(P_{Sat,2Refr} \ , \label{eq:T2Refr}$ $\eta_{Comp})$	$P_{2Refr} = P_{Sat,2Refr} = f(T_{3Refr})$	$f(P_{2Refr}, T_{2Refr})$	Compressor model.
6	3Refr	$T_{3Refr} = T_{Sat,3Refr} = T_C$	$P_{3Refr} = P_{Sat,3Refr}$	f(T _{Sat,3Refr} , X=0)	Heat exchanger model.
7	4Refr	$T_{4Refr} = T_{Sat,4Refr} =$ f(T _{8CO2} , PP)	$P_{4Refr} = P_{Sat,4Refr} = f(T_{4Refr})$	f(P _{4Refr} , h _{3Refr})	Isenthalpic expansion.

343 2.4.4. Proposed heat pump pressurization process integration with supercritical carbon dioxide cycle

344 During the multi-stage CO₂ compression, the temperature of the CO₂ at each compressor outlet can reach around 400 K, as is shown in Fig. 6. To make use of this temperature, an innovative scheme of CO_2 345 346 liquefaction and pumping integrated with the sCO_2 cycle, as displayed in Fig. 7 is proposed. In contrast to the ORC, the sCO₂ cycle is designed to utilize the intercooling heat, as no additional working fluid is 347 348 required to operate the sCO₂ cycle. As can be seen in Fig. 7, the heat source and cycle working fluid are 349 both CO₂, and therefore a better temperature match is achieved by employing an sCO₂ cycle. In addition, 350 as the captured CO_2 is liquefied using the HP system, therefore the high-pressure level required to operate 351 an sCO₂ cycle which is generally in the range of 150-250 bar can be attained in an energy-efficient manner by means of the pump. For the integrated design, CO₂ from the State 8CO₂ is initially pressurized to 200 352 353 bar and State $9CO_2$. The high-pressure CO_2 is later divided into three streams, the flowrates of which are given in Fig. 7. The splitting of the total flow is performed in such a way so that the temperature of the 354

streams after heating at States $10CO_2$, $10CO_2'$ and $10CO_2''$ is comparable. Thereafter, the streams are mixed (State $11CO_2$) and subsequently fed into a turbine to produce electric power.

For the sCO₂ power generation, the turbine power output is W_{Turb} and the power consumed by the compressor to recover the pressure from State 13CO₂ to 14CO₂ and complete the process is $W_{Comp2CO2}$. The net power of the sCO₂ cycle is expressed in Eq. (6).

360

$$W_{Net,Cycle} = W_{Turb} - W_{Comp2,CO2} \tag{6}$$

361 After the CO_2 has delivered power to the turbine, it is cooled down and subsequently compressed 362 to 150 bar (State 14CO₂), thereby completing its flow (Fig. 7). After integrating the cycle, the $W_{Net,PC}$ is 363 modified as follow:

364

$$W_{Net,PC} = W_{Comp,CO2} + W_{Pump} + W_{Comp,Refr} - W_{Net,Cycle}$$
(7)

365



Fig. 7. Heat pump system integrated with a supercritical CO₂ cycle.

366

367 2.5. Total equivalent work/performance indicator calculation

368 In the CCS process, two different energy expenditures are incurred: (i) the electrical power 369 consumed in the pumps and compressors; and (ii) the heat duty in the boiler. For a fair comparison, a 370 common basis for the two types of energy is needed, and therefore W_{Eq} is defined for a performance 371 comparison. The W_{Eq} for the base and the proposed configurations are calculated using Eq. (8):

$$W_{Eq} = \sum \eta_{Turb} * Q_i \left((T_i + 10[K] - T_{Sink}) / (T_i + 10[K]) \right) + W_{Eelctric}$$
(8)

where the turbine efficiency (η_{Turb}) is considered to account for the non-ideal expansion, T_i is the reboiler temperature (K), the temperature of the steam in the reboiler is 10 °C higher than T_i , Q_i is the reboiler duty for the base and proposed case (GJ/TonneCO₂) and $W_{Electric}$ is the net electric power consumed during the complete CCS process. All the power entities, electric power and heat, consumed in the base as well as the proposed case, are converted to W_{Eq} using Eq. (8). After calculating the total equivalent work for the base ($W_{Eq,BC}$) and the proposed case ($W_{Eq,PC}$), the net effect (ϕ_{Net}) of the proposed schematics is defined as:

$$\Phi_{Net} = (W_{Eq,BC} - W_{Eq,PC})/W_{Eq,BC}$$
(9)

The calculation flow chart of the complete CCS configuration proposed is summarized in Fig. 8.

380



Fig. 8. Calculation flowchart of the proposed carbon capture and storage process configuration.

381

382 **3. Results and discussion**

Initially, the BC was simulated to calculate the reboiler duty, which amounted to 3.36GJ/TonnesCO₂ for a CO₂ capture rate of 190 TonneCO₂/hr. The four-stage compressors, with a P_R of 3, were established as the BC for CO₂ pressurization. The BC consumes 16691.40 kW of electric power to increase the pressure of CO_2 from 1.9 to 150 bar. After quantifying the BC, the capture process design is optimized for: split fraction, flash pressure, stripper inter-heater location and stripper inter-heater solvent flowrate. The HP-assisted CO_2 compression integrated with the s CO_2 power cycle is optimized to yield the best performance.

390 3.1. Split ratio

391 The CO_2 -rich stream is split to enhance the heat integration within the CO_2 capture process. The 392 objective is to recover maximum heat from the top and bottom streams, leaving the stripper. The amount of heat recovered from any of these streams depends on the CO₂-rich solvent flowrate against the respective 393 394 heat exchanger. The split ratio to the economizer was varied from 0.65 to 0.74 in order to study the effect 395 on the stripper reboiler duty. Fig. 9 shows that 0.67 is the optimum split ratio, which corresponds to the 396 minimum reboiler duty. The split ratio below 0.67 reduces the CO₂-rich solvent flowrate to the economizer 397 and the heat recovered in it. Although heat recovery in the vapor heat exchanger increases due to a higher 398 CO₂-rich solvent flowrate, the sum of the total heat recovery in the economizer and vapor heat exchanger 399 is lower than at the split ratio of 0.67. Similarly, above the 0.67 split ratio, the economizer can recover more 400 heat, but the heat recovery in the vapor heat exchanger declines. The overall impact is the reduction in total 401 heat recovery. The split ratio of 0.67 is the optimum split ratio at which the overall heat recovered in the 402 economizer and vapor heat exchangers is at a maximum and the stripper reboiler duty is minimal.



Fig. 9. Reboiler duty variation with split fraction.

403 **3.2. Flash pressure**

Lean solvent from the stripper reboiler is flashed in a column to strip out the CO_2 , which is compressed back to the stripper pressure. Additional electrical energy is required for the compression of CO₂. The amount of energy required for compression depends on the flashing pressure. However, this additional energy increases the temperature of CO_2 which, upon returning to the stripper, not only adds

- 408 energy to the stripper but also acts as a mass and energy stripping agent and reduces the stripper duty. Fig.
- 409 10 shows the impact of flash pressure on the compression energy requirement and the stripper reboiler duty.
- 410 As the reboiler duty is in the form of heat energy and the compressor duty is in the form of electric work,
- 411 W_{Eq} , is used as the key parameter to determine the optimum flash pressure (Eq. (8)). The impact of the flash
- 412 pressure on W_{Eq} is shown in Fig. 11, which reveals that 1.3 bar is the optimum flash pressure and
- 413 corresponds to the minimum total W_{Eq} .



Fig. 10. Effect of the flash pressure on reboiler duty and compressor power.

Fig. 11. Effect of the flash pressure on equivalent work.

414 **3.3. Stripper inter-heater location**

Saturated steam, used as the reboiler heating utility, exits as condensate at 129 °C. To recover heat from the reboiler condensate, stripper inter-heater is introduced. The stripper has 20 stages. Rich amine is fed from the top (stage 1) while lean solvent leaves from the bottom (stage 20). Amine solvent from a higher stripper stage is withdrawn and heated in a stripper inter-heater using reboiler condensate, and is fed back to a lower stage. To find the optimal withdrawal and feedback location, the withdrawal and feedback stage was varied, and the stripper duty observed.

Fig. 12 shows the impact of withdrawal and feedback stage on the stripper reboiler duty. The liquid at the top stages of the stripper column is at a relatively lower temperature and, therefore, has a high temperature difference from the reboiler condensate and can thus recover more heat. Withdrawal from the lower stage results in a lower temperature difference from the reboiler condensate and, consequently, less heat can be recovered.

426 As the CO₂-rich solvent is heated in the inter-heater, the CO₂ gas and water vapors begin separating 427 from the solvent. When this vapor-liquid mixture is fed back to a lower stage, the vapor phase interacts 428 with more liquid coming down and strips more CO_2 from the CO_2 -rich solvent, thus reducing the reboiler

429 duty. Stage 2 is the optimum withdrawal location and stage 15 the optimum feedback stage. Feeding below

430 stage 15 does not induce any substantial change in reboiler duty.



Fig. 12. Stripper inter-heater location optimization.

431

432 **3.4. Stripper inter-heater flowrate**

Hot solvent from the stripper inter-heater, when fed back to the stripper, serves as a stripping agent. The effectiveness of the stripping agent improves at higher temperature and reduces the reboiler duty [40]. Eq. (10) shows that higher temperatures ($\Delta T_{solvent}$) can be achieved by reducing the solvent flowrate (m_{solvent}). However, due to the thermal degradation of the solvent, the solvent temperature in the stripper inter-heater should not exceed 120 °C.

In order to find the optimum flowrate at which the reboiler duty is at a minimum and the solvent temperature is below the degradation temperature (120 °C), the m_{solvent} to stripper inter-heater was reduced from 3000 kmol/hr to 1000 kmol/hr and its impact on the reboiler duty and solvent outlet temperature was evaluated. Fig. 13 shows that by reducing the inter-heater flowrate, the temperature of the solvent fed back to stripper increases, which also improves the stripping efficiency and reduces the reboiler duty. However, due to the solvent degradation temperature limit, 1750 kmol/hr. is considered the optimum m_{solvent}, which corresponds to a stripper inter-heater solvent temperature of below 120 °C and a lower reboiler duty.

$$Q = m_{solvent} \times c_p \times \Delta T_{solvent} \tag{10}$$



Fig. 13. Effect of stripper inter-heater flowrate on reboiler duty and solvent temperature.

447 **3.5. Optimizing the carbon dioxide pressurization process**

448 The proposed pressurization process aims to reduce the required electric power to raise the CO_2 449 pressure by replacing one or two stages of CO_2 compressors with an HP system, consequently reducing W_{Comp,CO2}. However, due to the application of HP, another source of electric power consumption becomes 450 involved in the form of the compressor of the HP, i.e. W_{Comp,Refr}. Therefore, in the proposed design, 451 452 W_{Comp,CO2} and W_{Comp,Refr} are in a trade-off relationship. Table 6 summarizes the performance of the proposed 453 design with the HP installed after the second and third stage of the compressors. Installing the HP after the 454 third stage of the compressor instead of the second will seemingly increase the W_{Comp,CO2}. However, as can be seen in the table, the W_{Comp,CO2} increases up to 46.49%. At the same time, however, the W_{Comp,Refr} is 455 reduced by 85.80% while the difference in W_{Pump} remains negligible. 456

Table 6. Performance evaluation with the heat pump installed after the second and the third stages of compressors.

New Design Liquefaction Pressure [kPa]		CO2 Multistage Compression (W _{Comp,CO2}) [kW]	CO ₂ Pump (W _{Pump}) [kW]	HP-Compressor (W _{Comp,Refr}) [kW]	
2-Stage CO ₂ Compressor & HP	$P_{5CO2} = 1710$	9191.50	1130.70	7581.70	

3-Stage CO ₂ Compressor & HP	$\mathbf{P} \qquad \mathbf{P}_{7\text{CO2}} = 5130 \qquad 13465.00$		1149.00	1076.60	
Percentage change	N/A	+46.49%	-1.62%	-85.80%	

The substantial decrease in $W_{Comp,Refr}$ with the HP installed after the third stage is due to the combined reduction in refrigerant flowrate (m_{Refr}) and the P_R across the HP's compressor. Table 7 tabulates the Q_{Eva}, the saturation pressure of the refrigerant in the evaporator and condenser, the resulting P_R and the m_{Refr}. Q_{Eva} decreases and the saturation temperature of the refrigerant in the evaporator increases with the increase in CO₂ liquefaction pressures. Therefore, the m_{Refr} and P_R both decrease, leading to a considerable reduction in W_{Comp,Refr}. The subsequent results and discussions are based on HP installed after the third compressor stage.

468

Table 7. Operating characteristics of the heat pump system installed after the second and the third stages ofcompressors.

New Design	Liquefaction Pressure	Evaporator heat load	Refr. sa pressur	turation re [kPa]	Refr. flow rate (m _{Refr}) [kg/s]	P _R across HP's comp
	[kPa]	$(\mathbf{Q}_{\mathrm{Eva}})$ [kW]	Evaporator	Condenser		
2-Stage CO ₂ Compressor & HP	$P_{5CO2} = 1710$	18461.54	171.16	1079.26	68.44	6.30
3-Stage CO ₂ Compressor & HP	$P_{7CO2} = 5130$	10983.51	642.90	1079.26	34.72	1.68
Percentage change	N/A	-40.51%	N/A	N/A	-49.27%	-73.33%

471

472 **3.6.** The integrated supercritical carbon dioxide cycle optimization

473Due to the advantages discussed earlier, the compression heat is recovered using an integrated sCO_2 474cycle (Fig. 7). For an sCO_2 cycle at a given turbine inlet temperature, an optimum P_R across the turbine is475required to maximize the cycle performance [41]. In the analysis, the turbine inlet pressure (P_{11CO_2}) is fixed476to 200 bar, and hence the turbine outlet pressure (P_{12CO_2}) is parametrically investigated in this study. Fig.47714 shows the variation of $W_{Net,Cycle}$ with P_{12CO_2} , while the individual variation of W_{Turb} and W_{Comp2,CO_2} with

478 P_{12CO2} is shown in Fig. 15. As can be seen in Fig. 15, the W_{Turb} varies steadily throughout; however, the

- 479 $W_{\text{Comp2,CO2}}$ experienced a sharp decrease at the start, with P_{12CO2}. The sudden reduction of $W_{\text{Comp2,CO2}}$ is due
- 480 to the operation of the compressor near the critical point where the properties of CO₂ substantially vary.
- 481 Fig. 16 shows the variation of CO_2 density at the compressor inlet with P_{12CO2} . The drastic initial increase
- 482 in CO_2 density results in a sharp reduction of $W_{Comp2,CO2}$, which causes the sCO₂ power cycle to behave in
- the manner presented in Fig. 14.



Fig. 14. Supercritical CO_2 power generation ($W_{Net,Cycle}$) variation with turbine outlet pressure (P_{12CO2}).



Fig. 15. Turbine power (W_{Turb}) and compressor power $(W_{Comp2,CO2})$ variation with turbine outlet pressure (P_{12CO2}) .



Fig. 16. Compressor inlet density variation with turbine outlet pressure (P_{12CO2}).

The progress of the CO₂ through the HP and sCO₂ cycle to the final state $14CO_2$ is revealed in the P-h diagram in Fig. 17. For each intercooling stage, a certain amount of heat is recovered while that remaining is ejected into the environment. A simple sCO₂ rather the recuperative sCO₂ cycle is integrated to generate power, due to a low outlet temperature of the turbine (T_{12CO2}).



Fig. 17. Pathway of CO₂ in the proposed pressurization process.

491 The integrated sCO₂ cycle generates a net power of 982.61 kW, with a cycle efficiency (η_{Cycle}) of 492 9.01% and a heat source temperature around 400 K. Furthermore, the new design utilized the CO₂ prior to 493 storage, with no additional working fluid inventory required to operate the cycle. Table 8 summarizes the 494 state of CO₂ and R290, with the HP installed after the third stage.

Table 8. State properties of CO₂ and R290 in the proposed pressurization process.

State	Fluid	Temperature [K]	Pressure [kPa]	Enthalpy [kJ/kg]
1CO ₂	CO ₂	303.16	190	2.54
$2CO_2$	CO_2	402.45	570	90.46
$3CO_2$	CO_2	348.74	570	40.17
3CO ₂	CO_2	303.16	570	-0.95
$4CO_2$	CO_2	403.03	1710	85.27
5CO_2	CO_2	340.74	1710	24.47
5CO ₂	CO_2	303.16	1710	-12.15
6CO ₂	CO_2	404.51	5130	68.83
7CO_2	CO_2	324.74	5130	-25.01
7CO ₂	CO_2	303.16	5130	-57.65
8CO ₂	CO_2	288.50	5130	-265.75
9CO ₂	CO_2	306.74	20000	-244.00
9CO_2	CO_2	306.74	20000	-244.00
9CO2 ^{//}	CO_2	306.74	20000	-244.00
10CO ₂	CO_2	394.14	20000	-35.47
$10 \text{CO}_2^{/}$	CO_2	391.21	20000	-41.31
10CO2 ^{//}	CO_2	390.47	20000	-42.80
11CO ₂	CO_2	392.33	20000	-39.06
12CO ₂	CO_2	318.87	7500	-71.04
13CO ₂	CO_2	303.16	7500	-215.00
14CO ₂	CO_2	320.17	15000	-201.70
1Refr	R290	288.50	642.91	595.3
2Refr	R290	313.17	1079.26	626.6
3Refr	R290	303.16	1079.26	279.4
4Refr	R290	283.50	642.91	279.4

496

497 **4. Performance comparison based on equivalent work:**

After optimizing the capture and pressurization process, the $W_{Eq,PC}$ is calculated using Eq. (8). Table 9 summarizes the power consumptions of the base and proposed case, as well as the ϕ_{Net} . The performance of the proposed system is reported at the optimized conditions. As Table 9 shows, the new design successfully reduces the boiler duty from 3.36 to 2.65 GJ/TonneCO₂. The modifications to the capture process include optimizing the split ratio in the splitter, adding a heat exchanger as the stripper inter-heater and adding a compressor for the HP. The optimization of the split ratio reduces the reboiler's

⁴⁹⁵

heat duty by achieving a better thermal match within the overhead heat exchanger and the economizer. For the stripper inter-heater, the condensate from the reboiler served as the heating utility and no additional heat source was required. The new capture process design involves an additional compressor in the HP, as discussed in section 3.1.2, which results in an additional electric power consumption of 1112 kW. The detailed thermodynamic state properties of the complete proposed system are provided in the supplementary material (Appendix A and B).

510 The electric power requirement for CO₂ pressurization was reduced from 16691 to 14708 kW using 511 the sCO₂ cycle-integrated HP liquefaction and pressurization system. The proposed CCS design results in 512 a significant saving of $\phi_{\text{Net}} = 15.8\%$ in equivalent work. The ϕ_{Net} shows the unified impact of heat and 513 electric power, while the individual saving on the heat duty and electric power is 21.1% and 5.22%, 514 respectively. This study is concerned with minimizing energy expenditure for the CCS process, which improves the prospects of CCS systems being deployed in the power plant sector. However, the reduction 515 in energy consumption is achieved by adding a heat exchanger and a compressor to the capture process and 516 517 assisting the compression chain with a HP refrigeration cycle and sCO₂ cycle. The proposed pressurization system has the advantage of reducing a CO_2 compressor stage, however, it involves an additional HP and 518 sCO₂ cycle footprint. To quantify the economic behavior of the proposed system would require detailed 519 520 cost modeling of the capture process, the CO₂ compression chain and HP system components. The current 521 paper focuses on the energy expenditure of the proposed system, while the future outlook entails an 522 economic assessment of the proposed design.

- 523
- 524

Table 9. Performance indicators of the base and the proposed system.

	Reboiler Heat Duty [GJ/Tonne CO ₂]	Electric Power in Capture Process [kW]	CO2 Compre- ssion [kW]	CO2 Pump [kW]	HP Compressor [kW]	sCO2 Power [kW]	Equival- ent Work
Baseline Case (BC)	3.36	N/A	16691	N/A	N/A	N/A	0.974
Proposed Case (PC)	2.65	1112	13465	1149	1077	983	0.820

525

526 **5.** Conclusion

527 This study designed and evaluated a novel carbon capture and storage process configuration. The 528 proposed configuration successfully reduced the energy expenditure of the carbon capture and storage 529 process by enhancing heat integration. During the capture process, effective heat integration was achieved 530 by incorporating three stripper modifications, namely: lean vapor compression, rich solvent splitting with 531 vapor heat recovery and reboiler condensate heat recovery using an inter-heater stripper in a single 532 flowsheet. The power required for the CO₂ pressurization was reduced by assisting the multistage compressors with a heat pump system. The multistage compressors then increased then CO₂ pressure to the 533 534 level needed for liquefaction. The CO₂ was then liquefied in the heat pump system and subsequently 535 pumped to the required pressure. To improve the CO_2 pressurization process, intercooling heat lost during 536 multistage compression was recovered by integrating the heat pump-assisted pressurization scheme with a 537 supercritical CO₂ power cycle.

538 The performance of the proposed configuration was evaluated for a range of key operating 539 parameters, i.e. split fraction (from 0.65 to 0.74), flash pressure (from 1.0 to 1.6 bar), stripper inter-heater 540 location (from 2 to 18 stages) and flow rate (1000 to 3000 kmol/hr), as well as CO₂ liquefaction pressure (1710 and 5130 kPa), and the low pressure level of the supercritical CO_2 cycle (from 70 to 100 bar). The 541 542 parametric investigation results indicated the energy expenditure of the proposed configuration was 543 minimized at the optimum values of the split fraction, flash pressure, stripper inter-heater location, stripper inter-heater flow rate, CO₂ liquefaction pressure, and pressure ratio across supercritical CO₂ cycle turbine. 544 545 The performance of the proposed configuration at the optimized conditions was quantified in terms of 546 equivalent work and it was concluded that 15.8% saving in equivalent work compared to the conventional 547 carbon capture and storage process was achieved using the proposed process configuration. The proposed 548 process configuration demonstrates technical superiority by significantly reducing the energy consumption 549 of both the sequestration and pressurization processes. However, this study has focused on the potential 550 energy savings without considering the capital costs of the proposed modifications, which should be taken 551 into account to ascertain the economic viability of the proposed design.

552

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