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3 Energy analysis of the cryogenic CO₂ capture

4 process based on Stirling coolers

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20

21 **Abstract**

22 In the existing coal-fired power plants, the energy penalty associated with CO₂
23 capture process is an important challenge. For this reason, energy analysis has been
24 widely used as a powerful tool to optimize the capture efficiency and reduce energy
25 consumption. In our previous work, a Stirling cooler based cryogenic CO₂ capture
26 system was outlined. Process simulation and energy analysis of the system was
27 undertaken in this research. The whole CO₂ capture process is composed of three
28 sections; pre-chilling, CO₂ anti-sublimation and storage. The energy consumption of
29 each section in the system was investigated in detail. The results show that when the
30 flow rate of flue gas (13 vol.% CO₂) is set at 5 L/min and the temperature of Stirling
31 cooler -1, 2 and 3 is set at -30, -120 and -120 °C, the energy consumption of the
32 pre-chilling, CO₂ anti-sublimation and storage sections are 15.58_{thermal} J/s, 30.48_{thermal}
33 J/s and 11.40_{thermal} J/s, respectively. The total energy consumption of the cryogenic
34 CO₂ capture system is 57.46_{thermal} J/s (equal to 689.52 J/L flue gas).

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36 **Keywords:** CO₂ capture, cryogenic, Stirling cooler, energy consumption

42 **1. Introduction**

43 At present, a number of large industrial power stations have become dominant
44 greenhouse gas (GHG) emitters, and coal-fired power plants have been paid wide
45 attention as one of the most important. According to World Energy Outlook 2009 of
46 the International Energy Agency (IEA), by 2030 coal will account for approximately
47 44% of world energy consumption [1]. In the face of this situation, integration of CO₂
48 capture and storage (CCS) technologies into CO₂ emission sources is an effective
49 strategy to mitigate increasing climate issues.

50 In recent decades, several CO₂ capture and storage technologies for
51 post-combustion power plants such as solvent absorption, sorbent adsorption,
52 membrane permeation and cryogenic fractionation have been developed [2]. Among
53 these techniques, the chemical absorption method has been identified widely in
54 research as the most mature technology, due to its significant advantage of being used
55 for different CO₂ sources even for dilute CO₂ concentrations (3 vol.% for gas turbine)
56 [3]. Adsorption is a promising alternative to the CO₂ absorption method. The main
57 technologies are thermal (TSA), electrical (ESA) or pressure/vacuum (PSA/VSA)
58 swing adsorption, which use solid adsorbents in a number of adsorption columns [4].
59 The common solid adsorbents are activated carbon, zeolite, CaO, etc [5]. Furthermore,
60 application of the polymeric membrane on CO₂ separation has been widely
61 investigated in recent years due to its significant characteristics (i.e. high selectivity
62 and high driving force) [6]. Cryogenic fractionation is a well known technique for air

63 separation and also has been utilized in oxy-fuel combustion CO₂ capture processes
64 [7,8]. The typical cryogenic CO₂ capture process contains single or twin column
65 distillations, Ryan Holmes process, controlled freezing (Exxon) [9-11]. Compared to
66 conventional low temperature processes, the Stirling cooler system can separate CO₂
67 from the diluted flue gas (typically 13% CO₂) and the capture process is implemented
68 under atmosphere [12,13].

69 Although research in CO₂ capture technologies has made remarkable progress, the
70 energy penalty for existing technologies is still high (around 50 \$ per ton of CO₂
71 avoided) [14]. The major challenge for existing CO₂ capture technologies is to lessen
72 the energy consumption of the capital and operational costs. For the absorption and
73 adsorption processes, a large amount of heat is required to regenerate the solvents and
74 sorbents [15,16]. Membrane and cryogenic methods are more suitable for high CO₂
75 concentration sources. When the CO₂ is diluted in the flue gas (lower than 30 vol.%),
76 the energy consumption of these processes will obviously increase [17,18]. Therefore,
77 growing attention has been paid to energy reduction of CO₂ capture and storage
78 technologies. For example, different chemical absorption process configurations have
79 been investigated to reduce energy consumption [19,20]. Meanwhile, novel materials
80 have been developed as absorbents and adsorbents to reduce the energy penalty of the
81 absorption and adsorption processes [21]. In 2012, Luis et al. summarized the current
82 situation of membrane CO₂ capture technologies [22]. As they pointed out, the
83 existing permeation processes are still expensive and few membrane modules are

84 commercially available. In 2011, Tuinier et al. evaluated the energy consumption of a
85 novel post-combustion CO₂ capture process based on cryogenic packed beds [18].
86 Their research indicated that 3.6 MJ electric energy (for per kg CO₂) would be
87 consumed to generate the required refrigeration condition. In addition, previous
88 research indicates that CO₂ capture is an intricate and integrated process, and various
89 forms of energy are utilized [23]. Therefore, energy analysis of the capture process
90 plays a significant role in the improvement of capture efficiency and the reduction of
91 energy penalty.

92 In light of these concerns, efforts are implemented in energy analysis to reduce the
93 energy penalty of CO₂ capture processes. The objective of the work is to simulate and
94 analyze the energy efficiency of the cryogenic CO₂ system based on Stirling coolers
95 (SCs). In order to achieve this aim, the detailed energy consumption of different
96 stages in the process has been investigated under different conditions (e.g., flow rate
97 of gas stream, temperature of Stirling cooler and CO₂ concentration).

98 The paper is structured as follows: Section 2 describes the base case of the
99 cryogenic CO₂ capture process. Section 3 analyzes the overall energy demand of the
100 process and simulates the thermodynamic characteristics of the cryogenic system.
101 Section 4 introduces detailed simulation work and the typical parameters that
102 influence the energy requirement of the system. Section 5 investigates the energy
103 requirements of different stages in the system. Economic evaluation and comparison
104 with other technologies are also carried out. Section 6 summarizes the main

105 conclusions and prospects for future work.

106 **2. Base case description**

107 The schematic for the anti-sublimation CO₂ capture process is shown in Fig. 1. The
108 main part comprises the Stirling coolers (SCs), vacuum pump, freezing tower, camera,
109 control panel and structure frame [24,25]. The whole process consists of 3 sections: 1)
110 pre-chilling tower; 2) CO₂ anti-sublimation tower; 3) storage column.

111 *2.1 Pre-chilling tower*

112 Initially, the flue gas is pre-chilled by SC-1 in the pre-chilling tower. There are two
113 important functions of the pre-chilling process. Firstly, the gas stream can be cooled
114 down to a low temperature which is beneficial to improve the total CO₂ capture
115 efficiency. Simultaneously, the pre-chilling process can effectively avoid the
116 mechanical damage caused by a sharp temperature drop. Secondly, in the pre-chilling
117 tower, the moisture in the feed gas condenses into water under low temperature
118 conditions. Owing to the subsequent hot gas stream there is no ice generated during
119 the whole process. The separated water flows out from the outlet to avoid clogging
120 the vessel which is a key issue of the anti-sublimation separation technology.
121 Meanwhile, the other gas flows into the CO₂ anti-sublimation tower.

122 *2.2 CO₂ anti-sublimation tower*

123 In the CO₂ anti-sublimation tower, SC-2 provides a cryogenic condition

(approximately -110°C), and the dry flue gas is chilled to below -100°C . According to the work of Clodic et al., the freezing point of CO_2 is related to its partial pressure in the gas mixture [26]. Fig. 2(a) shows the relationship between freezing point and CO_2 concentration. Typically for the flue gas (3~20 vol.% CO_2) in Fig. 2(b), the freezing point of CO_2 varies between $-112 \sim -97^{\circ}\text{C}$. Therefore, under low temperature in the CO_2 anti-sublimation tower, CO_2 in the flue gas could solidify into dry ice and frost onto the heat exchanger of SC-2 immediately. In this way, the capture of CO_2 from the feed gas is realized. It should be pointed out that the frosted CO_2 on the heat exchanger of SC-2 will adversely affect the heat and mass transfer process. For this reason, a motor driven scraper is used to scrape the deposited CO_2 down to the storage column.

2.3 Storage column

The last step of the whole capture process is to gather the captured CO_2 by SC-3. In the previous section, by spinning the scraping rod on the heat exchanger of SC-2, dry ice falls down into the storage column. Meanwhile, SC-3 provides a low temperature condition (below -78.5°C) to store the dry ice and prevent it from gasifying. On the other hand, residual gas (such as N_2) exhausts from the gas outlet without phase change. The detailed CO_2 deposition and storage process in the system is shown in Fig. 3. As latent heat with deposited CO_2 is substantial, we added a heat exchanger between the subsequent stream and frosted the CO_2 to recover it.

144 2.4 Cryogenic performance of cryogenic CO₂ capture system

145 In order to understand the intricate CO₂ capture process better and improve the
146 capture efficiency the cryogenic performances of the three stages in the system are
147 outlined in this section. Some geometrical assumption is put forward to simplify the
148 intricate process. The detail structure of the columns used in the system is depicted in
149 Fig. 4. The geometric dimension of pre-chilling, CO₂ anti-sublimation and storage
150 columns is listed in Table 1.

151 The distribution of temperature in the pre-chilling, CO₂ anti-sublimation and
152 storage columns is simulated in Fig. 5. The temperature field of the three columns is
153 labeled in the radial position. Because the function of the pre-chilling section is to
154 reduce the temperature of feed gas and separate moisture, the lowest temperature is
155 around -20 °C. By contrast, the minimum temperature in the CO₂ anti-sublimation
156 tower can drop to -105 °C to anti-sublimate the CO₂ from the flue gas. For the storage
157 column, for conditions below -78 °C (the frost point of the pure CO₂) must be
158 maintained in order to avoid frosted CO₂ sublimation.

159 3. Theoretical analysis

160 3.1 Overall energy consumption of the process

161 The energy flow diagram of the cryogenic CO₂ capture process is shown in Fig. 6.
162 First, the flue gas from the impurity removal units is introduced into the pre-chilling
163 section. In the next stage, SC-1 provides cold energy (Q_{SC-1}) to the pre-chilling tower.

Under low temperature conditions, the moisture in the gas stream can be condensed into water and separated from the outlet of the condensate tube. Then, the treated gas is introduced into the CO₂ anti-sublimation column, which is chilled by SC-2. With the cold energy Q_{SC-2} , the CO₂ in the gas stream can anti-sublimate into solid form (namely, dry ice) on the surface of the cold head. Simultaneously, the residual gas (mainly as N₂) exhausts from the system without phase change. The fallen CO₂ droplets are gathered and preserved by the cold energy from SC-3 (Q_{SC-3}). Finally, the cold energy of the captured CO₂ is recovered by heat exchangers. The heat loss of transmission during the three stages is represented by q_1 , q_2 and q_3 , respectively. Meanwhile, the latent heat loss associated with condensate water, residual gas and captured CO₂ is represented by q_4 , q_5 and q_6 , respectively.

3.2 Characteristics of flue gas

The mass flow rate of the simulated flue gas (φ_M) can be calculated as follows:

$$\varphi_M = \varphi_V \cdot \rho_g \quad (1)$$

where φ_V is the mass and volume flow rates of the gas mixture. ρ_g is the density of gas mixture and can be calculated as follows:

$$\rho_g = \omega_{N_2} \cdot \rho_{N_2} + \omega_{CO_2} \cdot \rho_{CO_2} + \omega_{H_2O} \cdot \rho_{H_2O} \quad (2)$$

here, ω_{N_2} , ω_{CO_2} and ω_{H_2O} are the percentage of N₂, CO₂ and water vapor in the flue gas, respectively. ρ_{N_2} , ρ_{CO_2} and ρ_{H_2O} are the density of N₂, CO₂ and H₂O, respectively.

184 The average heat capacity of the gas mixture (C_g) is expressed as follows:

$$185 \quad C_g = \omega_{N_2} \cdot C_{N_2} + \omega_{CO_2} \cdot C_{CO_2} + \omega_{H_2O} \cdot C_{H_2O} \quad (3)$$

186 here, C_{N_2} , C_{CO_2} and C_{H_2O} are the heat capacity of N_2 , CO_2 and water vapor,
187 respectively.

188 3.3 Energy consumption of the pre-chilling column

189 The total energy consumption of the pre-chilling column (Q_I) can be calculated by
190 the heat removal of SC-1 (H_{SC-1}).

$$191 \quad Q_I = \frac{H_{SC-1}}{\eta_{SC-1}} \quad (4)$$

192 here η_{SC-1} is the coefficient of performance (COP) for SC-1.

193 The heat removal of SC-1 can be divided into two parts: sensible heat of gas stream
194 (Q_{SI}) and latent heat of evaporation for water vapor (Q_{LI}). Hereinto, the sensible heat
195 for SC-1 (Q_{SI}) can be calculated as follows:

$$196 \quad Q_{SI} = \varphi_M \cdot C_g \cdot \Delta T_I \quad (5)$$

197 here ΔT_I is the temperature variation from the inlet to the outlet of the pre-chilling
198 column.

199 The calculation method for the latent heat of the condensate water (Q_{LI}) is
200 presented in Equation (6):

$$201 \quad Q_{LI} = \varphi_{m_{H_2O}} \cdot H_{H_2O} \quad (6)$$

202 here, H_{H_2O} is the vaporization heat of H_2O . The mass flow rate of H_2O ($\varphi_{m_{H_2O}}$) can
203 be calculated as follows:

$$\varphi_{m_{H_2O}} = \frac{\omega_{H_2O} \cdot \varphi_V \cdot M_{H_2O}}{V_M} \quad (7)$$

where M_{H_2O} is the mole mass of H_2O . Thus, the total energy consumption of the pre-chilling column (Q_1) can be calculated by the sum of the sensible heat and latent heat.

$$Q_1 = \frac{Q_{S1} + Q_{L1}}{\eta_{SC-1}} \quad (8)$$

3.4 Energy consumption of the CO_2 anti-sublimation column

For the CO_2 capture column, the total energy consumption (Q_2) can be expressed as:

$$Q_2 = \frac{H_{SC-2}}{\eta_{SC-2}} \quad (9)$$

where H_{SC-2} is the heat removal of SC-2. η_{SC-2} is the COP of SC-2.

The heat removal of SC-2 can be divided into the sensible heat of gas stream (Q_{s2}) and latent heat of anti-sublimation for CO_2 (Q_{L2}). The sensible heat (Q_{s2}) can be calculated as follows:

$$Q_{s2} = \varphi_M \cdot C_g \cdot \Delta T_2 \quad (10)$$

here, ΔT_2 is the temperature difference between the inlet and outlet of the CO_2 anti-sublimation column.

In addition, the latent heat (Q_{L2}) can be calculated as follows:

$$Q_{L2} = \varphi_{mCO_2} \cdot H_{CO_2} \quad (11)$$

here, H_{CO_2} is the heat of sublimation for CO_2 . The mass flow rate of CO_2 (φ_{mCO_2})

223 can be calculated as follows:

$$224 \quad \varphi_{m\ CO_2} = \frac{\omega_{CO_2} \cdot \varphi_V \cdot M_{CO_2}}{V_M} \quad (12)$$

225 The total energy consumption of the CO₂ anti-sublimation column (Q_2) is the sum
226 of the sensible heat and latent heat:

$$227 \quad Q_2 = \frac{Q_{s2} + Q_{L2}}{\eta_{SC-2}} \quad (13)$$

228 *3.5 Energy consumption of the storage column*

229 Due to no phase change in the storage column, the heat removal of SC-3 (H_{SC-3}) is
230 mainly used to maintain the preserving condition to avoid the sublimation of captured
231 CO₂.

$$232 \quad H_{SC-3} = \varphi_{m\ CO_2} \cdot C_{CO_2} \cdot \Delta T_3 \quad (14)$$

233 Thus, the total energy consumption (Q_3) of the storage column can be calculated as
234 follows:

$$235 \quad Q_3 = \frac{\varphi_{m\ CO_2} \cdot C_{CO_2} \cdot \Delta T_3}{\eta_{SC-3}} \quad (15)$$

236 here η_{SC-3} is the COP of SC-3.

237 **4. Methodology**

238 *4.1 Simulation work*

239 The simulation work was undertaken by using a commercial process simulator
240 (Aspen Plus software package, Version 7.3), Peng-Robinson equations of state are

241 reliable for three phase systems over a wide range of temperatures and pressures.

242 The condition of the flue gas is simulated as a typical coal-fired power plant and
243 listed in Table 2. In order to improve the purity of CO₂ products and avoid corrosion
244 of installation the impurities (i.e. SO_x, NO_x and ash) are often removed by the
245 pre-treatment units, such as flue gas desulphurization (FGD), selective catalytic
246 reduction (SCR) and electrostatic precipitators (ESP) [24]. In order to simplify the
247 simulation process, these units are not considered in this work.

248 *4.2 Parameter study*

249 According to analysis in the previous section, it was found that there are three
250 parameters (i.e., temperature of Stirling coolers, flow rate of flue gas and CO₂
251 concentration) that influence the energy consumption of the cryogenic system. The
252 Stirling cooler 2 and 3 (SC-2 and 3) are used to capture and storage solid CO₂ in the
253 whole process, and therefore their temperature is set at the minimum (-120 °C). The
254 influence of temperature on the energy consumption of the system is considered SC-1
255 in this work. Meanwhile, the influence of the parameters on the energy consumption
256 of the system is investigated at different levels.

257 The detailed conditions for the parameters are listed in Table 3. The performance of
258 the SC is depicted in Table 4. In addition, the detailed characteristics of the feed gas
259 (from a typical coal-fired power plant) have been introduced in our previous work
260 [27].

261 **5. Results and discussion**

262 *5.1 Effect of cryogenic temperature on SC-1*

263 The relationship of energy consumption with the temperature on SC-1 is described
264 in Fig. 7. The detailed energy variation of the different sections: (a) pre-chilling stage,
265 (b) CO₂ anti-sublimation stage and (c) storage stage, according with the temperature
266 of SC-1 is depicted in Fig. 8.

267 The results indicate that with decreasing temperature for SC-1, the energy
268 consumption of pre-chilling (as shown in Fig. 8(a)) and the storage column (as shown
269 in Fig. 8(c)) both increased. However, the energy consumption of the CO₂
270 anti-sublimation column (as shown in Fig. 8(b)) obviously decreased. When the
271 temperature of SC-1 varied from -60 °C to -20 °C, the energy consumption of
272 pre-chilling stage decreased from 18.78 to 14.50 _{thermal} J/s, and the energy
273 consumption of the storage column varied from 12.73 to 7.20 _{thermal} J/s. In contrast,
274 the energy consumption of the CO₂ anti-sublimation stage increased from 25.90 to
275 32.03 _{thermal} J/s. This can be explained by the fact that sufficient pre-treatment in the
276 pre-chilling tower is beneficial in decreasing the temperature of the flue gas, and part
277 of the cold energy can be preserved in the CO₂ anti-sublimation stage. It can also be
278 found that due to the flow rate and CO₂ concentration is constant, the variation of
279 energy consumption for the system according to the temperature of SC-1 is modest.

280 *5.2 Effect of the flow rate of gas stream*

281 The energy consumption of the system under different flow rate of the gas stream is
282 summarized in Fig. 9, and the detail of the energy variation of the three stages
283 (pre-chilling, CO₂ anti-sublimation and storage) is shown in Fig. 10.

284 The results indicate that with an increase in the flow rate, the energy consumption
285 of the pre-chilling column (as shown in Fig. 10 (a)) increased rapidly. When the flow
286 rate was set at 1 L/min, the energy consumption of pre-chilling stage was 2.95 thermal
287 J/s. When the flow rate increased to 5 L/min, the energy consumption of the
288 pre-chilling stage increased to 15.58 thermal J/s. Meanwhile, the energy consumption of
289 the CO₂ anti-sublimation column (as shown in Fig. 10 (b)) also increased according to
290 the flow rate. When the flow rate increased from 1 to 5 L/min, the energy
291 consumption of the CO₂ anti-sublimation stage varied from 6.18 to 30.48 thermal J/s.
292 Finally, the energy consumption of the storage column (as shown in Fig. 10 (c)) also
293 increased from 2.28 to 11.40 thermal J/s. The higher flow rate implies increasing
294 demand on the treatment capacity. Therefore, more cold energy needs to be consumed
295 to maintain the capture efficiency.

296 *5.3 Effect of the CO₂ concentration*

297 The energy variation of the whole cryogenic capture process with different CO₂
298 concentration is illustrated in Fig. 11. Meanwhile, the detailed variation of the energy
299 consumption for the three stages according to the concentration of CO₂ in the gas
300 stream is shown in Fig. 12.

301 The results indicate that with increasing CO₂ concentration, the energy
302 consumption of the pre-chilling stage (as shown in Fig. 12 (a)) increased slowly.
303 When the CO₂ concentration was set at 5 %, the energy consumption of the
304 pre-chilling stage was 15.38_{thermal} J/s. When the CO₂ concentration increased to 40 %,
305 the energy consumption of the pre-chilling stage increased to 16.38_{thermal} J/s. This is
306 because the function of the pre-chilling stage is to separate the moisture from the flue
307 gas. Therefore, the energy consumption of this part varied little with respect to the
308 CO₂ concentration. By contrast, the energy consumption of the CO₂ anti-sublimation
309 stage (as shown in Fig. 12 (b)) increased rapidly in association with the concentration
310 of CO₂. When the concentration increased from 5 to 40 %, the energy consumption of
311 the CO₂ anti-sublimation stage varied from 18.50 to 67.53_{thermal} J/s. Meanwhile, the
312 energy consumption of the storage stage (as shown in Fig 12. (c)) also increased from
313 4.20 to 40.15_{thermal} J/s. As the CO₂ concentration increased, the requirement for phase
314 change also increased. Thus, the energy consumption of the CO₂ anti-sublimation and
315 storage stages increased rapidly.

316 5.4 Economic evaluation

317 The techno-economic evaluation (capital and operational cost) of the system is
318 shown in Table 5. The total cost of the process consists of two parts: the capital cost
319 and operational cost. The capital cost depends on the modules, including FPSCs,
320 control panel and equipment frame. The capture process is carried out under the
321 cryogenic conditions, therefore the operational cost of deposition is also an important

322 component and determined by the electric power consumption.

323 *5.5 Comparison with existing processes*

324 The boundary conditions of the different processes are summarized in Table 6,
325 which indicates that the absorption, membrane and cryogenic factors (by Tuinier et al)
326 are based on a 600 MWe coal fired power plant. By contrast, the cryogenic process
327 developed by Clodic et al is tested in the pilot plant, and the adsorption process and
328 present work are carried out on a laboratory scale.

329 The performance comparison among the existing CO₂ capture technologies is
330 presented in Fig. 13. The comparison indicates that the cryogenic process exploited by
331 Tuinier et al. is most costly (4.5 MJ_{thermal}/kg CO₂) compared with other technologies.
332 This is because the cryogenic process is based on the consumption of electricity
333 (approximately 1.8 MJ_{electrical}/kg CO₂), and which is equal to 4.5 MJ_{thermal}/kg CO₂
334 (the efficiency of conversion is around 0.4). By contrast, it can be seen that the
335 present cryogenic process shows a promising CO₂ capture performance (1.37 MJ
336 thermal energy per capture of kg CO₂), and which is close to the performance of the
337 most mature absorption process (about 1.6 MJ_{thermal}/kg CO₂). The low energy
338 consumption of the present work can be attributed to the high efficiency of the SCs,
339 which is higher than the conventional refrigerators. However, it should be pointed out
340 that the performance of the exploited system has just been simulated, and this value
341 should be further tested in a real power plant.

342 5.6 Scale-up the cryogenic CO₂ capture process

343 Based on the experience gained from energy analysis and previous lab-scale studies,
344 the feasibility of scaling up the developed cryogenic CO₂ capture process can be
345 forecasted. To process the enormous flow of flue gas (600 kg/s in a typical coal-fired
346 power plant), the diameter of separation tower must be correspondingly increased [30].
347 Meanwhile, the number of Stirling coolers in different stages (pre-freezing, main
348 freezing and storage) should also be increased. The schematic diagram of the scale-up
349 installation is conceptually designed in Fig. 14. From the current situation, heat loss
350 would be an unavoidable challenge due to the huge temperature difference between
351 the internal and external of the installation and it can be overcome by an effective heat
352 recovery process.

353 6. Conclusion

354 In this work, the overall energy flow of the Stirling cooler based cryogenic CO₂
355 capture process was simulated. The theoretical analysis of the energy consumption for
356 each unit in the capture system was also undertaken. When the temperature of SC-1
357 varied from -20 to -60 °C, the total energy consumption of the cryogenic CO₂ capture
358 system increased from 53.73 to 57.41_{thermal} J/s. When the flow rate of flue gas
359 increased from 1 to 5 L/min, the total energy consumption of the cryogenic CO₂
360 capture system increased from 11.41 to 57.46_{thermal} J/s. When the CO₂ concentration
361 in the flue gas increased from 5 % to 40 %, the total energy consumption of the

362 cryogenic CO₂ capture system increased from 30.08 to 124.06 thermal J/s. The energy
363 analysis provides a reliable way to improve capture efficiency and reduce energy
364 penalty.

365 In future work, the dynamic operating process of system will be simulated, and
366 simultaneously a pilot plant test based on the developed system will also be carried
367 out. It should be noted that the sensible and latent heat of the separated components
368 (such as condensate water and residual gas) is substantial. Therefore, the heat
369 recovery units will be integrated into the system in the next step.

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375

Nomenclature

ΔT	Temperature, °C
C	Heat capacity, kJ/kg·°C
Q	Energy, J
q	Heat loss, J
H_{H_2O}	Heat of evaporation for water, kJ/mol

H_{CO_2}	Heat of sublimation for CO ₂ , kJ/mol	376
M	Mass, kg	377
V	Volume, L	378
<i>Greek letters</i>		
ω_{H_2O}	Percentage of H ₂ O in flue gas, %	379
ω_{CO_2}	Percentage of CO ₂ in flue gas, %	380
φ	Flow rate, L/min	381
ρ	Density, kg/m ³	382
η	Coefficient of performance	
<i>Abbreviations</i>		
<i>CCS</i>	CO ₂ capture and storage	383
<i>GHG</i>	Greenhouse gas	384
<i>SC</i>	Stirling cooler	385
<i>EC</i>	Energy consumption	386
<i>TSA</i>	Temperature swing adsorption	387
<i>ESA</i>	Electric swing adsorption	388
<i>PSA</i>	Pressure swing adsorption	389
<i>VSA</i>	Vacuum swing adsorption	
<i>COP</i>	Coefficient of performance	

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Figure captions

Fig. 1. Schematic of the developed anti-sublimation capture process.

Fig. 2. The relationship between freezing point ($^{\circ}\text{C}$) and CO_2 concentration (vol.%) (a); in the case of typical flue gas (b).

Fig. 3. The detailed CO_2 capture and storage stage in the system.

Fig. 4. The geometrical structure of the columns in the system.

Fig. 5. Temperature distributions in the pre-cooling tower, CO_2 anti-sublimation tower and storage column.

Fig. 6. The overall energy flow of the cryogenic CO_2 capture process.

Fig. 7. Influence of temperature of SC-1 on the energy consumption of the pre-chilling, CO_2 anti-sublimation and storage stages.

Fig. 8. Effect of the temperature of SC-1 on the energy consumption of the cryogenic CO_2 capture system. (a) Pre-chilling stage; (b) CO_2 anti-sublimation stage; (c) storage stage. The flow rate of the gas stream is set at 5 L/min. The temperature of SC-2 and 3 are both set at -120°C . The COP of SC-1, 2 and 3 are 1.1, 0.7 and 0.7, respectively. The concentration of CO_2 is set at 13% (in a typical the coal-fired power plant). The conversion efficiency from electric power to thermal energy is defined as 40%.

Fig. 9. Influence of flow rate on the energy consumption of the pre-chilling, CO_2 anti-sublimation and storage stages.

Fig. 10. Effect of flow rate on the energy consumption of the cryogenic CO₂ capture system. (a) Pre-chilling stage; (b) CO₂ anti-sublimation stage; (c) storage stage. The temperature of SC-1, 2 and 3 are set at -30°C, -120°C and -120°C, respectively. The COP of SC-1, 2 and 3 are 1.1, 0.7 and 0.7, respectively. The concentration of CO₂ is set at 13%. The conversion efficiency from electric power to thermal energy is defined as 40%.

Fig. 11. Influence of CO₂ concentration on the energy consumption of the pre-chilling, CO₂ anti-sublimation and storage stages.

Fig. 12. Effect of CO₂ concentration on the energy consumption of the cryogenic CO₂ capture system. (a) Pre-chilling stage; (b) CO₂ anti-sublimation stage; (c) storage stage. The temperature of SC-1, 2 and 3 are set at -30°C, -120°C and -120°C, respectively. The COP of SC-1, 2 and 3 are 1.1, 0.7 and 0.7, respectively. The flow rate of flue gas is set at 5 L/min. The conversion efficiency from electric power to thermal energy is defined as 40%.

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Table 1

Geometric dimensions of the three columns in the system.

Column	Geometric dimension (m)		
	OD*	ID*	L*
Pre-chilling	0.21	0.20	0.25
CO ₂ anti-sublimation	0.21	0.20	0.25
Storage	0.12	0.10	0.12

* OD, ID and L represent outside diameter, inside diameter and length of the column.

Table 2

Properties of the simulated flue gas (in a typical coal-fired power plant).

Properties	Value
Temperature	50 °C
Pressure	1.01×10^5 Pa
Composition of the feed gas	vol. %
CO ₂	13
N ₂	82
H ₂ O	5
SO _x	-
NO _x	-
Fly ash	-
CO ₂ recovery ratio	95 %
CO ₂ purity	99 %

Table 3

Parameters that influence the energy consumption of the system.

Parameters	Levels				
	#1	#2	#3	#4	#5
Temperature of SC-1 (°C)	-20	-30	-40	-50	-60
Flow rate of flue gas (L/min)	1	2	3	4	5
CO ₂ concentration (vol.%)	5	10	20	30	40

Table 4

Performance of the Stirling cooler (SC).

Performance	Stirling cooler
Power input	150 w
Cooling capacity	120 w (cold side temperature: -25 °C) 50 w (cold side temperature: -80 °C) 20 w (cold side temperature: -120 °C)
Frequency	≤10μm (peak to peak)
Working gas	Helium

Table 5

Technical and economic evaluation of the cryogenic CO₂ capture system.

Section	Equipment	Capital cost (\$)	Operational cost (\$/h)
Pre-chilling tower	FPSC-1	3750	0.03
CO ₂ anti-sublimation tower	FPSC-2	3750	0.056
	Scraper	300	0.0045
Storage tower	FPSC-3	3750	0.056
Others	Control panel	12500	0.019
	Frame	37500	-
Total direct investment	6.16×10 ⁴		
Total operational investment	2.79×10 ⁴		
Total fixed investment	8.95×10 ⁴		
Capture performance			
CO ₂ recovery	90%		
CO ₂ purity	99.9%		
Energy consumption	1.37 MJ _{thermal} /kg CO ₂		
Capture cost	35 \$/kg CO ₂		

* The reference flow rate of gas mixture is 5 L/min.

Table 6

The boundary condition of the compared CO₂ capture technologies.

Properties	Absorption		Adsorption	Membrane		Cryogenic	Cryogenic		The present work
	[26]		[4]	[27]		[24]	[17]		
Scale	600	MWe	Laboratory	600	MWe	Pilot plant	600	MWe	Laboratory
	coal	fired		coal	fired		coal	fired	
	power			power			power		
	plant			plant			plant		
Composition (vol. %)									
N ₂	86		90	87		81.4	86.5		82
O ₂	-		-	-		7	-		-
CO ₂	14		10	13		11.6	13.5		13
H ₂ O	-		-	-		-	-		5
Flow rate (kg/s)	666		0.67	602		3.45	635		0.17
Temperature (°C)	30		20	30		50	150		25
Pressure (bar)	1		1	1		1	1		1