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2	Study on the COP of Free Piston Stirling Cooler
3	(FPSC) in the anti-sublimation CO ₂ capture process
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22	Abstract

23	Free piston Stirling cooler (FPSC) is a promising alternative for the conventional
24	coolers and has been applied to various fields. In the previous research, a novel
25	cryogenic CO ₂ capture system based on FPSCs has been exploited. In order to
26	enhance the cryogenic CO ₂ capture efficiency, the investigation on the coefficient of
27	performance (COP) of the FPSC is carried out in this work. In detail, the influence of
28	different materials (aluminium and copper), size of cold head (length and diameter),
29	as well as ambient conditions (humidity and temperature) on the COP of the
30	cryogenic system were tested. The experiment results indicate that the material of cold
31	head should be selected at copper to increase the COP of CO ₂ capture system. The
32	length and diameter of cold head should be short and thick. In addition, the low
33	ambient temperature is benefit for the high COP. For the optimal conditions (the
34	material was copper, length and diameter were 180 and 40 mm, respectively), the
35	temperature of the cold head reached -140 °C, and the COP of the FPSC and the
36	cryogenic CO ₂ system was 0.82 and 0.70, respectively.
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39	Keywords: free piston Stirling cooler, COP, performance, CO ₂ capture
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	Nomenclature
	<i>L</i> Length of cold head, mm

Р	Pressure, Pa
Q	Cooling capacity, J
Ta	Ambient temperature, °C
T_c	Temperature of the cold head, $^{\circ}C$
V	Volume, m ³
W	Input power, J
h_a	Ambient humidity, %
m	Mass, kg
Abbreviations	
CCS	CO ₂ capture and storage
СОР	Coefficient of performance
FESC	Free piston Stirling cooler
LN2	Liquid nitrogen
LNG	Liquefied natural gas
PCC	Post-combustion CO ₂ capture

1. Introduction

49	According to the prediction of Intergovernmental Panel on Climate Change (IPCC),
50	by the year 2100, the atmosphere may contain up to 570 ppmv of CO ₂ , causing a rise
51	of mean global temperature of around 1.9 $$ $^{\circ}$ C and an increase in mean sea level of 3.8
52	m [1]. As an effective strategy to mitigating CO ₂ emissions, much attention has been
53	paid on post combustion CO ₂ capture (PCC) techniques. Nowadays, the most mature
54	post-combustion CO ₂ capture technology is based on CO ₂ absorption by aqueous
55	amine solutions and has been commercially utilized on the large CO ₂ emission
56	sources (i.e. coal-fired power plants, cement and steel plant) [2,3]. In the absorption
57	processes, CO ₂ reacts reversibly in an absorber with amines which are regenerated by
58	heating the solution in a stripper column [4]. Nevertheless, the biggest bottleneck for
59	the chemical absorption processes is that the regeneration of solvents is energy
60	penalty [5,6]. Meanwhile, the degradation of the aqueous amine solvents also leads to
61	an increasing cost [7]. In light of this situation, the alternative methods (such as
62	adsorption, membrane, cryogenic, microalgae and chemical looping) have also
63	attracted the attention of the scientists [8]. Compared to the other CO ₂ capture
64	technologies, cryogenic CO ₂ capture approach can achieve a high CO ₂ purity (above
65	99%) and which can minimize the compression and transport costs significantly [9].
66	Furthermore, the CO ₂ capture process can be achieved by the phase change, and there
67	is no utilization of chemical solvents. Consequently, a number of researches have
68	been driven toward this technique. [10-13].

As the critical part of the cryogenic CO₂ capture methods, several low temperature
sources have been utilized and investigated [14]. In 2002, Clodic et al. developed a

novel cryogenic CO₂ capture process by using the cold duty from liquid nitrogen 71 (LN2). The CO_2 in the flue gas can be recovered in the liquid form and which is 72 73 beneficial to the further compression and transport. However, the main disadvantage of the process is that the deposited CO_2 on the cold head would adversely affect the 74 75 heat transfer. Moreover, the moisture in the flue gas has to be separated beforehand to avoid the plugging by ice during the operation [15]. In 2011, Tuinier et al. exploited a 76 cryogenic packed bed taking advantage of the cold energy from liquefied natural gas 77 (LNG) regasification terminal. The moisture and CO₂ can be separated and collected 78 79 at the different locations in the process. However, the coefficient of performance (COP) of the system is typically 0.5, and thus about 3.6 MJ electric energy is required 80 and resulting in even higher thermal energy to capture per kg CO_2 [16]. 81

82 Free piston Stirling cooler (FPSC) is a new type cryogenic cooler attracting interest as the low temperature source [17]. Compared to the conventional coolers, FPSC can 83 use helium as regenerator, and avoid the increasing environmental issues (such as 84 85 ozone depletion) caused by CFCs, HCFCs and HFCs [18]. Meanwhile, high energy 86 efficiency and reliability are also the advantage of FPSC. For these reasons, FPSC has been utilized in the cryogenic CO₂ capture process in the previous work [19]. 87 However, it needs to point out that the original cooling region of FPSC is limit duo to 88 the size of itself. During the application process, it often needs to extend the cold head 89 for further refrigeration. Therefore, the investigation on the COP and cryogenic 90 91 temperature of FPSC is significant for improving the CO₂ capture efficiency.

92 The objective of this study is to investigate the characteristic of FPSC and

theoretically analyze the COP of the FPSC. The research also focuses on the influence 93 of the key parameters on the COP of FPSC, such as the material (aluminium and 94 95 copper) and size (length and diameter) of the cold head, as well as the ambient conditions (temperature and humidity). The structure of this paper is as follows. 96 Section 2 describes the cryogenic CO₂ capture process and deduces the COP of FPSC. 97 Section 3 shows the detailed experimental conditions. Section 4 investigates the key 98 parameters that influence the COP and cryogenic temperature of FPSC. Section 5 99 100 discusses the possibility of increasing the efficiency by the heat exchange between the 101 separated and incoming streams and the application in the large scale. Section 6 summarizes the main conclusions of this research. 102

103 **2. Base case description**

104 2.1. Cryogenic CO₂ capture process

The structure of designed cryogenic CO₂ capture system based on free piston 105 106 Stirling coolers (FPSC) has been introduced in our previous work [20, 21]. The whole capture process can be briefly described as follows: three Stirling coolers used in the 107 system are named as FPSC-1, FPSC-2 and FPSC-3, respectively. First, the flue gas is 108 introduced into the pre-freezing tower. Under the low temperature, the moisture in the 109 110 gas stream can condense and be separated. Then the dry flue gas flows into the main freezing tower. Under the cryogenic condition (approximately -110 $^{\circ}$ C), the CO₂ in 111 the gas stream frosts on the surface of the cold head of FPSC-2, and simultaneously 112 the other gas (such as N₂ and O₂) is exhausted without phase change. Finally, the 113

captured CO₂ is separated by the motor driven scraper and gathered in the storage 114 column to further compress for transport. The key parameters (such as flow rate, 115 116 temperature and operating time) that affect the capture performance are controlled and recorded by the control panel. 117 In addition, the detailed connection of FPSC-2, cold head and main-freezing tower 118 is shown in Fig. 1. The cold head is chilled by FPSC-2 to generate the required low 119 temperature condition (around -110 $^{\circ}$ C) in the main freezing tower. Therefore, during 120 the capture process, the CO₂ in the gas stream can be separated and frosted on the 121 122 surface of the cold head. It should be noted that in order to enhance the heat transfer efficiency, the vacuum interlayer is maintained between the internal of the tower and 123 the ambient surroundings. 124

125 2.2. Free piston Stirling cooler (FPSC)

126 2.2.1 Theoretical analysis

A schematic diagram of the FPSC is shown in Fig. 2. The FPSC may be defined as 127 a pressure vessel which operates by shuttling approximately 1 gram of helium back 128 and forth by the combined movements of two parts, namely the piston and the 129 displacer. While the piston that compresses the gas is driven by a linear motor, the 130 displacer is moved by the pressure difference. Heat exchanger and regenerator are 131 assembled to separate the compression and expansion spaces for the creation of a 132 thermal gradient which allows the FPSC to extract heat from the cold head and reject 133 heat to the region around hot head. This process is repeated many times per second 134

and can ultimately produce temperature differences between the cold and hot head.
During the whole process, heat can be moved from a remote location to the FPSC and
then rejected to the environment. The detailed working principle of FPSC has been
introduced by Berchowitz in 1992 [22].

139 2.2.2 Coefficient of performance (COP)

As an important quality to evaluate the performance of FPSC, the investigation on the COP is significant. The COP is defined as the ratio of the cooling capacity (Q_e) and the input power (W) of the system, and can be expressed as follows:

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$$COP = \frac{Q_e}{W}$$
(1)

144 where the input power (*W*) can be obtained from:

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$$W = \frac{\omega_0}{2\pi} \int_0^{2\pi} P_c dV$$
 (2)

here P_c and V are the pressure and volume of cylinder, and can be given by the following equations [22]:

148
$$P_c = \langle P \rangle + |P_c| \sin\phi$$
 (3)

$$149 \quad V = V_0 - A_p X_p \sin\phi \tag{4}$$

Taking equation (3) and (4) into equation (2), the input power (*W*) can be expressedas:

152
$$W = -\frac{\omega_0}{2} \alpha_T X_p X_d \sin\phi$$
(5)

where, X_d and X_p are the amplitude of the displacer and piston. α_T is the thermal coupling between the displacer motion and piston force, and it can be calculated as follows:

$$\alpha_T = A_p \frac{\partial P_c}{\partial x_d} \tag{6}$$

157 In addition, the cooling capacity (Q_e) is described as follows:

158
$$Q_e = -\frac{\omega_0}{2} \left\{ \alpha_p X_p \left(\frac{A}{A_R} - 1 \right) \sin\phi - K_{ext_d} X_p \frac{A}{A_R} \frac{m_p}{m_c} \sin\phi + C_{ext} \omega_0 X_d \right\} X_d$$
(7)

where *A* is the crosscut area of the cylinder. C_{ext} is incidental damping. It should be noted that heat transfer losses (Q_L) is inevitable. Among the heat losses, conduction (Q_{cond}) and regenerator (*H*) losses are the most significant.

$$Q_L = Q_{cond} + H \tag{8}$$

163 In conclusion, the COP of the FPSC can be expressed as the following equation:

164
$$COP = \frac{\alpha_P}{\alpha_T} \left(\frac{A}{A_R} - 1 \right) - \frac{A}{A_R} \frac{K_{ext}}{\alpha_T} \frac{m_P}{m_c} + \frac{C_{ext}}{\alpha_T} \frac{\omega_0}{\sin\phi} \frac{X_d}{X_P}$$
(9)

Simultaneously, the investigation on the coefficient of performance for the whole
system (COPS) is implemented to evaluate the performance of the system. The COPS
of the system can be calculated as follows:

168
$$COPS = \frac{Q_{C1} + Q_{C2} + Q_{C3}}{W_s}$$
 (10)

here Q_{C_1} , Q_{C_2} and Q_{C_3} are the cooling capacity of FPSC-1, 2 and 3, respectively. W_s is the input energy of the system.

171 **3. Experimental**

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The investigation of the COP are based on the experiments four parameters, namely material, length and diameter of the cold head, as well as the ambient humidity and temperature. Initially, the cold head of FPSC is tested with different materials (aluminium and copper). Then, the size (length and diameter) of the cold head is

investigated. The structure of the cold head with different materials (aluminium and 176 copper) is shown in Fig. 3. The length (L) of cold head is set at (180 mm and 270 177 178 mm), respectively. The diameter (D) of the cold head is investigated under 30 mm and 40 mm. It needs to point out that the selection of length and diameter is just 179 depending on the configuration of the system, and there is no special representation. 180 Additionally, during the operating process, the FPSC needs to intake the air from 181 ambient and exhaust the hot air simultaneously. Therefore, the influence of ambient 182 humidity (h_a) and temperature (T_a) on the COP of FPSC is also investigated. 183

184 **4. Results**

- 185 *4.1 Effect of the cold head on the COP*
- 186 *4.1.1 Material*

The relationship between the COP of FPSC and the material of the cold head is 187 investigated according with the chilling process (as shown in Fig. 4). With the 188 temperature reduction of the cold head, the cooling capacity of FPSC decreased, and 189 which led to the fall of COP. However, the aluminium cold head has a relative higher 190 COP decreasing rate compared to the copper one. It results that the FPSC with an 191 aluminium cold head has a lower COP than the copper one in the whole temperature 192 drop process. When the root temperature of the cold head dropped to -140 °C, the COP 193 194 of FPSC with the aluminium and copper cold head are 0.61 and 0.82 respectively. It can be deduced that the COP of the copper cold head is higher than the aluminium, 195 and which means a large cooling capacity can be obtained. 196

The effect of the material of cold head on the cryogenic temperature is depicted in 197 Fig. 5. From the results, it can be concluded that the cold head made by copper has a 198 199 lower temperature than aluminium. After 240 min, the root temperatures are -111.2 °C for the aluminium cold head and -111.3 $\,^{\circ}\!\!\!{\rm C}$ for the copper. Meanwhile, the front 200 temperatures of the aluminium and copper cold head are -78.09 °C and -97.82 °C. In 201 addition, the temperature decreasing rate for the copper cold head is higher than the 202 aluminium. For copper, the temperature drop from the root of the cold head to the 203 front is 13.38 $\,$ °C. By contrast, although the mass of the cold head by aluminium is 204 205 light, the thermal loss is great (with a temperature drop of 33.31 $^{\circ}$ C).

206 *4.1.2 Length*

207 The results in Fig. 6 show that the influence of the length of cold head on the COP of FPSC. It can be observed that with the increase of the length, the COP of FPSC 208 decreased obviously. However, for the aluminium cold head, the decreasing rate of 209 210 COP is higher than the copper one. When the length is extended from 180 mm to 270 mm, the COP dropped from 0.82 down to 0.59 (the root temperature of the cold head 211 was -140 $^{\circ}$ C). It can be explained by the fact that along with the increasing of the 212 213 length of the cold head, the heat loss also increase and the cooling capacity of the cold head reduced accordingly. Therefore, the COP of FPSC also dropped. 214

As the results in Fig. 7, the cryogenic temperature of the cold head decreases with the extension of the length. When the length of the cold head is set at 180 mm, the temperature drop from the root to the front is 9.92 °C. However, with the length extending to 270 mm, the temperature drop rose up to 15.73 °C. Furthermore, after
240 min, the front temperature of the cold head for 180 mm and 270 mm are
-84.08 °C and -73.17 °C. That is because that along with the extension of the length,
the temperature loss of the cold head also increases.

222 *4.1.3 Diameter*

As presented in Fig. 8, the COP variation process with different diameters was investigated. The COP of FPSC reduced according with the temperature drop of the cold head. With the expanding of the diameter of the cold head (from 30 mm to 40 mm), the COP of FPSC increased obviously. Meanwhile, the decreasing rate of the COP was decelerated. When the root temperature of the cold head decreased to -140 \degree , the COP was 0.52 (diameter of 30mm) and 0.59 (diameter of 40 mm), respectively.

In addition, the relationship between the diameter and cryogenic temperature of the 230 cold head was depicted in Fig. 9. From the results, it can be concluded that a large 231 diameter is beneficial to reduce temperature loss. When the diameter of the cold head 232 is set at 30 mm, the lowest temperature for the root and front of the cold head is 233 234 -85 °C and -64.72 °C. The temperature drop is 20.28 °C from the root to the front side. By contrast, when the diameter is set at 40 mm, the lowest temperature for the root 235 and front sides is -88.9 $^{\circ}$ C and -73.17 $^{\circ}$ C respectively. The temperature drop can be 236 237 reduced to 15.73 °C. For the larger diameter (40 mm), both of the root and front temperature are lower than the smaller one (30 mm). It indicated that expanding the 238

diameter of the cold head is beneficial to improve heat transfer efficiency and theCOP of FPSC.

241 4.2 Effect of the ambient conditions on the COP

The effect of ambient temperature on the COP of FPSC is summarized in Fig. 10. 242 243 From the result, it can be observed that the COP of FPSC increased with the decreasing of ambient temperature. When the ambient temperature varied from 8 $\,$ $\,$ $\,$ $\,$ $\,$ $\,$ 244 to 28 °C, the COP of FPSC reduced from 0.7 to 0.6. That is for the reason that during 245 the refrigeration process of FPSC, it absorbs cool air from surrounding and 246 simultaneously exhausts warm air. A low ambient temperature is favor of increasing 247 temperature difference from the cold head of FPSC to the warm side. Thus, in order to 248 249 improve the performance of FPSC, the ambient temperature should be dropped as low as possible. The influence of ambient humidity on the COP of FPSC is shown in Fig. 250 11. From the results in the figure, it shows that with the ambient humidity increasing 251 from 20 % to 75 %, the COP of FPSC varied in the range of 0.62 to 0.68. It indicated 252 that the influence of ambient humidity on the COP of FPSC is not significant. 253

4.3 COP of the cryogenic CO₂ capture system

The COP variation of the FPSCs (FPSC-1, 2 and 3) and the whole system is shown in Fig. 12. From the results, it can be found that the COPs of three FPSCs were different. That is for the reason that due to the different functions in the cryogenic CO_2 capture system, the length of the cold head of the FPSCs is also different. When the temperature of the cold head for FPSC-2 was cooled to -140 °C, the COPs of

260	FPSC-1, 2 and 3 were 0.82, 0.78 and 0.79, respectively. Meanwhile, due to the other
261	energy consumption units (such as vacuum pump, control panel and scraper), the COP
262	of the whole system was 0.70.

263 **5. Discussion**

The COP of the Stirling cooler and the developed cryogenic CO_2 capture system has been tested under various conditions (including material, length and diameter of the cold head, as well as the ambient temperature and humidity) to achieve the optimal performance of the system. Simultaneously, the temperature of the cold head was also investigated to evaluate the cryogenic performance of the Stirling cooler. It can be concluded that the COP of the Stirling cooler is higher (about 0.82 at -140 °C) than the common refrigerators (around 0.5 at -140 °C).

Although the cryogenic CO₂ capture process has several advantages, it still suffers some limitations need to be overcome. It should be noticed that the whole capture process is based on the low temperature condition, and thus the latent heat associated with the separated components (i.e. the condensate water, exhaust gas and especially captured CO₂) would be substantial. For the future work, the cold energy of these components should be recovered by the heat exchangers, and then the total energy consumption of the system could be further reduced.

Owing to the flow rate of flue gas in the real power plants is higher than the laboratory scale, the scaling up of the capture capacity of the system is very significant. It can be achieved by utilizing the high power Stirling coolers. Meanwhile,

more Stirling coolers can be integrated into the system to improve the chilling ability of each parts (such as pre-freezing, main freezing and storage towers). However, it should be pointed out that with the increase of the amount of the Stirling coolers, the phenomenon of oscillation of the system may become increasing serious. Therefore, the influence of the oscillation phenomenon on the COP of Stirling cooler should be studied in the future research.

287 **6.** Conclusion

In the present work, the investigation on the COP of the FPSC was carried out. In order to improve the COP of FPSC in the cryogenic CO_2 capture process, the key parameters (material, length and diameter of the cold head, as well as the ambient temperature and humidity) were also investigated. Based on the experimental results, the conclusions are summarized as follows:

1) From the experiment of the materials, it can be concluded that when the cold
head was made by copper, the COP of FPSC was 0.82 with -140 °C of the cold head.
By contrast, the COP of FPSC for the aluminium cold head was 0.61.

2) For the different lengths of the cold head (270 mm and 180 mm), the COP of
FPSC was 0.59 and 0.82, respectively. Furthermore, when the diameter of the cold
head was set at 30 mm, the COP of FPSC was 0.52. By contrast, with the diameter
thickening to 40 mm, the COP of FPSC increased to 0.59.

300 3) In addition, the ambient temperature also presented a significant influence on the
301 COP of FPSC. When the ambient temperature varied from 8 °C to 28 °C, the COP of

FPSC reduced from 0.7 to 0.6. Nevertheless, the effect of ambient humidity on the
COP is not significant. With the ambient humidity increasing from 20 % to 75 %, the
COP of FPSC varied in the range of 0.62 to 0.68.
4) Although the FPSCs in the system had high COPs, the COP of the system was

around 0.70 due to other energy consumption units (such as vacuum pump and controlpanel).

308 Acknowledgement

We thank Mr. Yamano and Mr. Yamasaki of Tanabe Engineering Corporation fortheir technological assistance.

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Figure captions:

- 370 Fig. 1. The detailed connection of FPSC-2, cold head and main-freezing tower. (The gray area
- 371 represents the vacuum condition of the interlayer in the system.)
- **Fig. 2.** The detailed configuration of FPSC.
- 373 Fig. 3. The cold head of FPSC with different materials (aluminium and copper).
- Fig. 4. COP variation of FPSC with the root temperature of the cold head under different material
- 375 (L=180 mm; D=40 mm).
- Fig. 5. The effect of the material of cold head on the cryogenic temperature (L=180 mm; D=40 mm).
- **Fig. 6.** COP variation of FPSC with the root temperature of cold head under different length (*L*)
- 379 (material of cold head is copper; D=40 mm).
- **Fig. 7.** The relationship of length (*L*) and cryogenic temperature (T_c) of the cold head (material of
- 381 cold head is copper; D=40 mm).
- **Fig. 8.** COP variation of FPSC with the root temperature of cold head under different diameter (*D*)
- 383 (material of cold head is copper; L=270 mm).

Fig. 9. The relationship of diameter (*D*) and cryogenic temperature (T_c) of the cold head (material of cold head is copper; *L*=270 mm).

- **Fig. 10.** COP variation of FPSC with the different ambient temperature (T_a) (material of cold head is copper; L=180 mm; D=40 mm).
- **Fig. 11.** COP variation of FPSC with different ambient humidity (h_a) (material of cold head is
- 389 copper; L=180 mm; D=40 mm).
- **Fig. 12.** COP variation of the FPSCs and system with the root temperature of FPSC-2's cold head
- 391 (material of cold head is copper; L=180 mm; D=40 mm). COP-1, COP-2, COP-3 and COPS
- 392 represent the coefficient of performance for FPSC-1, FPSC-2, FPSC-3 and the whole system,
- 393 respectively.

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421 Fig. 1. The detailed connection of FPSC-2, cold head and main-freezing tower. (The gray area

422	represents the	vacuum condition	of the	interlayer in	the system.)
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Fig. 2. The detailed configuration of FPSC.







Fig. 4. COP variation of FPSC with the root temperature of the cold head under different material

^{450 (}L=180 mm; D=40 mm).





463 Fig. 5. The effect of the material of cold head on the cryogenic temperature (L=180 mm; D=40

464 mm).





Fig. 6. COP variation of FPSC with the root temperature of cold head under different length (L)





494	Fig. 7. The relationship of length (L) and cryogenic temperature (T_c) of the cold head (material of
495	cold head is copper; D=40 mm).
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Fig. 8. COP variation of FPSC with the root temperature of cold head under different diameter (D)

510 (material of cold head is copper; L=270 mm).





523	of cold head is copper; L=270 mm).
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Fig. 10. COP variation of FPSC with the different ambient temperature (T_a) (material of cold head

- 538 is copper; L=180 mm; D=40 mm).



Fig. 11. COP variation of FPSC with different ambient humidity (h_a) (material of cold head is

553 copper; L=180 mm; D=40 mm).





Fig. 12. COP variation of the FPSCs and system with the root temperature of FPSC-2's cold head
(material of cold head is copper; L=180 mm; D=40 mm). COP-1, COP-2, COP-3 and COPS
represent the coefficient of performance for FPSC-1, FPSC-2, FPSC-3 and the whole system,
respectively.